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► To cite this version:

Dominique Cluzel, Philippe Boulvais, Marion Iseppi, Didier Lahondère, Stephane Lesimple, et al.. Slab-derived origin of tremolite-antigorite veins in a supra-subduction ophiolite; the Peridotite Nappe (New Caledonia) as a case study.. *International Journal of Earth Sciences*, 2020, 109 (1), pp.171-196. 10.1007/s00531-019-01796-6 . insu-02369790

HAL Id: insu-02369790

<https://insu.hal.science/insu-02369790>

Submitted on 28 Nov 2019

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1 **Slab-derived origin of tremolite-antigorite veins in a supra-subduction ophiolite; the Peridotite**
2 **Nappe (New Caledonia) as a case study.**

3

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17 **Abstract**

18 Hydration of mantle peridotites provides information on the exhumation history and on the fluid
19 regime accompanying exhumation of these rocks (reservoirs involved, fluid/rock ratios, temperature
20 of interaction). Highly depleted harzburgites and dunites of the Peridotite Nappe of New Caledonia
21 are crosscut by fractures, which have been pervasively serpentinized, producing lizardite, brucite,
22 magnetite and minor chrysotile in a near-static environment, probably by sea water circulating in
23 cooling joints. This event is generally referred to as “primary serpentinization”. In a next step, already
24 serpentinized joints were re-opened to produce tension and shear cracks sealed by higher-
25 temperature synkinematic fibrous minerals. Locally, tremolite-bearing veins and pockets, which do
26 not display evidence for void infill, were generated by metasomatic replacement of the wall rock
27 peridotite. Most veins only contain fibrous antigorite but some display tremolite-antigorite
28 intergrowths or even pure tremolite. The latter rocks yield high contents of Ca and incompatible
29 elements, which contrast with the overall depletion of the peridotite host rock and suggest
30 contribution of an external source. Whole-rock geochemical and isotopic features ($^{87}\text{Sr}/^{86}\text{Sr}$, $\delta^{18}\text{O}$ and
31 δH) suggest that antigorite veins, which bear highly radiogenic Sr isotope signatures, were strongly
32 influenced by fluids emitted by the subducted slab and associated sediments. In contrast, the
33 geochemical and isotopic signatures of tremolite-bearing rocks suggest a genetic link with Early
34 Eocene supra-subduction dykes and fluids that leached them. Calcium, strontium and REE-bearing
35 oxidized fluids reacted with already serpentinized fracture wall rock as shown by fibre nucleation,
36 chromite alteration and high Cr contents in tremolite. The bulk of syn-tectonic fluid-rock interaction
37 associated with shear and tensile fracture development probably occurred at the onset of Eocene
38 intra-oceanic subduction, when the buoyant lower plate (the South Loyalty Basin) obliquely forced its
39 way beneath the nascent Loyalty fore-arc. Mobile elements extracted from supra-subduction dykes
40 temporarily enriched the circulating fluids and generated tremolite, antigorite, Mg-chlorite and
41 magnetite instead of antigorite and magnetite solely. Tremolite crystallization probably ceased due
42 to the exhaustion of Ca-rich fluids in a globally cooling fore-arc environment.

43

44 **Keywords**

45 Ophiolite, tremolite, antigorite, veins, metasomatism, supra-subduction, dykes

46

47 **1. Introduction**

48 Most ultramafic ophiolites are interpreted to represent supra-subduction mantle (Pearce et al.
49 1984) having recorded multiple events related to oceanic accretion, cooling, basin inversion and fore-
50 arc evolution. Fluid and melt release through dehydration, decarbonation and partial slab melting
51 within subduction zones is responsible for profound modifications of the supra-subduction mantle,
52 which are overprinted on previous metasomatic events. Hydration of supra-subduction mantle
53 peridotites (Wang et al. 2009; Mothersole et al. 2017) results in the occurrence of hydrous silicates of
54 the serpentine and amphibole groups, which are difficult to distinguish from those formed near the
55 ridge during the cooling of the ocean lithosphere (Mével 2003; Morishita et al. 2009). In contrast,
56 melts appearing during the early stages of arc formation (fore-arc melts) display diagnostic features
57 that may be a clue for identifying supra-subduction events.

58 The ultramafic allochthon of New Caledonia, termed Peridotite Nappe (Avias 1967; Fig. 1), is
59 formed of partly or totally serpentinized peridotites, which have evolved from a back-arc basin ridge
60 into a fore-arc setting, before being obducted (Cluzel et al. 2012). The Peridotite Nappe is crosscut by
61 a spaced vein network, which contains fibrous minerals that mainly belong to the serpentine group
62 (antigorite, chrysotile) and more rarely to the amphibole group (anthophyllite, tremolite). Most
63 antigorite and tremolite occurrences correspond to synkinematic fibrous seals associated with the
64 opening of tension cracks, or motion along micro-faults. However, some tremolite occurrences
65 appear in irregular streaks and pockets without obvious link to faults, and partly correspond to
66 metasomatized peridotites, which contain remains of their protolith such as chromite streaks
67 (Lahondère et al. 2012).

68 In weakly serpentinized lherzolites of the Peridotite Nappe, calcium content is relatively high (1.5
69 wt% to 2.2 wt% CaO) (Secchiari et al. 2016). It is lower in un-serpentinized harzburgites (0.3 wt% to
70 0.6 wt% CaO) (Secchiari et al. 2019). However, occurrence of such fresh rocks is extremely rare and
71 the bulk of the Peridotite Nappe underwent 40% to 100% serpentinization (lizardite mesh) and
72 contain less than 0.1 wt% CaO (0.01%-0.1%), a general feature of serpentinized peridotites
73 (Kodolányi et al. 2000). In spite of this, tremolite, a Ca-rich amphibole, is not unusual in serpentinized
74 ultramafic rocks and occurs in tectonic melanges, unroofed abyssal peridotites, transform fault zones
75 and fore-arcs (e.g., Kodolányi et al. 2000). Tremolite occurrences and hence Ca input have been
76 attributed to mobilization from the peridotite itself or, alternatively, external sources. As far as an
77 external source is needed, tremolite crystallization may result from the interaction of serpentinite
78 with Ca-rich metasediments in mélanges (i.e., nephrite jade; Adams et al. 2007) or with mafic rocks in
79 subduction zones (Marocchi et al. 2010), from the recrystallization of dykes crosscutting
80 serpentinites (e.g., rodingites; Bach and Klein 2009; Harlow and Sorrensen 2005; Koutsovitis et al.
81 2013; Sivell and Waterhouse 1986), from mobilization of Ca from pyroxenes during serpentinization
82 (Python et al. 2011), or "hydrothermal" remobilisation of Ca from calcite veins coming from sea

83 water at shallow depth (Agrinier et al. 1996). In New Caledonia, in addition to crosscutting veins,
84 tremolite locally appears in the lower crustal section of the Peridotite Nappe in greenschist-facies
85 mylonitic gabbro-norite cumulates (Ouen Island “jade”; Maurizot and Lesimple 2012). However, in
86 these rocks tremolite displays strong preferred orientation but never shows asbestiform texture. In
87 contrast, tremolite occurrences dealt with in this article are always located in serpentinized
88 peridotite, away from any mafic crustal rock.

89 In a general fashion, fibrous crack seals are nucleated on wall rock and the nature of minerals that
90 fill the progressively opening spaces depends strongly on the composition of the host rock. There is a
91 consensus on the fact that synkinematic minerals, which form the crack seals, come from pressure-
92 or shear-solution and are in equilibrium with pore fluids. Thus, synkinematic mineral growth is at
93 least partly fed by dissolved elements that are not carried over large distances (e.g., Baghat and
94 Marshak 1990; Beach 1974, 1977; Bons and Jessel 1997; Durney 1972, 1976; Pollard and Segall 1987;
95 Rutter 1983). Accordingly, only serpentine minerals (antigorite, lizardite, chrysotile, polygonal
96 serpentine, depending upon P-T conditions) are expected to be in equilibrium with the more or less
97 deeply serpentinized peridotite wall rock. This assertion has been confirmed by recent studies
98 (Andreani et al. 2007; Kurat et al. 2011); therefore, owing to the very low Ca content of depleted
99 peridotite, the occurrence of faults and cracks filled with tremolite in the Peridotite Nappe raises the
100 question of the composition and the origin of the associated fluids.

101 No associated sediments are known in the Peridotite Nappe that could be a source for Ca. Scarce
102 carbonate veins are documented in the Peridotite Nappe (e.g. Lahondère et al. 2012; Mothersole et
103 al. 2017) and clearly post-date the bulk of serpentinization. Generally these veins are composed of
104 magnesite or hydromagnesite, and although aragonite has been observed locally (Lahondère et al.
105 2012) it is related to late (post-obduction ?) joints infill of supergene origin.

106 Therefore, there are three internal and external possible sources:

- 107 i. Calcium may come directly from fluids emitted by the subduction zone.
- 108 ii. Although harzburgites and dunites of the Peridotite Nappe are strongly depleted (even
109 un-serpentinized rocks - a very rare occurrence though - yield very low Ca contents), they could be a
110 source of Ca for tremolite, provided the leaching of large amounts of rock.
- 111 iii. Early Eocene felsic dykes (up to 17 wt% CaO; Cluzel et al. et al. 2006) emplaced in already
112 fractured and serpentinized rocks commonly display disequilibrium features related to syn- and post-
113 magmatic tectonics (Rodgers, 1973a, b, 1974; Cluzel et al. 2006; see below), which suggest easy fluid
114 circulation through the fracture network and a possible source for Ca.

115

116 Tracking the origin of the tremolite-crystallizing fluids has been attempted using four methods: i)
117 geological characterization of the geometry and kinematics of the veins; ii) major elements, trace
118 elements, and REE geochemistry of tremolite-bearing veins; iii) Sr isotopes (tracer of Ca origin); and

119 iv) oxygen ($^{18}\text{O}/^{16}\text{O}$) and hydrogen (D/H) isotope ratios. As far as possible, tremolite features have
120 been compared with that of fibrous minerals of the serpentine group (antigorite), which are
121 commonly associated with tremolite in peridotite massifs.

122

123 **2. Geological setting**

124 The geology of New Caledonia is characterized by: i) a pre-Late Cretaceous basement formed of
125 amalgamated arc-derived terranes (Meffre 1995), which relate to the Southeast Gondwana margin;
126 ii) a Late Cretaceous to Late Eocene sedimentary cover; iii) a Eocene high-pressure metamorphic belt,
127 exposed in the northern part of the island; and, iv) two large allochthonous terranes emplaced during
128 the Eocene, the Poya Terrane and the Peridotite Nappe, which together represent almost one half of
129 the island's surface (Fig. 1) (Cluzel et al. 2012; Maurizot et al. in press a).

130 The Peridotite Nappe of New Caledonia (Avias 1967), results from obduction of the Loyalty Basin
131 lithosphere over the continental Norfolk/New Caledonia Ridge (Collot et al. 1987). It originally
132 covered most of the island; however, several phases of erosion isolated tectonic klippe spread along
133 the west coast and a larger unit termed "Massif du Sud" in the south of the island. A highly sheared
134 serpentinite sole, 20-200 m thick, indicates top-to-the-southwest emplacement kinematics (Quesnel
135 et al. 2013, 2016), while thrust-related deformation rapidly decreases upward.

136

137 The Peridotite Nappe is dominantly composed of harzburgite (>80%), dunite and minor lherzolite
138 (in northern massifs only) that represent a prominently depleted supra-subduction mantle
139 lithosphere (Marchesi et al. 2009; Pirard et al. 2013). Owing to their depletion in incompatible
140 elements compared to primitive mantle composition, residual harzburgites probably underwent
141 about 20%-25% partial melting (Prinzhofer 1981; Secchiari et al. 2019). They are overlain by dunite,
142 pyroxenite, websterite and gabbro-norite cumulates (Prinzhofer 1981). Cumulate websterite and
143 gabbro-norite probably crystallized in equilibrium with depleted melts intermediate between boninite
144 and island-arc tholeiite (Marchesi et al. 2009; Pirard et al. 2013; Cluzel et al. 2016; Secchiari et al.
145 2018), which at present are not preserved. Whole rock and mineral chemical constraints on
146 lherzolites allow a polyphase evolution to be drawn, from re-enrichment by circulating melts during
147 oceanic accretion to mantle metasomatism during subduction (Marchesi et al. 2009; Secchiari et al.
148 2016; Spandler and Pirard 2013; Ulrich et al. 2010).

149 The inception of intra-oceanic subduction that eventually led to obduction is time-constrained by
150 the granulite-facies "metamorphic sole", dated at 56 Ma (Cluzel et al. 2012a). Afterwards, the
151 Peridotite Nappe was crosscut by a series of dykes and sills emplaced at ca. 55-50 Ma (Early Eocene;
152 Cluzel et al. 2006), which are not present in the underlying Poya Terrane. These intrusive rocks
153 comprise minor dolerite dykes, and a variety of medium to coarse grain rocks (sills and dykes), the
154 compositions of which vary from ultramafic (pyroxenite and hornblendite) to felsic (diorite,

155 leucodiorite and granite). Dolerite dykes crosscut all the levels of the Peridotite Nappe, including
156 gabbro cumulates, while felsic and intermediate dykes and sills are apparently restricted to the
157 ultramafic part of the allochthon. Dolerites display supra-subduction affinities (island-arc tholeiites),
158 and likely represent the product of partial melting of “normal” supra-subduction mantle. In contrast,
159 the majority of felsic or intermediate dykes come from slab melts formed by partial melting of
160 diverse mafic oceanic crust rocks including gabbro cumulates; some others, however, may be
161 referred to as boninite-series generated by hydrous melting of highly depleted mantle wedge
162 peridotites enriched by slab melts and fluids (Cluzel et al. 2006, 2016; Secchiari et al. 2018).

163 Thin tremolite coatings around high-temperature minerals (olivine/orthopyroxene) and
164 crosscutting tremolite needles, which have been rarely found in unserpentinized harzburgites
165 (Secchiari 2016; Secchiari et al. 2019), signal early Ca input by circulating fluids or melts. Rocks of the
166 Peridotite Nappe are serpentinized at various degrees (0% to 100%) and this early tremolite
167 crystallization did not survive the development of lizardite-brucite (\pm chrysotile) mesh and tremolite
168 now only appears in syntectonic or replacement veins in serpentinized peridotites.

169 The development of serpentinization at large scale is crucial for Ni ore-forming processes during
170 tropical weathering (e.g. Orloff 1968; Pelletier 1996; Butt and Cluzel 2013; Maurizot et al. in press b)
171 because Ni substitutes Mg to form Mg-Ni silicates (the garnieritic ore) and has been systematically
172 described by mining geologists. Since the work of Orloff (1968) in New Caledonia, there has been a
173 consensus about the timing and origin of serpentinization, which was considered to be primarily
174 related to Eocene obduction and decreasing in intensity from the serpentinite sole upwards.
175 However, this view, which fits well with serpentinization at a large scale, is challenged by the local
176 observations of unserpentinized peridotites in which incipient serpentinization develops downwards
177 independently of the distance to the tectonic sole.

178 Thus, three main episodes of serpentinization may be defined: i) primary (intra-oceanic)
179 serpentinization is thought to have formed during cooling and hydration of mantle lithosphere; ii)
180 secondary, upward decreasing serpentinization was likely due to the subduction/obduction process;
181 iii) finally, continuing (minor?) serpentinization accounts for the local occurrence of hyper-alkaline
182 springs (Monin et al. 2014), somewhat similar to the Lost City oceanic geothermal field (Rivizzigno et
183 al. 2001). The products of present serpentinization are unknown and could only be reached by
184 drilling.

185 Serpentine formed by “primary” serpentinization and that due to subduction/obduction are
186 dominated by lizardite and somewhat difficult to discriminate where they overprint each other. The
187 only difference holds in the large amount of chrysotile, which is apparently a feature of the tectonic
188 sole.

189 With the exception of the tectonic sole, the Peridotite Nappe is weakly deformed and displays m-
190 to dm-spaced joints with only scarce evidence for shearing motion. Most of these joints have

191 serpentinized boundaries mm- to dm-thick, composed of sheeted seams of dark cryptocrystalline
192 lizardite and brucite crosscut by minor chrysotile veins. From the serpentinized fractures outwards,
193 lizardite seams are connected to the intra-crystalline serpentine mesh of olivine, which forms 40% to
194 100% of the bulk rock volume. In extremely scarce locations, peridotites have escaped pervasive
195 serpentinization and allow the observation of its progressive development. These weakly
196 serpentinized areas display upward, downward and laterally increasing amount of serpentinization
197 without strong associated deformation. Fresh olivine grains are crosscut by closed micro-cracks (Fig.
198 2a), which probably resulted from cooling-related differential shrinkage. Interconnection of these
199 micro-cracks with lizardite-bearing well-organized fractures (Fig. 2b) suggests that these structures
200 have been controlled by larger-scale joint development (Fig. 2c & 2d). Serpentinization progressed
201 from the fracture walls outward without significant relative motion and the occurrence of almost
202 undeformed mineral grains such as pyroxene (bastite) or chromite forming bridges across the
203 serpentinized fractures show that the latter were not significantly open and serpentinization was
204 dominantly static. The process of serpentinization may have varied through time and from place to
205 place and secondary minerals show some compositional variation due to protolith, fluid
206 composition, temperature and variation of the fluid/rock ratio (for a review, see Agrinier et al. 1996;
207 Frost et al. 2013 and references therein). It is worth noting that “primary” serpentinization (i.e.
208 lizardite-forming) left chromite grains unaltered; in contrast, chromite grains in antigorite-tremolite
209 veins frequently display chromian magnetite pseudomorphs (see below).

210 In contrast with static (or primary) serpentinization, which never gave birth to Ca-bearing silicates,
211 the crystallization of fibrous antigorite (and minor chrysotile) and tremolite occurred when
212 serpentinized fractures were reactivated by opening perpendicular or oblique to fracture walls filled
213 with synkinematic fibrous minerals (Fig. 3). Antigorite-bearing veins are widespread in the Peridotite
214 Nappe (e.g., Lahondère et al. 2012; Quesnel et al. 2016). They commonly reach several metres in
215 length and bear fibrous crack seals several mm to dm long. The angle between fibres and vein walls is
216 generally acute (45-25°) and denote oblique opening. Vein boundaries are generally composed of
217 dark lizardite, on which antigorite fibres are nucleated (Fig. 3b); but in some other localities, the vein
218 boundaries are made of porcelain-like whitish serpentine seams, which may result from the leaching
219 or recrystallization of lizardite vein walls.

220 Antigorite-tremolite and pure tremolite veins display the same characteristics; however, tremolite
221 fibres are shorter and commonly perpendicular or strongly oblique to vein walls (Fig. 3c & 3d). In all
222 cases the development of antigorite and tremolite post-dated the lizardite-bearing “primary”
223 serpentinization.

224

225 **3. Analytical procedures**

226 Because it may have fibrous, asbestiform habits (Dorling and Zussman 1987), and frequently
227 shows intergrowth with antigorite (this study), tremolite was sometimes overlooked in the field.
228 Therefore, lab analysis (i.e., polarized light microscopy, XRD, IRS, Raman spectrometry) was often
229 necessary to distinguish the various asbestiform minerals.

230 Raman data were acquired with a Renishaw inVia spectrometer of the Geology Lab. of Ecole
231 Normale Supérieure de Paris. The spectrometer is fitted with a Physics Argon laser delivering
232 monochromatic coherent light beam of 20mW at 514.5 nm (green laser). The laser beam is focused
233 through a Leica DMLM microscope fitted with 4 short focus lenses (x5, x20, x50 (Numerical
234 aperture=0.75), x100 (NA=0.90)) and 2 long focus lenses (x50 and x100). In the most effective
235 configuration, the resolution was in the order of 1 μ m for 1mW power at the sample surface so that
236 any irradiation damage may be avoided. The Rayleigh diffusion (< 100 cm⁻¹) was eliminated by
237 dielectrical filters. The Raman signal was then spread with a network of 1800 lines mm⁻¹ (or 3000
238 lines mm⁻¹) and measured with a CCD RENCAM detector of 400 x 576 pixels cooled by Peltier effect.
239 The microspectrometer is fitted with a motorized Prior Scientific stage, which allowed moving across
240 the sample

241 Geochemical whole-rock analyses were performed at Service d'Analyse des Roches et Minéraux
242 (CNRS-CRPG, Vandoeuvre, France). Major elements were analysed by ICP-AES, trace and Rare Earth
243 elements by ICP-MS (a description of analytical procedures and detection limits may be obtained on
244 the SARM web site: <http://helium.cprg.cnrs-nancy.fr/SARM/>).

245 Single minerals major elements concentrations have been determined on polished thin sections
246 with a Cameca SX five electron microprobe (W beam source) of the analytical platform of University
247 of Orleans/CNRS/BRGM.

248 Strontium isotope analyses have been carried out at Clermont-Ferrand isotope laboratory of
249 Laboratoire Magmas et Volcans (UMR 6524 CNRS-Université de Clermont-Ferrand, France) by TIMS
250 using a Thermo Fischer Triton instrument, corrected for mass fractionation by normalisation to
251 ⁸⁶Sr/⁸⁸Sr = 0.1194. Strontium sample decomposition and chemical separation followed the procedure
252 developed by Pin and Santos Zalduegui (1997). Strontium isotope ratios were corrected for in-situ
253 decay at 53 Ma, based upon the average U-Pb zircon age of dykes (Cluzel et al. 2006). Concentrations
254 of Sr are given in μ g.g⁻¹. The ⁸⁷Sr/⁸⁶Sr ratio measured for the NIST SRM 987 standard during the
255 period of analyses gave a mean value of 0.710238 \pm 9 (2 σ , n = 30).

256 Oxygen isotope compositions were measured on dried silicates reacted using BrF₅, following the
257 method of Clayton and Mayeda (1963). Isotopic compositions were measured on CO₂ with a VG
258 Optima triple collector mass spectrometer. Measured isotopic compositions were normalized to NBS
259 28 standard (9.6 ‰). The reproducibility is 0.25‰.

260 Hydrogen isotope compositions were analyzed at the CRPG UMR 7358 CNRS-UL (Nancy, France)
261 using an Eurovector Elemental Analyzer (EA) coupled to a VG Isoprime Isotope Ratio Mass

262 Spectrometer (IRMS), following the procedure described in Lupker et al. (2012), Bell and Ihinger
263 (2000) and Bigeleisen et al. (1952). Powdered samples were loaded into tin capsules and degassed at
264 120 °C under vacuum for 48 h in a degassing canister to remove adsorbed water. After dehydration,
265 samples were transferred under a N₂ dry atmosphere in a customized, sealed, automatic sampler
266 pre-flushed with He in order to avoid any contact with atmospheric moisture. This sampler was then
267 connected to the EA-IRMS system. Samples were combusted at 1450 °C on an EA glassy Carbon
268 reaction tube, filled with glassy carbon chips, leading to the reduction of hydroxyl groups to H₂. After
269 chromatographic separation, the recovered H₂ was introduced in the IRMS source and analyzed for
270 hydrogen isotopic composition. Three different internal standards (Muscovite: MuscD65, Phlogopite:
271 Mica-Mg and a fine grained marine sediment from the Bay of Bengal: SO188) were included during
272 the sample analyses. Values of δD are reported in standard per mil (‰) notation relative to SMOW
273 (Standard Mean Ocean Water). The overall, long term, 1σ, reproducibility of the method on silicate
274 powders is given at 2‰ for δD.

275 Mineral abbreviations used in figures of this article follow recommendations by the IUGS
276 Subcommittee on the Systematics of Metamorphic Rocks: Web version 01.02.07 according to Kretz
277 (1983) completed by Siivola and Schmid (2007).

278

279 **4. Veins and dykes petrography and structure**

280 **4.1. Tremolite-antigorite veins**

281 In addition to widespread lizardite mesh and lizardite-coated joints formed in relatively static
282 conditions, which can be referred to “primary serpentinization” (see above), peridotites of the
283 Peridotite Nappe are crosscut by tension and shear cracks filled with fibrous serpentine and
284 amphibole. They appear independently of their location in the Peridotite Nappe and especially have
285 no relation to the tectonic sole. Acicular/fibrous crack seals composed of antigorite (± chrysotile),
286 tremolite, or both, are nucleated on lizardite seams (Fig. 3c, 3d, 4d, 5) and thus post-date them.

287 Fibrous amphiboles (anthophyllite and tremolite) are mainly present in Poro, Thio and Tiebaghi
288 areas, but also appear in many other sites (Fig 1). In the Poro area (east coast) crack seals are filled
289 with tremolite often associated with antigorite (Fig. 6), whereas in the Tiebaghi area vein infills are
290 dominated by anthophyllite (Lahondère et al. 2012). In most cases, Raman analysis only allowed the
291 determination of intimately intergrown tremolite and antigorite (Fig. 6 and 7). In the Massif du Sud
292 crack infills comprised tremolite, anthophyllite and Mg-chlorite. Anthophyllite reaction rims generally
293 appear at the margin of some felsic dykes and around peridotite enclaves within the same dykes.
294 Crack seals with Mg-rich amphiboles appear closely related to felsic dykes in the Thio area (east
295 coast). Microfaults with synkinematic fibrous tremolite and anthophyllite coatings crosscut dykes
296 related to the lower Eocene dyke complex (Thio, Bogota Peninsula) (Lahondère and Maurizot 2009 ;
297 Lahondère et al. 2011, 2012). Thus, field evidence and radiochronologic data from felsic dykes

298 suggest that amphibole-bearing veins, which are more or less associated with felsic magmatic dykes
299 formed after 55 Ma.

300 Most synkinematic fibrous or acicular antigorite and tremolite crystals that appear in cm- to dm-
301 thick open cracks and dikelets are:

- 302 - either strongly oblique or perpendicular to fissure walls and thus resemble tension cracks
303 (Fig. 3c)
- 304 - or, slightly oblique to fissure walls and very much resemble to pull-apart crack seals
305 associated with micro-faulting (oblique slip) (Fig. 3d)

306 These well-known tectonic features of brittle and semi-brittle rocks, which are extensively used
307 for structural analysis, are generally filled with quartz, calcite and other minerals, depending on the
308 composition of host rock (quartzite, limestone, etc.), which dominantly come from pressure/shear
309 solution. Therefore, analogous process is postulated in serpentinized peridotites in which serpentine
310 minerals are formed at medium to low temperature in the presence of water. In any case, these
311 open tension or shear cracks are dilatational features, which likely record low lithostatic, or
312 alternatively, relatively high fluid pressure.

313 In many places, tensile and shear fractures are closely associated and their fibres are aligned in
314 the same direction, which represents the direction of maximum finite stretch ("X" axis of the local
315 strain ellipsoid). When the vein thickness is larger than 5-10 cm, tremolite fibres are frequently
316 organised in radiate or sheaf-like aggregates (Fig. 3a), which likely record fracture opening faster
317 than synkinematic crystal growth. In some occurrences, first phase synkinematic fibrous crystals are
318 micro-folded or crenulated during a second tectonic phase with different kinematic axes. It is worth
319 noting that in this case, the second tectonic event generated antigorite only. Tremolite also appears
320 in poly-mineral associations with antigorite, Mg-chlorite and magnetite in heaps and reaction
321 structures.

322

323 **4.2. Lower Eocene dykes**

324 Lower Eocene supra-subduction magmas intruded the pre-existing fracture network as shown by
325 the zig-zag geometry of some dykes (Cluzel et al. 2006); therefore, a relationship with
326 serpentinization may be suspected (e.g., Python et al. 2011). However, not all dykes display lizardite
327 walls and some of a zigzag geometry, display serpentinized and unserpentinized walls, depending
328 upon fracture orientation. The only (scarce) occurrences of reaction rims around Eocene dykes are
329 composed of relatively coarse-grain anthophyllite or Mg-chlorite. Therefore, lizardite "black walls"
330 formed before dyke intrusion and cannot be related to peridotite-magma interaction.

331 In a few locations (Poro, Kopeto) dm- to m-thick "dykes" are composed of whitish massive
332 material dominated by tremolite. Samples from these localities display high Ti and Al contents (eg.
333 sample POR1b, see below) and are probably deeply altered HFSE-depleted felsic dykes (Cluzel et al.

334 2012), which contain no zircons that could be dated. Remarkably, the two tremolite types,
335 synkinematic and reaction-type coexist on these outcrops, a fact that suggests a genetic relationship.
336

337 **4.3. Structural analysis of lower Eocene dykes and tremolite-antigorite veins**

338 Crosscutting dykes are found at all levels of the ophiolite, except in gabbronorite cumulates,
339 which are crosscut by IAT dolerites only. Strongly fragmented dykes are also present in the
340 porphyroclastic serpentinite sole, whereas they are well preserved in the rest of the Peridotite
341 Nappe. It is worth noting that in contrast with Late Oligocene granitoids (Cluzel et al. 2005; Paquette
342 and Cluzel 2007) Eocene dykes never crosscut the parautochthonous or autochthonous basement of
343 the ophiolite, a feature consistent with their pre-obduction character. Most dykes trend NW-SE (av.
344 N140°E) and NNE-SSW (av. N30°E), and some sills intruded parallel to the peridotite foliation. Their
345 orientations are roughly consistent with the main structural trends of the Peridotite Nappe and
346 locally, sub-parallel to antigorite and tremolite-bearing veins, but they rarely coexist. Most
347 ultramafic, mafic and felsic intrusive rocks are medium to coarse grained and locally display
348 pegmatite texture; in contrast, dolerites are fine-grained and often display chilled margins. Thin
349 medium-temperature reaction rims with anthophyllite, talc, and chlorite, develop around most
350 coarse-grained felsic dykes. In contrast, chilled margins of the younger dolerite dykes unambiguously
351 show that they were emplaced at lower host rock temperatures. Locally (Massif du Sud, Ouen Isl.),
352 coarse-grained diorite and hornblendite dykes are severely deformed and display ductile strain
353 features including high-temperature foliation marked by amphibole preferred orientation, boudinage
354 and stretching, plagioclase-hornblende banding and locally isoclinal folding (Ouen Isl.), evolving
355 internally toward lower-temperature mylonite and cataclastic mylonite. Shallow dipping stretching
356 lineation and/or hornblende preferred orientation suggest strike-slip or oblique slip tectonics; i.e.,
357 sinistral-reverse motion along N150°E trending dykes (Soret et al. 2016). U-Pb dating of zircon gave
358 magmatic ages at ~55-52 Ma (Cluzel et al. 2006) close to ⁴⁰Ar/³⁹Ar hornblende cooling ages at ~55 Ma
359 (Soret et al. 2016). The occurrence of internal ductile textures suggests synkinematic intrusion in a
360 transcurrent tectonic regime. This event is thus slightly younger than amphibolites of the
361 metamorphic sole, which record subduction initiation at ca. 57-56 Ma (Cluzel et al. 2012; Soret et al.
362 2016). It is worth noting that IAT dolerite dykes, which occur at about the same level of the Peridotite
363 Nappe never display such tectonic features and thus post-date this event at ca. 50 Ma (Cluzel et al.
364 2012; Cluzel and Jourdan unpubl. ⁴⁰Ar/³⁹Ar data). These data also suggest significant unroofing of the
365 Peridotite Nappe during the 55-50 Ma interval.

366 Most dykes apparently escaped the mineral transformations that commonly affect the host
367 peridotite and have well preserved magmatic mineralogy; however, some display secondary
368 development of epidote, albite and green amphibole (Rodgers 1973a, 1973b; Rodgers and Bevan
369 1974; Maurizot and Lesimple 2012) depending upon local thermal anomalies and/or fluid-rock ratio.

370 Therefore, the elemental mobility associated with greenschist alteration is likely to have occurred
371 during or soon after dyke emplacement.

372 The oblique arrangement of the kinematic axes deduced from antigorite and tremolite-antigorite
373 shear veins (Iseppi 2018) is consistent with the development of the oblique subduction postulated on
374 the basis of paleogeographic and tectonic evidence (Cluzel et al. 2001; Maurizot and Cluzel 2014).
375 Meanwhile, the development of “extensional” strain (i.e. shallow dipping stretching axis) in the
376 upper plate of the subduction/obduction system is consistent with stress orientation in the upper
377 plate in shallow subduction settings (Chemenda et al. 2000; Espurt et al. 2008; Gutscher et al. 2000;
378 McCaffrey et al. 2000; McNulty and Farber 2002). Overall, it is consistent with the tectonic features
379 that may develop at subduction inception (Fig. 15).

380 At island scale, the fracture network of the Peridotite Nappe shows a N130°E dominant trend,
381 associated with subordinate N090°E, N045°E and N000°E directions (Leguéré 1976; Moutte and Paris
382 1977) that control most geomorphological features. At outcrop scale and near the base of the nappe,
383 peridotites are affected by dense and complex fracture sets, which are probably due to several
384 superimposed tectonic events during obduction and exhumation to shallow depth. Brittle failure at
385 low lithostatic pressure accounts for changing stress ellipsoids and complex joint patterns. In
386 contrast, antigorite and tremolite-antigorite veins, which characteristically have several meters to
387 several tens of meters spacing and have been filled at relatively high temperature, display consistent
388 structural orientations and likely originated from the same tectonic events independently of
389 obduction.

390 Tremolite and antigorite appear in tension cracks, strike-slip, normal or reverse faults related to
391 the frictional reactivation of pre-existing fractures. Inversion of fracture data from the Massif du Sud
392 yields a sub-horizontal stretching axis of the strain ellipsoid with a roughly N-S ($\pm 20^\circ$) orientation. In
393 contrast, in the klippe of the northwest coast (Koniambo and Tiebaghi), the stretching axis is
394 oriented WNW-ESE (Iseppi 2018). In general, the shortening axis, although variably oriented, is
395 steeply dipping. The strain ellipsoid denotes transtensional setting in the upper plate of the
396 subduction consistent with inception of oblique subduction (Gurnis et al. 2004; Gutscher et al. 2000).

397

398 **5. Analytical results**

399 **5.1. Whole-rock geochemistry**

400 Sampling has been undertaken in several nickel open pit mines throughout New Caledonia and
401 ten samples from the Poro mine (east coast) judged representative, have been selected for chemical
402 analysis (Table 2). Seven of these samples had been identified in the field as tremolite and three as
403 antigorite taken for comparison. It appeared thereafter that most samples were not pure mineral
404 phases but actually a mix of tremolite and other minerals (talc-chlorite, antigorite). By comparison

405 with theoretical tremolite, only two samples are close to the pure end-member with only a limited
406 amount of Fe/Mg substitution, which accounts for the pale green colour of some prismatic crystals.

407 In a general fashion, trace elements contents in serpentinized harzburgite and dunite are very
408 low; similarly, REE contents of most antigorite samples are extremely low and below the detection
409 limit of routine analysis. However, the tremolite-bearing samples have contents higher than
410 serpentines; specifically, REE could have been analysed with a good accuracy by ICP-MS analysis. The
411 specific behaviour of Eu is worth noting; REE patterns of tremolite-bearing veins display positive Eu
412 anomalies (Fig. 8), which contrast with the negative anomaly of most felsic dykes due to feldspar
413 fractionation.

414

415 **5.2. Mineral chemistry**

416 Microprobe analyses of syntectonic tremolite needles/fibres on polished thin sections show some
417 variability without clear connection with location or textural features. On the Fe/(Mg+Fe²⁺) vs. total
418 Si binary diagram (Leake 1968, Leake et al. 1997; Hawthorne et al. 2012), the analysed amphiboles
419 (Table 3) dominantly plot in the domain of tremolite and alternatively in the tremolite-hornblende
420 domain (Fig. 9) with a strong negative correlation between Mg# and total Si. Chromium content also
421 displays a close negative correlation with Si content (Fig. 10a) and a positive correlation with X_{FeA} (Fig.
422 10b), which both are indicative of increasing T° of crystallization. High-Mg chlorites associated with
423 tremolite in some veins (0.29 < Fe_{tot} < 0.34; 9.67 < Mg < 9.92) may be termed penninites according to
424 Hey's classification (Hey, 1954); they are referred herein to as Mg-chlorites.

425

426 **5.3. Strontium isotopes**

427 The ⁸⁷Sr/⁸⁶Sr ratio is a possible marker of the origin of Ca in tremolite because Ca and Sr display
428 similar geochemical behaviour. The very low Ca and Sr content of antigorites makes the analysis
429 difficult as Sr contents are often below the detection limit of routine analysis (0.1 ppm). Therefore,
430 more analyses have been performed on purified minerals to get more accurate data to build the Sr
431 vs. ⁸⁷Sr/⁸⁶Sr diagram. On the Sr and Ca vs. ⁸⁷Sr/⁸⁶Sr diagrams (Fig. 11; Table 4), tremolite-bearing veins
432 plot in a narrow array with a negative slope (Fig. 11). The ⁸⁷Sr/⁸⁶Sr ratios of almost pure tremolite
433 (CaO= 9-12 wt%; Sr= 4-9 ppm) are in the range of 0.7042-0.7045. In contrast, antigorites, which have
434 very low Ca (0.002 wt%-0.01 wt% CaO) and Sr contents (0.03 ppm-1.4 ppm) display a wider range of
435 highly radiogenic ⁸⁷Sr/⁸⁶Sr ratios (0.7062-0.7116) without clear correlation with Ca or Sr contents.

436

437 **5.4. Oxygen and hydrogen isotopes**

438 Tremolite and serpentine minerals likely were in isotopic equilibrium with circulating/pore water
439 during their crystallization; therefore, isotopic compositions of minerals reflect that of water
440 (Méheut et al. 2010). Fresh tremolite and antigorite were carefully selected by hand picking and

441 analysed for oxygen and hydrogen isotopes, compared to weathered specimens and data from Ni-
442 rich and Ni-poor supergene silicates: "garnierite" (an informal mixture of Ni-rich serpentine, talc and
443 smectite) of New Caledonia (Amisse et al. 2010); and deweylite (Wenner and Taylor 1974) (Table 5).
444 Isotopic ratios were plotted on the δD vs. $\delta^{18}O$ diagram (Fig. 12). Fresh tremolites (av. $\delta^{18}O = 6.8\text{‰}$;
445 $\delta D = -54.8\text{‰}$) and antigorites (av. $\delta^{18}O = 6.4\text{‰}$; $\delta D = -46.0\text{‰}$) from this study plot in a narrow field
446 similar to that of "continental antigorites" of Wenner and Taylor (1974); in contrast, tremolite and
447 antigorite from the weathering profile of the same sites, have lower δD (-60‰ to -110‰) at
448 somewhat constant $\delta^{18}O$. Fresh tremolite and antigorite have higher $\delta^{18}O$ and lower δD than
449 supergene silicates (Amisse et al. 2010).

450 Serpentinites of the tectonic sole characteristically display supergene isotopic signatures (Ulrich
451 2010), which may due to infiltration of meteoric waters during obduction (Quesnel et al. 2013, 2016)
452 or alternatively, post-obduction (i.e. post-kinematic) rock-water interaction. In any case, field
453 evidence shows that tremolite-bearing veins predate and have no genetic relationship to the brittle
454 shearing and fracture development related to obduction. Therefore, fresh tremolite and antigorite
455 veins have been most likely formed in endogenous conditions and did not interact with meteoric
456 water.

457

458 **6. Discussion**

459 Tremolite- and antigorite-bearing veins from the Peridotite Nappe of New Caledonia display
460 geochemical and isotopic features that drastically contrast with their host rocks. Prominent
461 enrichment of tremolite-bearing veins in some major and trace elements compared to serpentinitized
462 peridotites, and the synkinematic character of fracture infill suggest that they likely formed at depth
463 through a metasomatic process in connection with minor tectonic events. Their geochemical and
464 isotopic composition thus results from the interaction of several potential solid and fluid sources, the
465 role of which may vary through time. Solid sources are represented by the variably serpentinitized
466 peridotite host rocks and the lower Eocene dykes which crosscut them, while fluid sources may be
467 the fluids that circulated in the supra-subduction mantle; i.e., mostly aqueous fluids released by slab
468 dehydration.

469

470 **6.1. Textural features, relative chronology and temperature constraints**

471 Much attention has been paid to ultramafic terranes having undergone HP-LT metamorphism in
472 subduction/collision zones. For example, the stability fields of serpentine minerals have been mainly
473 defined at high pressure (e.g., Debret et al. 2013; Deschamps et al. 2013; Schwartz et al. 2013;
474 Padrón-Navarta et al. 2010, 2013). In contrast, data from low-pressure ultramafic terranes are
475 scarce, except those retrieved by ocean drilling programs, which generally correspond to serpentinite
476 bodies exhumed at magma-poor extensional systems or at transform faults. The Peridotite Nappe of

477 New Caledonia is a supra-subduction ophiolite (see geological setting above and Fig. 15) and
478 although there are no precise P-T constraints available, its evolution lacked high pressure conditions
479 and is probably similar to “oceanic” terranes at least during the first steps.

480 Syntectonic antigorite and tremolite-antigorite bearing veins crosscut and frequently reactivate
481 (re-open) lizardite veins formed during “primary” pervasive serpentinization. Instead of static
482 pseudomorphic replacement of the high-temperature parageneses (olivine + Opx) by relatively low-
483 temperature lizardite and brucite, they record shearing deformation due to a low-strain tectonic
484 event. Some tremolite-bearing veins display final crystallization of antigorite. Thus, a rough
485 chronology may be established as follows: 1) early tremolite (mostly erased); 2) static lizardite (\pm
486 chrysotile); 3) synkinematic antigorite or antigorite + tremolite, 4) synkinematic antigorite solely, 5)
487 supergene minerals.

488 In spite of some uncertainty on boundaries, this mineral sequence allows some constraints on the
489 evolution in temperature to be proposed. The upper stability limit of lizardite (\pm chrysotile) at low
490 pressure (< 0.4 GPa) is established to about 300-320°C (Evans, 2004), while antigorite stability starts
491 at about 320°C and persists up to 500-550°C (Wenner and Taylor, 1971). At low to medium pressure
492 (0.1-0.5 GPa) tremolite appears at 350-550°C and disappears at 400-650°C depending on the CO₂
493 partial pressure (Jenkins 1983; Chernosky et al. 1998; Evans 2004; Evans et al. 2000, 2013).
494 Therefore, the relative chronology above records: 1) a net increase in temperature after primary
495 serpentinization, during the formation of tremolite-antigorite-bearing veins; 2) a gentle temperature
496 decrease at the end of tremolite crystallization; and 3) a final temperature decrease down to
497 supergene conditions (ca. 50-35°C).

498 The total thickness of the Peridotite Nappe does not exceed 2 km; therefore, a depth-dependent
499 variation of temperature is unlikely and with the exception of chrysotile, which seems to be more
500 concentrated near the tectonic sole, there is no specific location for tremolite- or antigorite-bearing
501 veins.

502

503 **6.2. Selective input of incompatible trace-elements record of tremolite-bearing veins**

504 Tremolite-bearing veins are generally enriched by about 10 to 100 times in REE and incompatible
505 elements compared to peridotite host rock and commonly display positive Eu (Fig. 8) and negative
506 Zr-Hf anomalies (Fig. 13).

507 Positive Eu anomalies have been described in peridotite-hosted hydrothermal fluids (Allen and
508 Seyfrid 2003, 2005; Schmidt et al. 2007) and in serpentines of mid-oceanic ridges (German et al.
509 1999; Douville et al. 2002; Paulick et al. 2006, Ningthoujam et al. 2012) and thus, early hydrothermal
510 circulations could be a source for REE. However, Eu anomalies do not appear in serpentinized
511 dunites and harzburgites of the Peridotite Nappe and only secondarily silicified and carbonated rocks
512 of the serpentinite sole display bulk REE enrichment (~ 10 times) especially in LREE (Ulrich 2010; Fig.

513 7). Rare Earths Elements patterns of moderately silicified and carbonated serpentines
514 characteristically display Ce negative anomalies. This feature is diagnostic of REE enrichment of
515 supergene origin because Ce^{4+} is residual during oxidizing alteration and accumulates with hematite
516 on top of the weathering profile (Braun et al. 1990); therefore, supergene fluids circulating from the
517 weathering profile were probably Ce-depleted. Thus, high REE content of tremolite-bearing veins is
518 likely unrelated to supergene alteration and of probable hydrothermal origin, the hydrothermal
519 fluids having interacted with feldspar in order to acquire a positive Eu anomaly. Owing to the relative
520 chronology established above, REE enrichment most likely appeared after primary serpentinization,
521 during subsequent water-wall rock interaction.

522 Although tremolite-bearing veins have obviously not been generated by magmatic processes, the
523 contrasting behaviour of Eu compared to other REE is possibly a key for REE provenance. The REE
524 patterns of tremolite-bearing veins analysed in this study, display a positive Eu anomaly, which has
525 the same order of magnitude regardless of the bulk REE content. Eu anomalies in tremolite-bearing
526 veins may have been caused by precipitation of tremolite from solutions which had already
527 developed Eu anomalies. Therefore, a positive Eu anomaly in tremolite may possibly come from
528 reaction with fluids having altered a plagioclase-rich rock such as a gabbro cumulate or a felsic dyke.

529 Assessing the enrichment of tremolite veins in some trace-elements compared to
530 peridotites/serpentinites is not always possible because some trace-element abundances in the
531 latter are below the standard detection limit (Secchiari et al. 2019; table 2). When measurable, this
532 enrichment is not uniform and prominent depletion appears in HFSE (Nb, Ta, Zr and Hf), some of
533 which are below the detection limit of standard ICPMS whole rock analysis (Table 2). HFSE depletion
534 is a common feature of subduction zone magmatism, and generally interpreted to result from the
535 occurrence of HFSE-retaining residual minerals in the subducting slab (e.g. Garrido et al. 2005; Holm
536 et al. 2016; Munker et al. 2004 ; Rubatto et al. 2003; Woodhead et al. 2011); consequently, HFSE-
537 depleted fluids are transferred toward the mantle wedge. Therefore, fluids directly emitted by the
538 subduction could have circulated in the supra-subduction mantle (Fig. 14b) and generated tremolite-
539 antigorite veins with a similarly HFSE-depleted signature. However, strontium, oxygen and hydrogen
540 isotopic features of tremolite are not consistent with direct slab derivation and point to a
541 “magmatic” signature (see 6.4 and 6.6 below).

542 In contrast to most subduction zone magmatic rocks, Eocene supra-subduction dykes of the
543 Peridotite Nappe dominantly display strong positive Zr and Hf anomalies (~10 times) and contain
544 abundant magmatic zircons (Cluzel et al. 2006). Hydrothermal fluids circulating in fractures hosting
545 Eocene dykes may be relatively depleted in certain elements contained in refractory minerals and
546 this is possibly the case for Hf and Zr captured by zircon and other Hf- and Zr-retaining minerals. At
547 moderate temperature within the tremolite stability domain (<600°C), Ca and other mobile elements
548 may have been leached from dykes, while magmatic zircons remained unaffected, thus resulting in

549 Zr-Hf depletion in circulating fluids. In contrast, leaching of peridotites or felsic cumulates
550 (gabbro-norites), which do not contain zircon would not result in such a feature and thus can be
551 discarded. Preferential Ca leaching may also be expected by autometamorphism of some dykes. By
552 the end of crystallization, supercritical hydrous fluids originally contained in the magma (2% to 10%
553 depending upon magma composition) are ejected at sub-solidus temperatures. This process was
554 likely responsible for the appearance of secondary amphibolite to greenschist facies parageneses in
555 some dykes and favoured the development of ductile tectonic features (banding, preferred
556 orientation, isoclinal folding, mylonitization) within some dykes, which did not appear in the
557 peridotite host

558

559 **6.3. Origin of antigorite veins, constraints from Sr isotopes.**

560 The $^{87}\text{Sr}/^{86}\text{Sr}$ ratio in harzburgites and lherzolites of the Peridotite Nappe, which are the most
561 common wall rocks of antigorite and tremolite-bearing veins, covers a relatively large range (0.7049-
562 0.7077) (Secchiari 2016; Secchiari et al. 2019) and a certain role of “primary” serpentinization may be
563 suspected. However, there is no correlation between the LOI, which could represent the degree of
564 primary serpentinization and Sr isotope ratio of un-serpentinized (LOI= 0%-0.7%; $(^{87}\text{Sr}/^{86}\text{Sr})_i = 0.7077$ -
565 0.7054) and moderately serpentinized lherzolites and harzburgites (LOI= 6.0%-10.7%; $(^{87}\text{Sr}/^{86}\text{Sr})_i =$
566 0.7075-0.7049) (Secchiari et al. 2016; 2019). Thus, a moderate amount of primary serpentinization
567 does not significantly change the Sr isotope ratio of peridotites. Therefore, serpentinization cannot
568 be directly responsible for the radiogenic character and variability of the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of New
569 Caledonia’s peridotites, which are thus likely due to slab-derived magma input (Secchiari et al. 2019).

570 As far as shear- or pressure-solution origin is considered for antigorite veins, the Sr isotope
571 composition of antigorite is expected to reflect the variability of peridotite wall rock, pending some
572 influence of circulating fluids. However, the $^{87}\text{Sr}/^{86}\text{Sr}$ of antigorite veins is much higher than that of
573 peridotites (up to 0.7117), close to the present ocean water composition and even much higher than
574 that of lower Eocene waters (Murthy and Beiser 1968; Elderfield 1986; Veizer 1989; McArthur et al.
575 2012). These values are slightly lower than that of hemi-pelagic terrigenous sediments (e.g.
576 McLennan et al. 1990; Hemming et al. 2007) and infer sediment-water mixing/leaching. Thus, the
577 highly radiogenic $^{87}\text{Sr}/^{86}\text{Sr}$ ratios from antigorite veins (Fig. 11) may reflect some influence of
578 subducted sediments on the fluids emitted by the subduction zone, which infiltrated the supra-
579 subduction mantle and were involved in antigorite crystallization. The nature of involved sediments
580 remains unknown; however, Late Cretaceous sediments similar to those included in the Poya
581 Terrane; abyssal argillites associated with basalts and hemipelagic turbidites of the Kone Facies, are
582 good candidates for the source of high $^{87}\text{Sr}/^{86}\text{Sr}$ fluids. At that stage, there are no constraints on the
583 mechanisms and P-T conditions of fluid release from the slab.

584

585 **6.4. Origin of Ca in tremolite-bearing veins, constraints from Sr isotopes.**

586 Tremolites from veins and pockets plot in a narrow field with low $^{87}\text{Sr}/^{86}\text{Sr}$ (0.7042-0.7045), that
587 clearly differ from the isotopic signatures of lherzolites and harzburgites; thus, a direct origin of Ca by
588 leaching of peridotites is unlikely. Alternatively, tremolite seems to have been in equilibrium with
589 “magmatic” fluids (Fig. 11); a feature consistent with $\delta^{18}\text{O}$ and δH values (see below), even though a
590 certain influence of ocean-like water cannot be excluded. The “sedimentary” signature of antigorite
591 veins, possibly derived from subduction-related fluids (see 6.3 above), was probably diluted in
592 tremolite-bearing veins, which contain 100 to 1000 times more Sr than antigorite.

593 A comparison with exhumed serpentinites of the Atlantis Massif (Boschi et al. 2008) shows that
594 similar trends may exist in oceanic ridge environments; variable Ca-Sr contents at high and constant
595 $^{87}\text{Sr}/^{86}\text{Sr}$ ratio record metasomatism from oceanic water/sediments, while additional Ca-Sr
596 enrichment with decreasing $^{87}\text{Sr}/^{86}\text{Sr}$ signals some influence of (crustal ?) magmatic rocks (Fig. 11).

597 It is unlikely that tremolite formed by direct reaction of felsic magma with serpentinized wall rock,
598 because in all observed reaction rims around dykes, anthophyllite and/or Mg-chlorite are formed
599 instead of tremolite. In the frequent absence of direct contact between felsic dykes and tremolite
600 veins or pockets, it may be suggested that tremolite merely formed by reaction between hydrous
601 fluids released by the subduction zone and having leached the still hot felsic dykes and the
602 serpentinized wall rocks of active shears or tension cracks. This may account for the compositional
603 variability, REE contents 10 times lower than that of dykes and the presence of large amounts of
604 compatible elements taken from fracture walls. Thus, three end-members are likely involved in the
605 formation of tremolite: 1) an aqueous fluid emitted from the subduction zone and somewhat
606 influenced by sediments; 2) a REE-poor and compatible elements-rich ultramafic end-member that
607 displays variable Sr isotope signature; and 3) a REE-rich magmatic end-member with a mantle-like
608 isotopic signature, which very much resembles Lower Eocene dykes, or fluids evolved from these
609 dykes.

610

611 **6.5. High Cr contents and wall rock involvement**

612 An important geochemical feature of New Caledonia tremolites is the high content of compatible
613 trace elements such as Cr (Fig. 9). Cr is relatively immobile in hydrothermal and supergene systems
614 and should appear in tremolite with moderate contents only. Whole-rock analysis of tremolite-
615 bearing veins revealed high Cr contents (2,800 – 3,300 ppm), which may be due to Cr contained in
616 newly crystallized (syn-kinematic) minerals, or alternatively, to residual chromite. However,
617 microprobe analyses of tremolites display up to 2.6 wt% Cr_2O_3 (Table 3). Cr content correlates
618 negatively with total Si (Fig. 10a) and positively with Fe content ($X_{\text{FeA}} = 100(\text{Fe}+\text{Mn})/(\text{Fe}+\text{Mn}+\text{Mg})$)
619 (Fig. 10b), a feature that suggests increasing Cr incorporation with higher temperature. The
620 polyminerally tremolite-bearing veins and pockets coming from the replacement of peridotite along

621 fractures contain residual streaks of chromite, which display evidence for Cr mobility. Altered
622 chromites are generally composed of a residual chromite core rimmed by a microcrystalline
623 association of chromite and Cr-rich magnetite (Fig. 13). Fe-Cr substitution giving birth to Cr-rich
624 magnetite may be responsible for Cr mobility and its incorporation into tremolite. Therefore, high Cr
625 contents in tremolite may be explained by an intervening Cr-rich end-member during water rock
626 interaction, which is most likely the peridotite wall rock.

627 The mobility of Cr in hydrous fluids strongly depends on its oxidation state; whereas Cr³⁺ species
628 are relatively immobile, Cr⁶⁺ are highly mobile (e.g. James, 2003). Thus chromite dissolution and
629 formation of Cr-rich magnetite rims around inherited chromite grains needs oxidizing fluids
630 consistent with the preferential leaching of Eu from felsic dykes and occurrence of positive Eu
631 anomalies in tremolite. Oxidized character is a feature of fluids emitted by subduction zones that
632 involve highly serpentinized upper mantle rocks (Evans et al. 2017). Nothing is known about the
633 degree of primary serpentinization of the lower plate of the subduction/obduction system of New
634 Caledonia, which has totally disappeared in the subduction zone, whereas its partial preservation in
635 the exhumed eclogitic melange (Pouébo Terrane) is problematical. However, considering that the
636 degree of serpentinization of the upper and lower plates might have been identical prior to
637 subduction inception, it may be postulated that an important part of the serpentinization of the
638 Peridotite Nappe was acquired prior to subduction and is unrelated to obduction.

639

640 **6.6. Origin of the tremolite- and antigorite-precipitating fluids**

641 The $\delta^{18}\text{O}$ and δD of hydrous minerals result from complex balance of several parameters: the
642 isotopic composition of the wall rock, the water/rock ratio, mineral-water fractionation coefficients,
643 isotope ratios of circulating water, and temperature. In this study, a probable temperature of
644 tremolite-antigorite formation can be estimated in the range 300-450°C (end of section 6.2). Under
645 these temperatures, tremolite and serpentine minerals should display comparable $\delta^{18}\text{O}$ values as an
646 equilibrium isotopic fractionation of about $\delta^{18}\text{O}_{\text{Trem-Serp}} = -0.3\text{‰}$ is expected (Zheng, 1993). The mean
647 $\delta^{18}\text{O}$ values depicted above (6.8‰ and 6.4‰ for tremolite and antigorite, respectively,
648 corresponding to $\delta^{18}\text{O}_{\text{Trem-Serp}} = +0.4\text{‰}$) are somewhat at variance with the theoretical equilibrium.
649 In fact, in the crystallizing sequence (end of section 6.2), we argue for a slight temperature decrease
650 at the end of tremolite crystallization. All other things equal, this should imply a small increase of the
651 isotopic fractionation between tremolite and water, and thus an increase in the $\delta^{18}\text{O}$ value of
652 tremolite. The fact that tremolite is actually slightly enriched ($\delta^{18}\text{O} = 6.8\text{‰}$) in ^{18}O relative to
653 antigorite ($\delta^{18}\text{O} = 6.4\text{‰}$) is consistent with a small temperature decrease.

654 It remains that, at first order, both minerals likely crystallized in equilibrium with a unique
655 aqueous reservoir. Assuming then that fluid was in excess at the time of vein formation (i. e., the
656 fluid/rock ratio is isotopically infinite, which was very likely in the case of the studied veins), the $\delta^{18}\text{O}$

657 value of the invading fluid can be estimated between 6.5‰ and 8.5‰ (using the fractionation factors
658 between H₂O and serpentine and antigorite between 300 and 450°C from Zheng, 1993). These
659 values are consistently expected for the fluids involved in supra-subduction contexts, like the one
660 prevailing for the formation of the studied veins. Indeed, these fluids, originally derived from
661 seawater, underwent significant isotopic exchange with crustal and mantle rocks, as shown by the Sr
662 isotope and REE systematics of vein minerals, which results in increasing their initial, low $\delta^{18}\text{O}$ value.
663 A magmatic contribution to the fluid budget that would add a high- $\delta^{18}\text{O}$ component (about 7‰) to
664 the fluid is permitted by the data.

665 From the observations presented above, it appears that lizardite formed during the primary
666 serpentinization did not significantly recrystallize when the rock went into the antigorite stability
667 field. It should be kept in mind that lizardite and antigorite may coexist at low pressure depending
668 upon serpentine composition (e.g., Caruso and Chernosky 1979; O’Hanley et al. 1989; Schwartz et al.
669 2013). However, antigorite fibres in the study are nucleated on lizardite “black walls” and partly
670 originated from the destabilization of lizardite as shown by the accumulation of iron oxide at the
671 lizardite-antigorite interface (Fig. 4c & 4d). However, the pervasive replacement of “primary”
672 lizardite by antigorite would require an unlikely large water/rock ratio given the relative
673 impermeability of lizardite coatings and restriction of fluid circulation along open fractures.

674 It is worth noting that some serpentine samples from open pit mines did not escape tropical
675 weathering and yield strongly negative δD ratios similar to that of supergene silicates. In order to
676 evaluate the effect of weathering, four samples of the same steep antigorite vein were taken across
677 the weathering profile of Vulcain mine. Upward decreasing δD values ($-38.7\text{‰} > \delta\text{D} > -107.2\text{‰}$; Table
678 5) unambiguously show that only unweathered antigorites taken in the bedrock retain their original
679 isotopic signature. In addition, rocks of the serpentinite sole display strongly negative δD ($-$
680 $80\text{‰} < \delta\text{D} < -105\text{‰}$) (Fig. 12; Ulrich 2012); thus, these otherwise unweathered rocks have been
681 profoundly modified by fluids of supergene origin as also shown by their negative Ce anomaly.

682

683 7. Conclusion

684 The formation of tremolite- and antigorite-bearing veins recorded an elevation of temperature
685 compared to the initial oceanic (primary) serpentinization of peridotites, which was marked by the
686 formation of lizardite and minor chrysotile; therefore, a heat source is needed as well as a source for
687 Ca. This event was associated with shear/tensile reactivation of pre-existing joints already coated by
688 lizardite.

689 The widespread antigorite-bearing low-Ca veins bear highly radiogenic $^{87}\text{Sr}/^{86}\text{Sr}$ signatures, which
690 record some influence of subducted sediments. In contrast, this “sedimentary” signature is diluted in
691 high-Ca tremolite, which probably comes from Ca input of direct or indirect magmatic origin. The Sr
692 isotope signature of tremolite, the REE abundance, the $\delta^{18}\text{O}$ and δD values, all suggest that the Ca

693 source was the supra-subduction Early Eocene felsic dyke system and thus indirectly, the subducted
694 slab. Europium positive anomaly and paradoxically high contents of some compatible elements (Cr)
695 suggests that Ca was carried by oxidized hydrothermal fluids that leached the dykes and interacted
696 with Cr-rich wall rock (serpentinized peridotite). These features provide a constraint on the timing of
697 tremolite crystallization, because supra-subduction dykes intruded peridotites within a narrow span
698 of time at ca. 55-50 Ma (U-Pb zircon; Cluzel et al. 2006). Since these dykes are mostly slab melts and
699 boninite-series magmas that formed shortly after subduction inception (Cluzel et al. 2012, 2016),
700 already existing fractures were reactivated in an oblique-extensional regime under relatively high
701 fluid pressure and gave way to magma and laterally/upward to Ca-bearing fluids when the young and
702 buoyant slab had to force its way beneath the nascent fore-arc lithosphere (Fig. 14). The heat source
703 needed for crystallizing tremolite and antigorite after lizardite was likely provided by the injection of
704 supra-subduction slab melts. The coexistence of tremolite and antigorite in the same fracture fillings,
705 and thereafter crystallization of antigorite solely, could signal the exhaustion of magmatic fluids,
706 while synkinematic crystallization associated with small-scale fault motion and tension cracks was
707 continuing. It also signals that tremolite and antigorite veins were related to the same tectonic event,
708 whereas only fluid composition differed through time.

709 Therefore, independent of decreasing-upward serpentinization due to the obduction of the
710 Peridotite Nappe, the development of tremolite-antigorite veins may be related to a three-step
711 evolution: i) the “oceanic” period (Late Cretaceous-Paleocene), during which static serpentinization
712 developed along cooling joints (Fig. 14a); ii) at subduction inception (latest Paleocene), when the
713 injection of slab-derived supra-subduction magmas and fluids, together with semi-brittle shearing
714 due to tectonic deformation of the upper plate of the system generated the tremolite-antigorite vein
715 system (Fig. 14b); finally, iii) the fluid-assisted melting of the supra-subduction mantle generated
716 strongly depleted melts, the fractionation of which resulted in gabbronorite cumulates (Fig. 14c) that
717 now represent the only remnants of fore-arc crust.

718

719 **Acknowledgements**

720 This study is a contribution of CNRT Nickel et son Environnement research programs « Amiante-
721 serpentinisation et fibrogenèse dans les massifs de péridotite de Nouvelle-Calédonie » and
722 « Ophiostruct-Connaissance de l’ophiolite, structure et géophysique aéroportée ». T. Rigaudier
723 (CRPG, Vandœuvre-lès-Nancy, France) is warmly thanked for assistance in isotope analyses. We
724 acknowledge Professor Katy Evans and an anonymous reviewer for their constructive reviews.

725

726

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1085

1086 **Figure captions**

1087

1088 Figure 1: Geological sketch map of New Caledonia.

1089

1090 Figure 2: a: Unserpentinized peridotite with olivine grains crosscut by cooling (?) micro-cracks
1091 (Kopeto mine). b: incipiently serpentinized peridotite with development of lizardite along
1092 parallel joints (Me Maoya Massif). c: outcrop picture and conceptual model (d) of centripetal
1093 development of 'primary' serpentinization (lizardite) in blocks of harzburgite delineated by a
1094 pre-existing joint network (roadside near Poro mine, east coast). Ol: olivine; Chr: chromite; Lz:
1095 lizardite

1096

1097 Figure 3: Tremolite-antigorite veins. a: mixed (successive ?) development of palisadic and radiating
1098 structures in a composite tremolite vein (Poro mine); b: antigorite crack seal developed against
1099 older lizardite-coated fracture walls; c: tension crack filled with tremolite crack seal; 3d: thin
1100 section of small-scale shear joint (micro-fault) showing oblique synkinematic tremolite fibres,
1101 note the chronological relationship of light brown lizardite veins (1) and tremolite crack seal
1102 (2). Tr: tremolite; Lz: lizardite; Atg: antigorite; Hzb: harzburgite

1103

1104 Figure 4: Outcrop picture (a) and conceptual model (b) of relationships between pressure solution,
1105 shear solution, and synkinematic crystal growth in tension and shear (pull apart) cracks. c:
1106 shear fracture (micro-fault) filled with syn-kinematic antigorite fibres (Atg) nucleated on pre-
1107 existing lizardite (Lz), magnetite grains (Mag) were formed by Fe liberation during syn-tectonic
1108 decomposition of lizardite; d: same features as c to show prominent magnetite formation
1109 (dark seams and streaks) at the boundary of newly formed antigorite (Atg) and pre-existing
1110 lizardite.

1111

1112 Figure 5: Microphotographs of synchronously crystallizing hydrous silicates in tension cracks of partly
1113 serpentinized peridotite. a-b: intergrowths of tremolite, antigorite and Mg-chlorite in a tension
1114 crack (a: planar light; b: crossed nicols). c-d: border of a tension crack showing syn-crystallizing
1115 tremolite and antigorite nucleated against lizardite of the wall rock (c: planar light; d: crossed
1116 nicols). Tr: tremolite; Chl: chlorite; Atg: antigorite; Lz: lizardite; Ol: olivine.

1117

1118 Figure 6: SEM image of intimately intergrown tremolite (Tr) and antigorite (Atg) fibres (sample#
1119 NC581DLA)

1120

1121 Figure 7: Raman spectra of tremolite and antigorite from sample# NC581DLA (see Fig. 6). Intensity is
1122 given in arbitrary units (a.u.).

1123

1124 Figure 8: Comparison of chondrite-normalized REE patterns (Evensen et al. 1978) to show the hook
1125 shape and relatively high REE contents of tremolite veins compared to New Caledonia
1126 peridotites (Secchiari 2016), early Eocene dykes (Cluzel et al. 2006) and serpentinites of the
1127 tectonic sole (with negative Ce anomaly; Ulrich, 2010)

1128

1129 Figure 9: Chemical classification (Leake, 1968; Leake et al. 1997) of microprobe analyses of single
1130 minerals from tremolite-bearing veins and pockets; the data form a continuous array spanning
1131 from high-Si tremolite to tremolite hornblende.

1132

1133 Figure 10: Hydrothermal incorporation of Cr in tremolite. a: Cr vs. Si plot of tremolite showing the
1134 increasing Cr molecular content with decreasing total Si content at increasing temperature
1135 (from tremolite to tremolite-hornblende. b: Cr₂O₃ (wt%) and X_{FeA} covariation similarly showing
1136 the positive correlation of Cr content and Fe ratio of tremolite.

1137

1138 Figure 11: a: Plot of Sr vs. ⁸⁷Sr/⁸⁶Sr to compare Sr isotopic composition of tremolite and antigorite
1139 (this study) to unserpentinized peridotites (Secchiari et al. 2018, 2019), gabbro-norites
1140 (Secchiari et al. 2016), Early Eocene dykes (Cluzel et al. 2006) from the Peridotite Nappe, and
1141 Atlantis Massif serpentinites (Boschi et al. 2008). Ocean waters compositions are from Murthy
1142 and Beiser 1968, Elderfield (1986), Veizer (1989) and McArthur et al. (2012); b: plot of Ca vs.
1143 ⁸⁷Sr/⁸⁶Sr, same symbols as in 11a.

1144

1145 Figure 12: δ¹⁸O and δD isotopic ratios of fibrous serpentines and tremolites of Poro site. "Oceanic"
1146 serpentinites after Agrinier et al. (1995, 1997); supergene silicates field ("garnierite") after
1147 Amisse et al. (2010) and deweylite after Wenner and Taylor (1974), serpentinite sole of the
1148 Peridotite Nappe after Ulrich (2010).

1149

1150 Figure 13: a: SEM image of altered chromite grain from peridotite isolated within a tremolite-
1151 antigorite replacive "vein". The grain is formed of a residual core of primary chromite and a
1152 cortex of micro-granular chromian magnetite and low-Cr chromite. b & c sketch drawing to
1153 show the metasomatic replacement of peridotite wall rock by a mixture of tremolite and
1154 antigorite with partly altered chromite

1155

1156 Figure 14: Geodynamic model of subduction inception showing the timing of tremolite-antigorite
1157 veins development.

1158

1159 Table 1: Raman shift data from Raman spectra of two samples of tremolite-antigorite veins from
1160 Poro mine (data from Lahondere et al. 2012) uncertainty is $\pm 2\text{cm}^{-1}$

1161 Table 2: Whole rock major and trace-elements composition of representative tremolite-antigorite
1162 veins of Poro Ni mine

1163 Table 3: Microprobe analyses of tremolites

1164 Table 4: Strontium isotopic data for tremolite-antigorite and antigorite veins from Poro Ni mine (east
1165 coast New Caledonia). $^{87}\text{Sr}/^{86}\text{Sr}$ normalization value = 0.710245

1166 Table 5: Oxygen and hydrogen isotopic compositions of fibrous veins from Poro nickel mine (east
1167 coast New Caledonia)

1168

1169 Table 1: Raman shift data from Raman spectra of two samples of tremolite-antigorite veins from
 1170 Poro mine (data from Lahondere et al. 2012) uncertainty is $\pm 2\text{cm}^{-1}$
 1171
 1172

| NC225DL(2)-B | NC226DL | NC225DL(2)-B | NC226DL |
|---------------------|----------------|---------------------|----------------|
| Tremolite | Tremolite | Antigorite | Antigorite |
| 124 | - | - | - |
| 164 | 166 | 233 | 233 |
| 181 | 184 | - | - |
| 226 | 227 | 381 | 378 |
| 372 | 375 | - | - |
| 398 | 395 | - | - |
| 419 | - | - | - |
| 532 | - | 687 | 691 |
| 656 | - | 1047 | 1046 |
| 676 | 676 | - | - |
| 751 | - | 3666 | 3666 |
| 933 | - | 3698 | 3699 |
| 1031 | 1032 | | |
| 1066 | 1066 | | |
| 3663 | 3662 | | |
| 3676 | 3676 | | |

1175

1176

| sample | POR1a tremol | POR1b tremol | POR2a antigor? | POR2c tremol | POR5 tremol | POR6 tremol | POR7b tremol | POR 8c antigor? | POR10 antigor | POR 13 antigor? |
|-----------|-----------------|-----------------|-------------------|-----------------|----------------|----------------|-----------------|--------------------|------------------|--------------------|
| Pr | 0.33 | 0.394 | < D.L. | 0.203 | 0.198 | 0.251 | 0.198 | 0.025 | 0.012 | 0.224 |
| Rb | < D.L. | < D.L. | < D.L. | < D.L. | < D.L. | < D.L. | < D.L. | < D.L. | < D.L. | < D.L. |
| Sc | 13 | < D.L. | < D.L. | 25.39 | 13.29 | 10.86 | 11.87 | < D.L. | < D.L. | < D.L. |
| Sb | < D.L. | < D.L. | < D.L. | < D.L. | < D.L. | < D.L. | < D.L. | < D.L. | < D.L. | < D.L. |
| Sm | 0.321 | 0.365 | < D.L. | 0.196 | 0.135 | 0.186 | 0.149 | 0.026 | < D.L. | 0.211 |
| Sn | < D.L. | < D.L. | < D.L. | < D.L. | < D.L. | < D.L. | < D.L. | < D.L. | < D.L. | < D.L. |
| Sr | 6.963 | 7.57 | < D.L. | 3.947 | < D.L. | 14.19 | 7.77 | < D.L. | < D.L. | 3.968 |
| Ta | < D.L. | < D.L. | 0.018 | < D.L. | < D.L. | < D.L. | < D.L. | < D.L. | < D.L. | < D.L. |
| Tb | 0.047 | 0.055 | < D.L. | 0.027 | 0.02 | 0.028 | 0.021 | < D.L. | < D.L. | 0.031 |
| Th | < D.L. | < D.L. | < D.L. | < D.L. | < D.L. | < D.L. | < D.L. | < D.L. | < D.L. | < D.L. |
| Tm | 0.028 | 0.03 | < D.L. | 0.014 | 0.011 | 0.024 | 0.02 | 0.002 | 0.001 | 0.02 |
| U | < D.L. | < D.L. | < D.L. | < D.L. | < D.L. | < D.L. | < D.L. | < D.L. | < D.L. | < D.L. |
| V | 103.3 | 110.6 | 28.97 | 72.11 | 42.59 | 190.9 | 173.1 | 14.23 | 13.57 | 73.97 |
| W | < D.L. | < D.L. | < D.L. | < D.L. | < D.L. | < D.L. | < D.L. | < D.L. | < D.L. | < D.L. |
| Y | 1.734 | 1.885 | < D.L. | 0.833 | 0.647 | 1.108 | 0.951 | < D.L. | < D.L. | 1.154 |
| Yb | 0.194 | 0.22 | < D.L. | 0.091 | 0.08 | 0.178 | 0.148 | 0.008 | 0.008 | 0.139 |
| Zn | 97.97 | 52.38 | < D.L. | 54.83 | 22.71 | < D.L. | 23.35 | 17.43 | 16.71 | < D.L. |
| Zr | 1.533 | 1.765 | < D.L. | < D.L. | < D.L. | < D.L. | < D.L. | < D.L. | < D.L. | 2.458 |

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1181 Table 3: microprobe analyses of tremolites

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| Spot# | Na2O | MgO | SiO2 | Al2O3 | K2O | CaO | TiO2 | Cr2O3 | FeO | MnO | NiO | Total |
|-------|-------|--------|--------|-------|-------|--------|-------|-------|-------|-------|-------|--------|
| 87 | 0.504 | 23.918 | 56.915 | 1.412 | 0.000 | 12.453 | 0.000 | 0.38 | 1.593 | 0.086 | 0.013 | 97.275 |
| 88 | 1.042 | 22.125 | 54.209 | 3.489 | 0.017 | 11.725 | 0.045 | 0.901 | 2.142 | 0.000 | 0.171 | 95.865 |
| 89 | 0.801 | 23.956 | 56.247 | 2.415 | 0.049 | 12.307 | 0.000 | 0.732 | 1.737 | 0.003 | 0.082 | 98.329 |
| 1 | 0.19 | 24.867 | 58.846 | 0.378 | 0.018 | 13.291 | 0.000 | 0.061 | 1.108 | 0.073 | 0.145 | 98.977 |
| 12 | 0.009 | 24.539 | 58.69 | 0.08 | 0.002 | 13.463 | 0.029 | 0.086 | 1.025 | 0.006 | 0.153 | 98.081 |
| 21 | 0.904 | 23.675 | 55.909 | 2.864 | 0.051 | 12.282 | 0.082 | 0.195 | 1.914 | 0.15 | 0.082 | 98.107 |
| 22 | 0.804 | 23.614 | 54.598 | 2.766 | 0.006 | 12.381 | 0.000 | 0.714 | 1.879 | 0.024 | 0.283 | 97.068 |
| 23 | 0.184 | 24.552 | 58.604 | 0.473 | 0.006 | 12.956 | 0.025 | 0.003 | 1.26 | 0.114 | 0.236 | 98.412 |
| 24 | 0.255 | 24.571 | 57.662 | 0.598 | 0.002 | 12.913 | 0.000 | 0.048 | 1.406 | 0.000 | 0.179 | 97.634 |
| 25 | 0.722 | 23.759 | 56.39 | 2.132 | 0.000 | 12.798 | 0.000 | 0.627 | 1.647 | 0.129 | 0.095 | 98.299 |
| 27 | 0.231 | 24.425 | 58.317 | 0.605 | 0.072 | 13.249 | 0.031 | 0.126 | 1.195 | 0.000 | 0.124 | 98.376 |
| 28 | 0.768 | 24.121 | 56.579 | 2.116 | 0.002 | 12.369 | 0.000 | 0.168 | 1.865 | 0.084 | 0.211 | 98.284 |
| 34 | 1.063 | 24.586 | 50.703 | 2.29 | 0.05 | 11.555 | 0.000 | 0.61 | 1.721 | 0.126 | 0.262 | 92.967 |
| 36 | 0.298 | 25.178 | 57.362 | 0.533 | 0.000 | 11.859 | 0.046 | 0.237 | 2.183 | 0.157 | 0.159 | 98.011 |
| 37 | 0.731 | 23.781 | 56.45 | 2.202 | 0.000 | 12.512 | 0.000 | 0.522 | 1.83 | 0.033 | 0.047 | 98.108 |
| 38 | 0.71 | 23.563 | 56.467 | 2.263 | 0.05 | 12.477 | 0.036 | 0.522 | 1.682 | 0.000 | 0.024 | 97.794 |
| 39 | 1.254 | 23.265 | 54.974 | 4.231 | 0.041 | 11.967 | 0.091 | 0.735 | 2.583 | 0.134 | 0.064 | 99.34 |
| 40 | 0.565 | 23.974 | 56.975 | 1.938 | 0.023 | 12.617 | 0.017 | 0.402 | 1.708 | 0.001 | 0.000 | 98.218 |
| 42 | 1.036 | 23.633 | 53.843 | 3.516 | 0.015 | 12.027 | 0.000 | 1.233 | 2.307 | 0.000 | 0.000 | 97.612 |
| 48 | 1.04 | 23.518 | 54.855 | 3.351 | 0.011 | 12.173 | 0.012 | 0.768 | 2.039 | 0.115 | 0.074 | 97.957 |
| 49 | 1.141 | 23.469 | 54.485 | 3.629 | 0.012 | 12.311 | 0.078 | 0.796 | 2.123 | 0.138 | 0.112 | 98.294 |
| 50 | 0.665 | 23.907 | 56.915 | 1.97 | 0.043 | 12.684 | 0.000 | 0.352 | 1.529 | 0.184 | 0.129 | 98.378 |
| 53 | 0.914 | 23.647 | 55.907 | 2.844 | 0.053 | 12.298 | 0.032 | 0.539 | 1.963 | 0.018 | 0.136 | 98.352 |
| 54 | 1.163 | 22.896 | 53.848 | 3.769 | 0.014 | 12.004 | 0.000 | 0.825 | 2.163 | 0.135 | 0.301 | 97.119 |
| 55 | 1.17 | 23.237 | 54.081 | 3.487 | 0.021 | 12.375 | 0.000 | 0.629 | 2.16 | 0.108 | 0.021 | 97.289 |
| 56 | 0.962 | 23.511 | 55.354 | 3.152 | 0.002 | 12.383 | 0.000 | 0.606 | 2.183 | 0.102 | 0.25 | 98.505 |
| 59 | 1.047 | 23.452 | 54.974 | 3.265 | 0.006 | 12.539 | 0.000 | 0.544 | 1.897 | 0.116 | 0.159 | 97.999 |
| 80 | 0.735 | 23.803 | 55.714 | 2.296 | 0.000 | 12.616 | 0.034 | 0.648 | 1.648 | 0.121 | 0.19 | 97.805 |
| 82 | 0.68 | 23.865 | 56.148 | 2.267 | 0.002 | 12.407 | 0.000 | 0.67 | 1.782 | 0.008 | 0.182 | 98.012 |
| 83 | 0.522 | 29.048 | 51.846 | 1.628 | 0.015 | 8.143 | 0.000 | 0.491 | 2.593 | 0.08 | 0.226 | 94.593 |
| 85 | 0.969 | 23.976 | 55.769 | 2.777 | 0.015 | 12.058 | 0.008 | 0.77 | 2.019 | 0.037 | 0.216 | 98.614 |
| 96 | 0.864 | 23.749 | 55.321 | 2.717 | 0.035 | 12.513 | 0.048 | 0.833 | 1.704 | 0.111 | 0.000 | 97.895 |
| 97 | 0.647 | 25.043 | 57.255 | 1.912 | 0.000 | 10.987 | 0.000 | 0.507 | 2.293 | 0.09 | 0.085 | 98.818 |
| 98 | 0.746 | 24.14 | 56.104 | 2.259 | 0.055 | 12.465 | 0.007 | 0.656 | 1.68 | 0.061 | 0.213 | 98.388 |
| 99 | 0.573 | 24.48 | 57.347 | 1.641 | 0.026 | 12.188 | 0.045 | 0.57 | 1.656 | 0.094 | 0.004 | 98.623 |
| 107 | 0.192 | 25.294 | 57.056 | 0.575 | 0.027 | 12.034 | 0.058 | 0.201 | 2.166 | 0.165 | 0.15 | 97.918 |
| 111 | 0.7 | 24.236 | 56.928 | 2.031 | 0.017 | 12.5 | 0.000 | 0.605 | 1.583 | 0.000 | 0.216 | 98.816 |
| 112 | 1.034 | 23.895 | 55.36 | 3.147 | 0.025 | 11.606 | 0.074 | 0.793 | 1.72 | 0.022 | 0.026 | 97.701 |
| 114 | 0.693 | 24.658 | 56.873 | 1.904 | 0.023 | 11.447 | 0.000 | 0.453 | 2.245 | 0.181 | 0.111 | 98.587 |
| 131 | 1.041 | 23.147 | 54.364 | 3.395 | 0.019 | 12.352 | 0.000 | 0.895 | 1.887 | 0.136 | 0.032 | 97.267 |
| 132 | 0.539 | 24.785 | 55.089 | 2.585 | 0.021 | 11.722 | 0.000 | 0.599 | 1.736 | 0.013 | 0.123 | 97.212 |
| 133 | 0.245 | 28.133 | 55.568 | 0.359 | 0.008 | 9.6 | 0.000 | 0.092 | 1.516 | 0.159 | 0.000 | 95.679 |

| | | | | | | | | | | | | |
|-----|-------|--------|--------|-------|-------|--------|-------|-------|-------|-------|-------|--------|
| 134 | 0.17 | 30.601 | 51.269 | 2.507 | 0.009 | 7.19 | 0.000 | 0.307 | 1.718 | 0.111 | 0.102 | 93.984 |
| 135 | 0.174 | 27.053 | 56.341 | 0.243 | 0.04 | 10.506 | 0.03 | 0.095 | 1.533 | 0.036 | 0.013 | 96.066 |
| 149 | 0.22 | 25.962 | 56.11 | 0.594 | 0.000 | 11.748 | 0.009 | 0.189 | 1.724 | 0.079 | 0.084 | 96.72 |
| 150 | 1.457 | 22.342 | 52.332 | 5.442 | 0.032 | 12.595 | 0.027 | 1.189 | 2.16 | 0.144 | 0.024 | 97.743 |
| 175 | 0.006 | 24.879 | 58.634 | 0.049 | 0.000 | 13.236 | 0.000 | 0.013 | 1.361 | 0.015 | 0.203 | 98.396 |
| 184 | 0.065 | 24.378 | 58.639 | 0.167 | 0.014 | 12.87 | 0.000 | 0.16 | 1.64 | 0.073 | 0.04 | 98.047 |
| 185 | 0.573 | 24.648 | 56.837 | 1.682 | 0.022 | 12.041 | 0.000 | 0.481 | 1.861 | 0.135 | 0.381 | 98.662 |
| 186 | 0.557 | 24.127 | 56.995 | 1.924 | 0.017 | 12.753 | 0.000 | 0.51 | 1.774 | 0.045 | 0.024 | 98.724 |
| 187 | 0.712 | 24.163 | 56.97 | 2.167 | 0.05 | 12.293 | 0.000 | 0.559 | 1.988 | 0.041 | 0.23 | 99.174 |
| 188 | 0.44 | 25.599 | 56.112 | 1.463 | 0.000 | 10.51 | 0.000 | 0.396 | 1.747 | 0.082 | 0.738 | 97.086 |
| 189 | 0.715 | 24.465 | 56.55 | 2.379 | 0.000 | 12.155 | 0.000 | 0.661 | 1.774 | 0.000 | 0.083 | 98.782 |
| 190 | 0.655 | 24.285 | 56.226 | 2.24 | 0.047 | 11.875 | 0.000 | 0.659 | 2.028 | 0.174 | 0.516 | 98.705 |
| 191 | 0.64 | 27.375 | 54.089 | 2.1 | 0.003 | 9.989 | 0.000 | 0.604 | 2.485 | 0.019 | 0.291 | 97.596 |
| 192 | 0.796 | 23.771 | 55.874 | 2.474 | 0.059 | 12.275 | 0.000 | 0.654 | 1.917 | 0.000 | 0.164 | 97.984 |
| 194 | 0.575 | 25.642 | 57.97 | 1.873 | 0.04 | 9.999 | 0.047 | 0.568 | 2.58 | 0.183 | 0.106 | 99.583 |

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1184 Table 4 : Sr isotopic compositions of tremolite-antigorite veins

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| sample | Field name | CaO wt% | Sr ppm | ⁸⁷ Sr/ ⁸⁶ Sr |
|--------|--------------------|---------|--------|------------------------------------|
| POR 1a | Tremolite mix | 7.19 | 6.963 | 0.704262 |
| POR 1b | metasomatized dyke | 12.56 | 7.57 | 0.704417 |
| POR 2a | antigorite ? | 0.02 | < DL | 0.707058 |
| POR 2c | Tremolite mix | 5.07 | 3.947 | 0.704402 |
| POR 5 | Tremolite mix | 1.82 | < DL | 0.705369 |
| POR 6 | tremolite | 11.52 | 14.19 | 0.703894 |
| POR 7b | tremolite | 11.16 | 7.77 | 0.704239 |
| POR 8c | antigorite ? | 0.01 | < DL | 0.706264 |
| POR 10 | antigorite | 0.01 | < DL | 0.708290 |
| POR 13 | antigorite ? | 0.02 | 3.968 | 0.702853 |
| Sr341 | antigorite | 0.002 | 0.058 | 0.709741 |
| Sr343 | tremolite | 9.6 | 8.885 | 0.704207 |
| Sr344 | antigorite | 0.002 | 0.086 | 0.709701 |
| Sr346 | antigorite | 0.004 | 1.428 | 0.707945 |
| Sr347 | antigorite | 0.002 | 0.036 | 0.709546 |
| Sr349 | antigorite | 0.002 | 0.351 | 0.709687 |
| Sr350 | antigorite | 0.002 | 0.070 | 0.710002 |
| Sr353 | antigorite | 0.002 | 0.173 | 0.709261 |
| Sr354 | tremolite | 4.675 | 5.250 | 0.704542 |
| Sr355 | antigorite ? | 0.113 | 0.617 | 0.708086 |
| Sr356 | tremolite | 9.422 | 9.239 | 0.704205 |
| Sr357 | antigorite | 0.002 | 0.183 | 0.711584 |
| Sr359 | tremolite | 5.248 | 4.318 | 0.704350 |
| Sr360 | tremolite | 3.386 | 3.979 | 0.704529 |

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1188 Table 5: Oxygen and hydrogen isotopic compositions

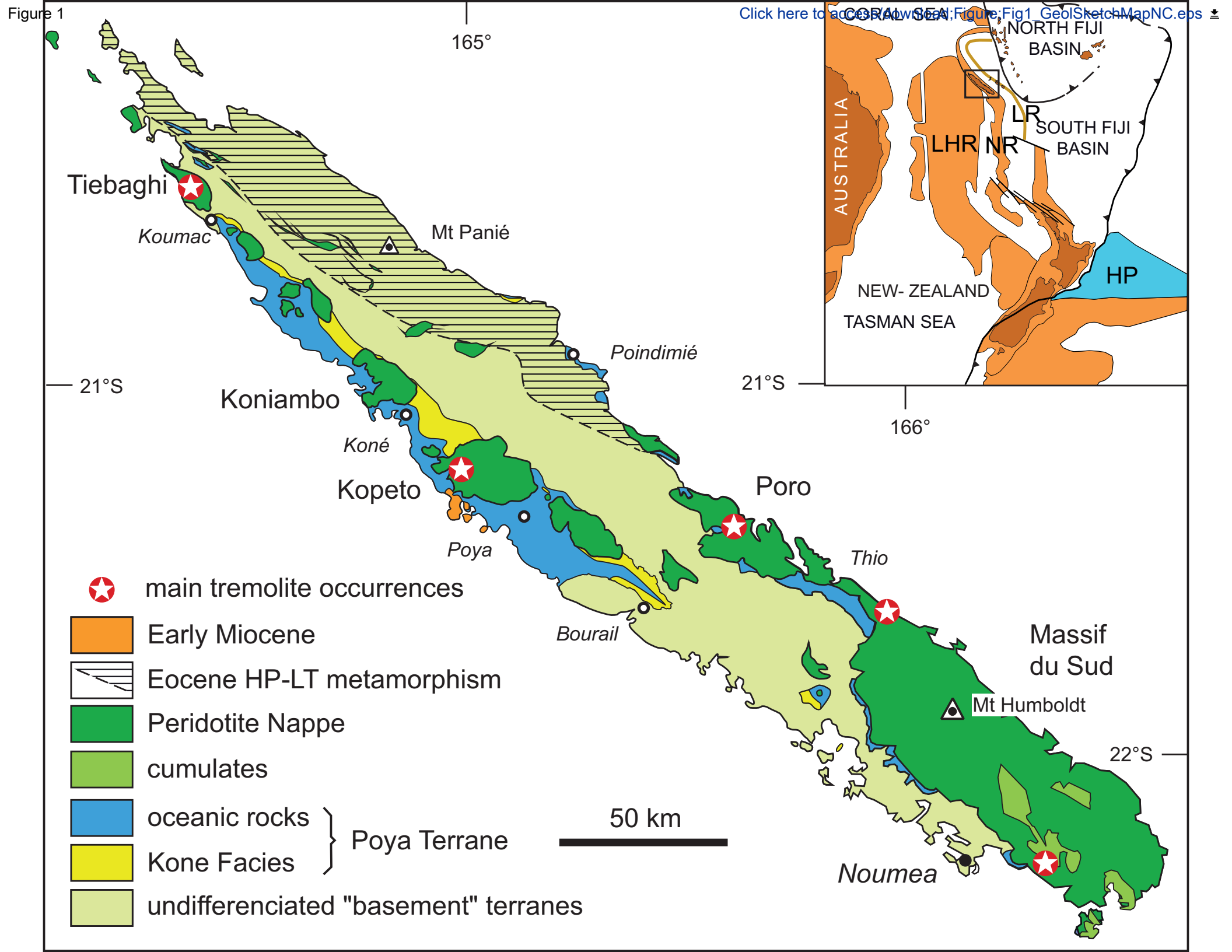
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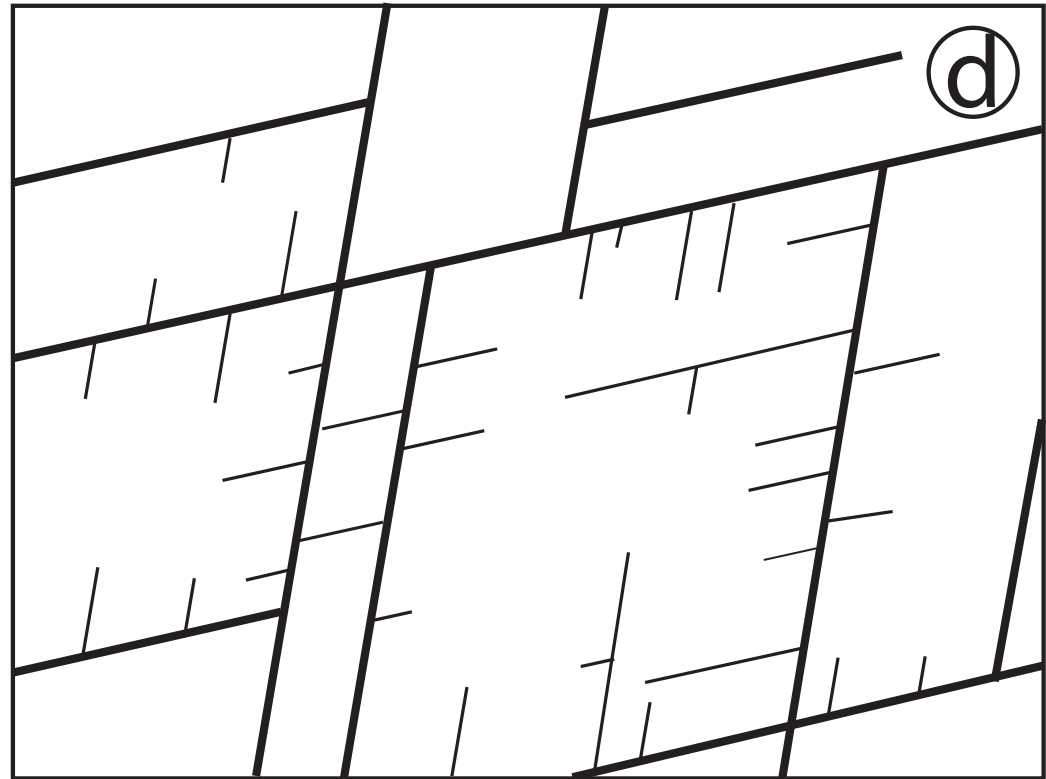
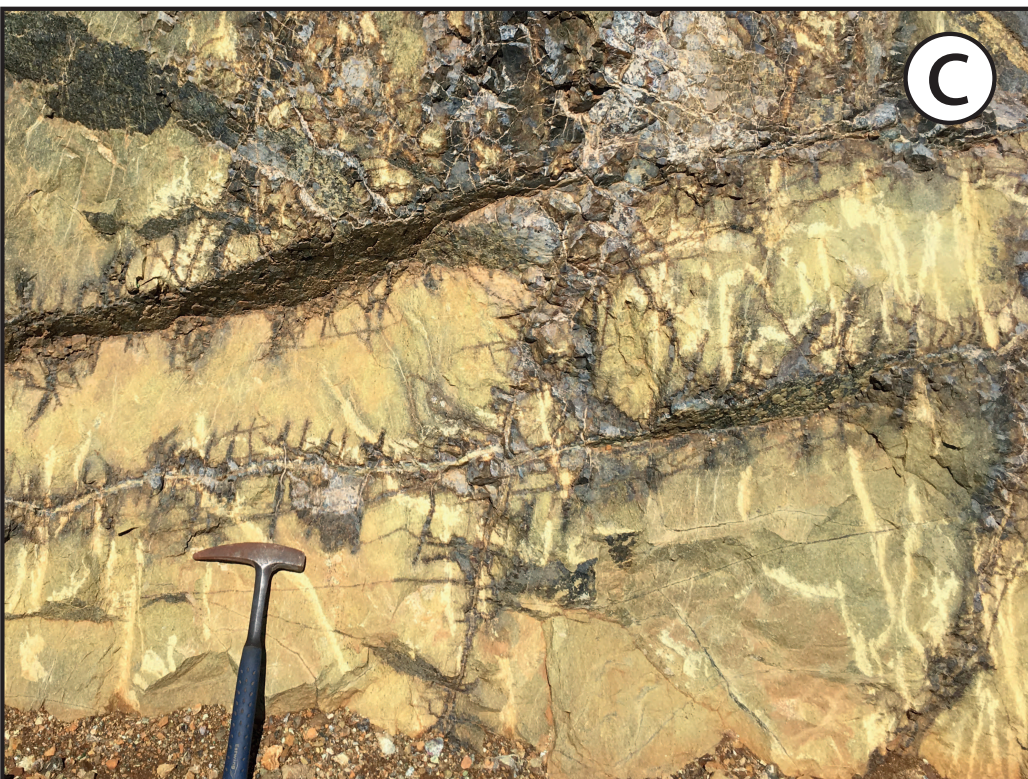
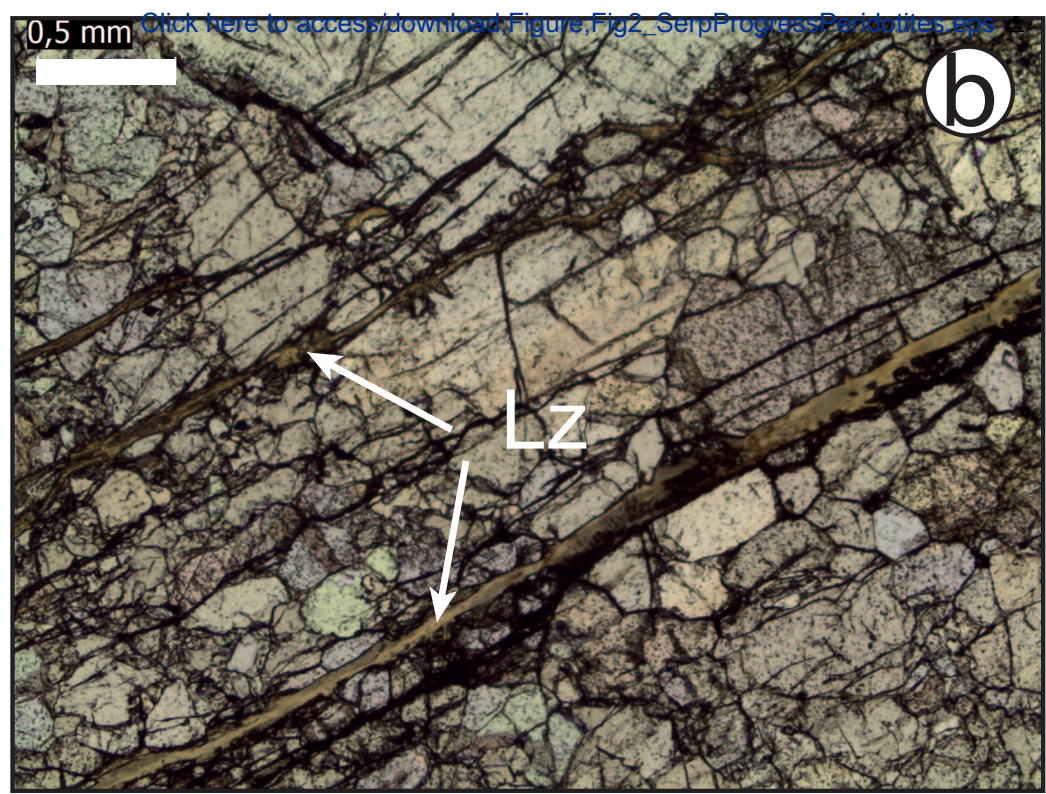
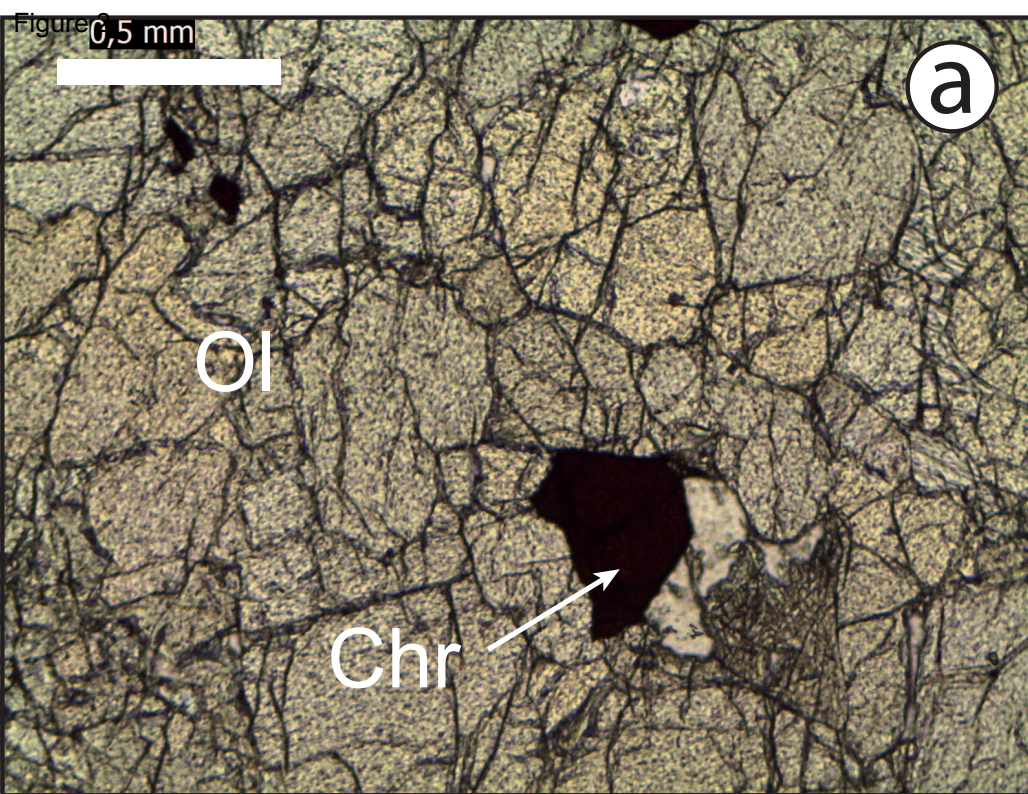
| sample. | location | Type | $\delta^{18}\text{O}$ | | δD | |
|---------|----------|--|-----------------------|------|------------------|-----|
| | | | (‰ SMOW) | ± | (‰ SMOW) | ± |
| POR1a | Poro | tremolite | 7.2 | 0.25 | -58.3 | 1.2 |
| POR1b | Poro | tremolite | 7.2 | 0.25 | -46.8 | 1.8 |
| POR2a | Poro | tremolite | 7.1 | 0.25 | -82.8 | 2.2 |
| POR2b | Poro | tremolite | 6.2 | 0.25 | -47.2 | 1.1 |
| POR2c | Poro | tremolite | 6.3 | 0.25 | -45.3 | 1.4 |
| POR3b | Poro | antigorite | 6.3 | 0.25 | -47.3 | 2.1 |
| POR4 | Poro | Mg-chlorite (penninite) | 3.3 | 0.25 | -17.5 | 2.9 |
| POR5 | Poro | tremolite | 8.8 | 0.25 | -60.6 | 3.9 |
| POR6 | Poro | tremolite (?) | 6.5 | 0.25 | -51.5 | 1.6 |
| POR7b | Poro | tremolite (?) | 5.2 | 0.25 | -46.0 | 2.1 |
| POR8 | Poro | antigorite | 6.3 | 0.25 | -50.6 | 2.0 |
| POR8b | Poro | antigorite | 2.7 | 0.25 | -41.8 | 3.0 |
| POR8c | Poro | antigorite (?) | 6.6 | 0.25 | -52.7 | 2.1 |
| POR10 | Poro | antigorite | 6.3 | 0.25 | -40.4 | 1.0 |
| POR12 | Poro | antigorite | 6.7 | 0.25 | -45.7 | 1.1 |
| | | slickensided porcelain-like | | | | |
| POR14 | Poro | serpentine | 8.8 | 0.25 | -105.8 | 0.9 |
| | | porcelain-like serpentine in tensional | | | | |
| POR15 | Poro | crack seal | 8.3 | 0.25 | -107.7 | 1.1 |
| VUL 4-1 | Vulcain | weathered antigorite | 4.5 | 0.25 | -61.2 | 2.7 |
| VUL 4-2 | Vulcain | weathered antigorite | 6.9 | 0.25 | -107.2 | 0.4 |
| VUL 4-3 | Vulcain | unweathered antigorite | 5.8 | 0.25 | -38.7 | 4.8 |
| VUL 4-4 | Vulcain | slightly weathered antigorite | 6.0 | 0.25 | -42.1 | 2.0 |

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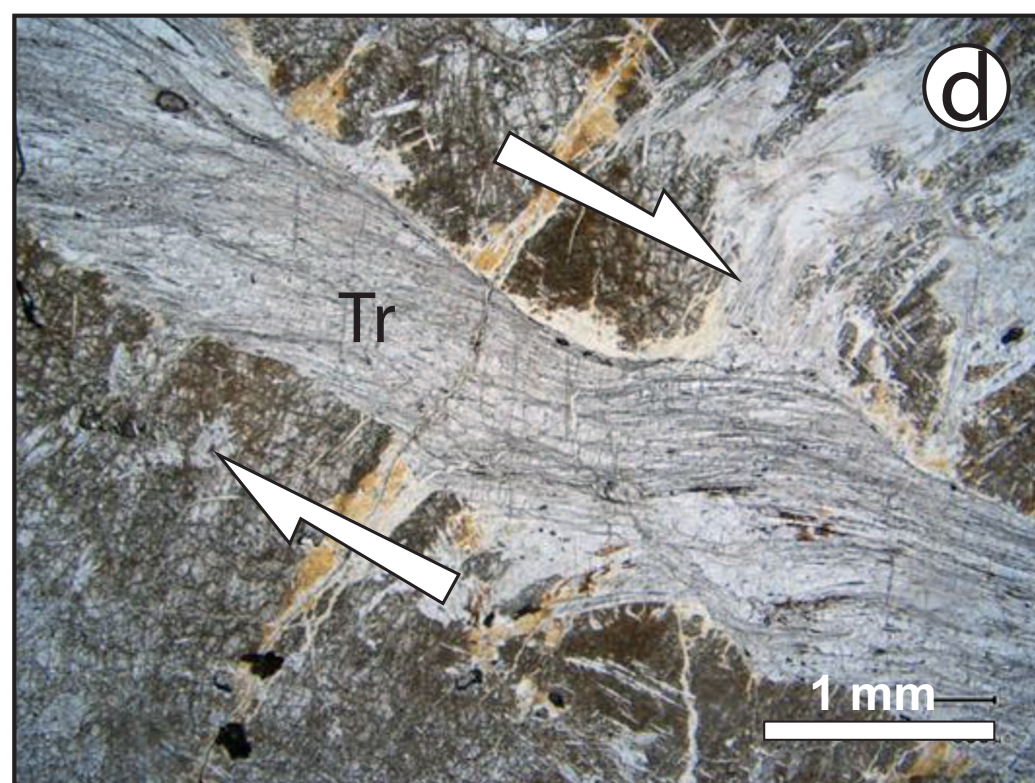
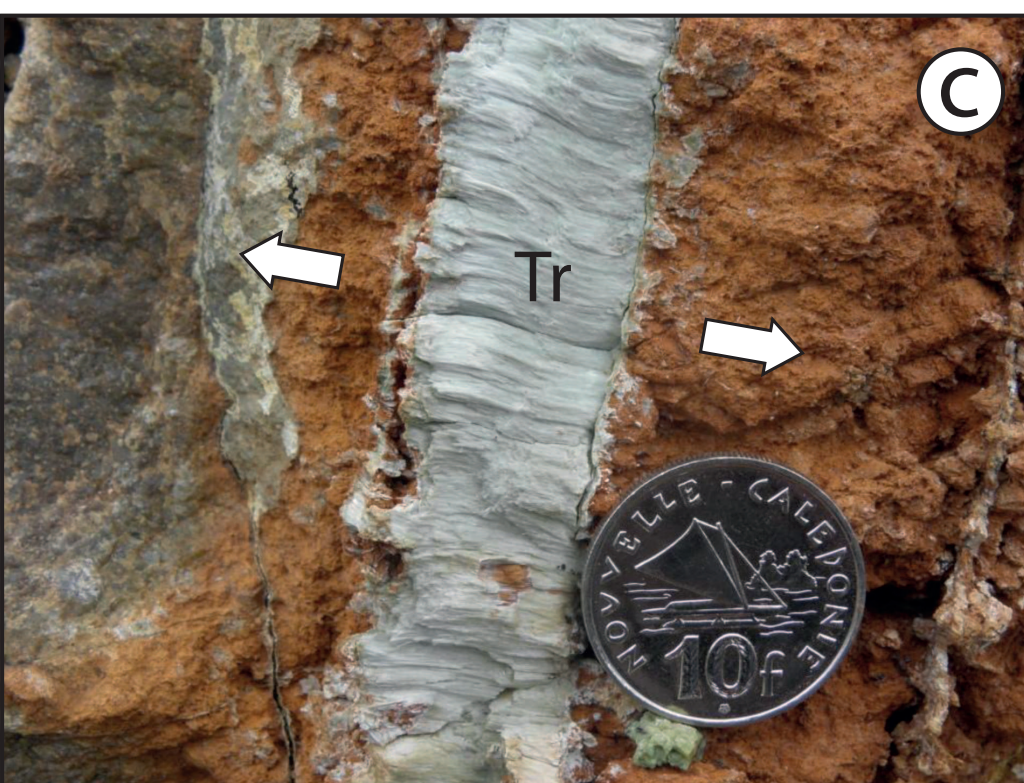
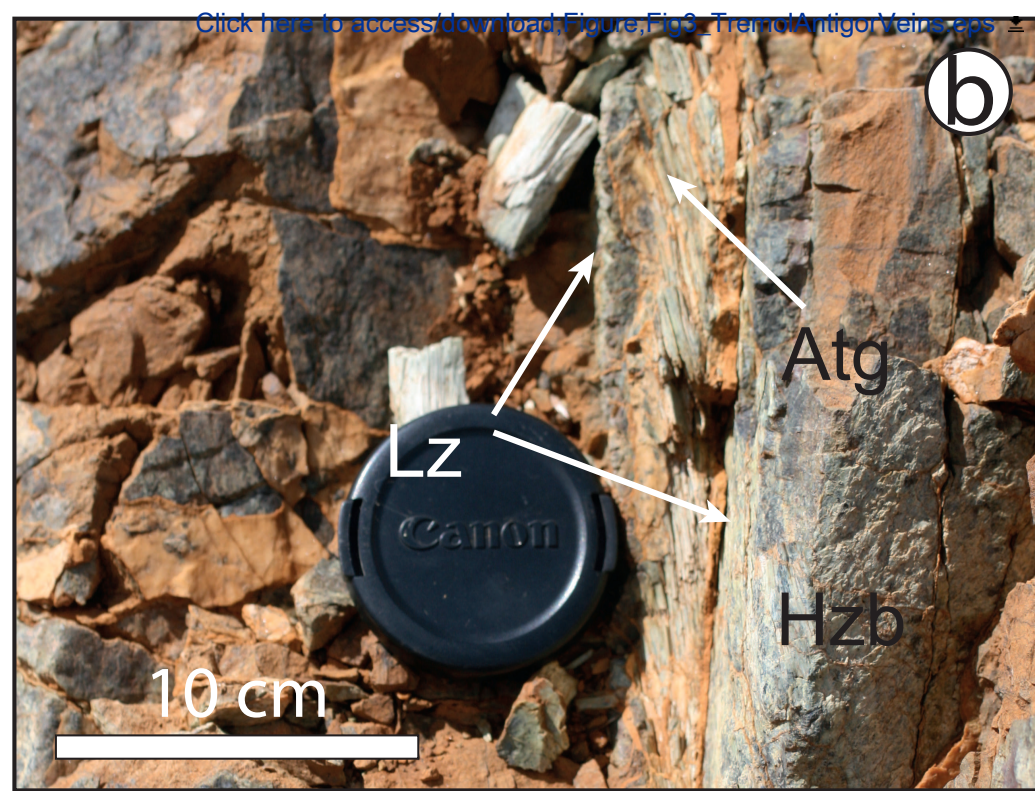
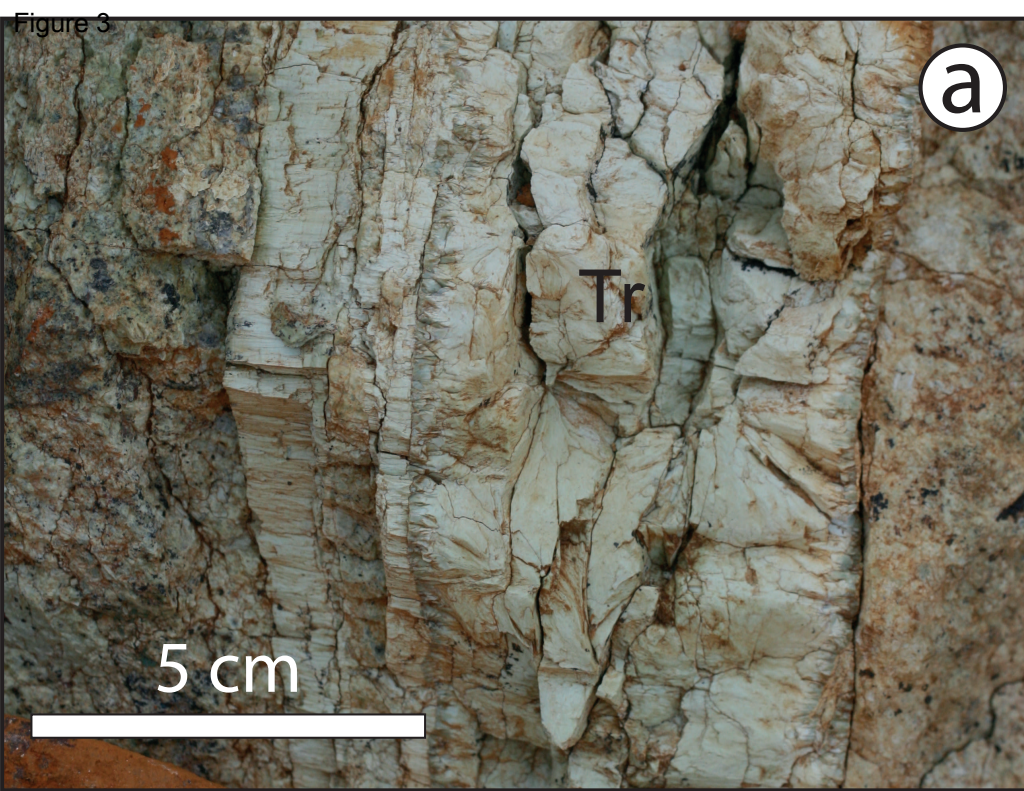


Figure 4

[Click here to access/download;Figure;Fig4_ShearPressSolution.eps](#)

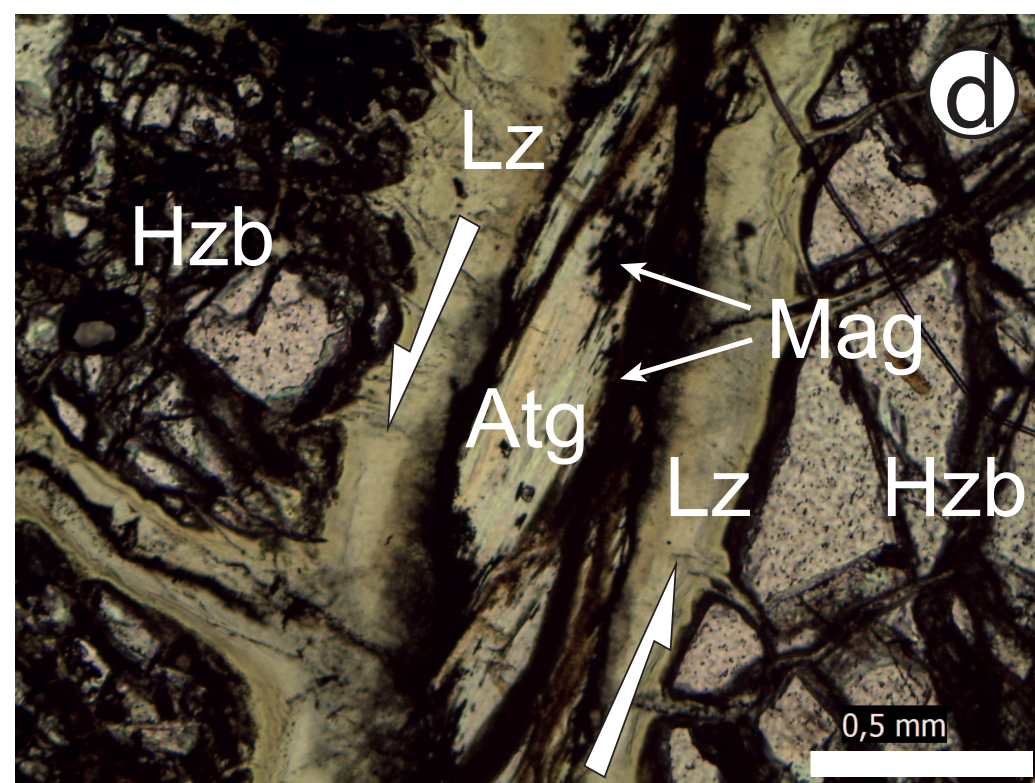
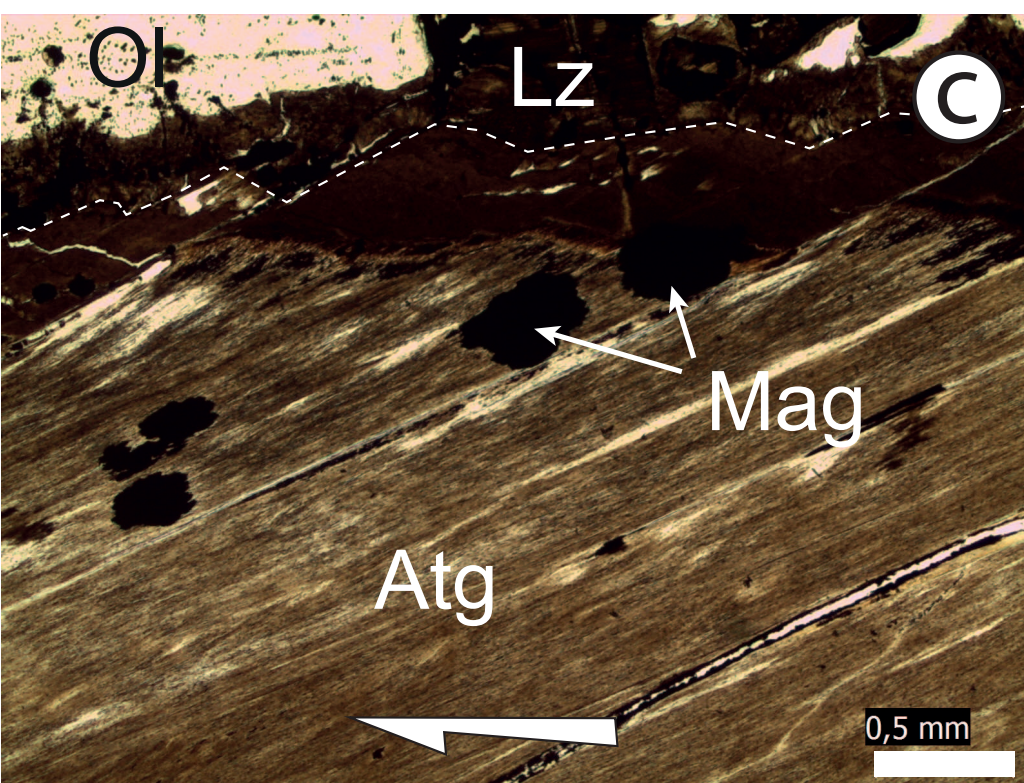
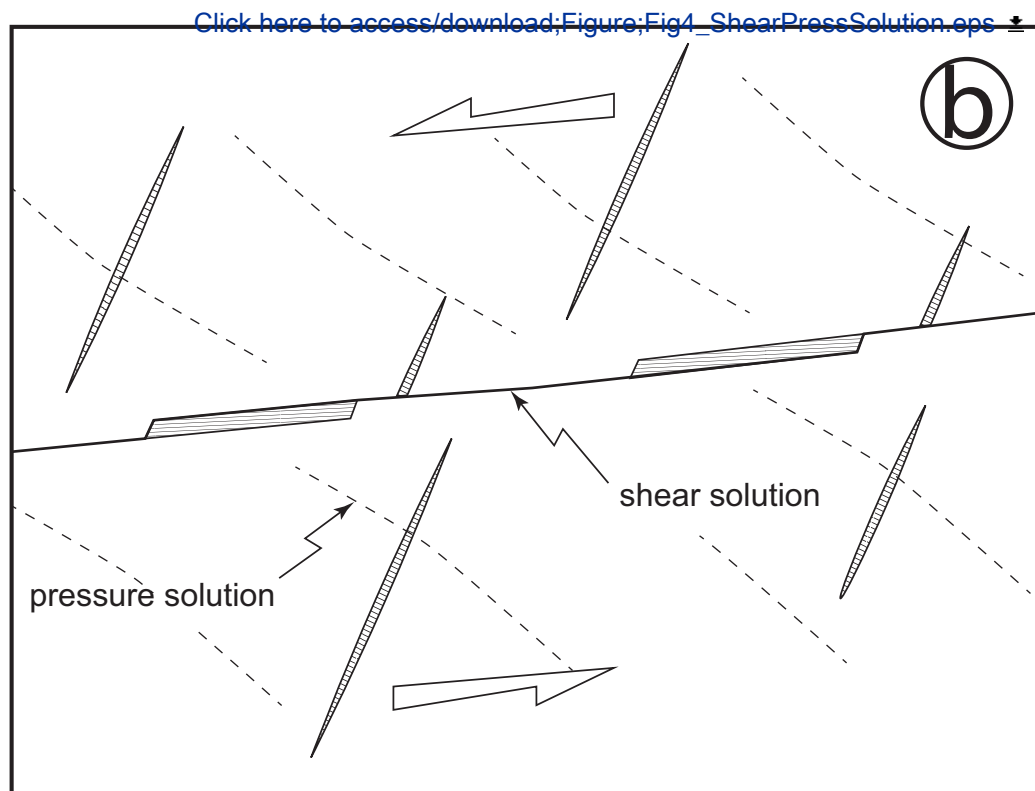
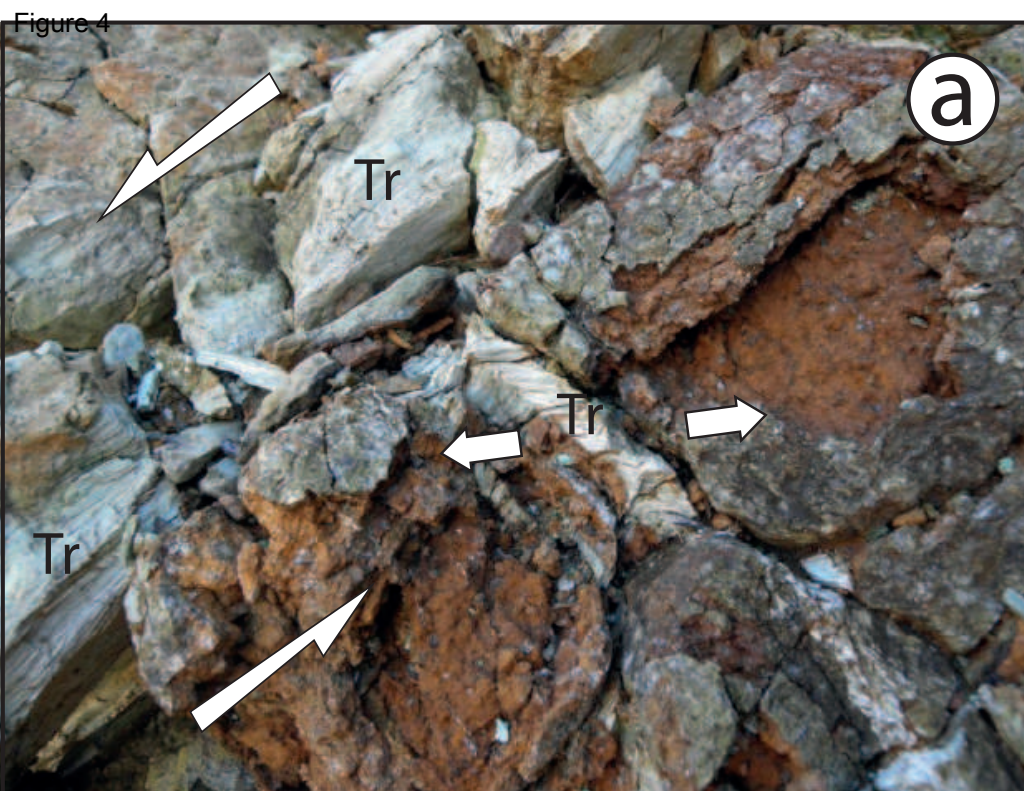
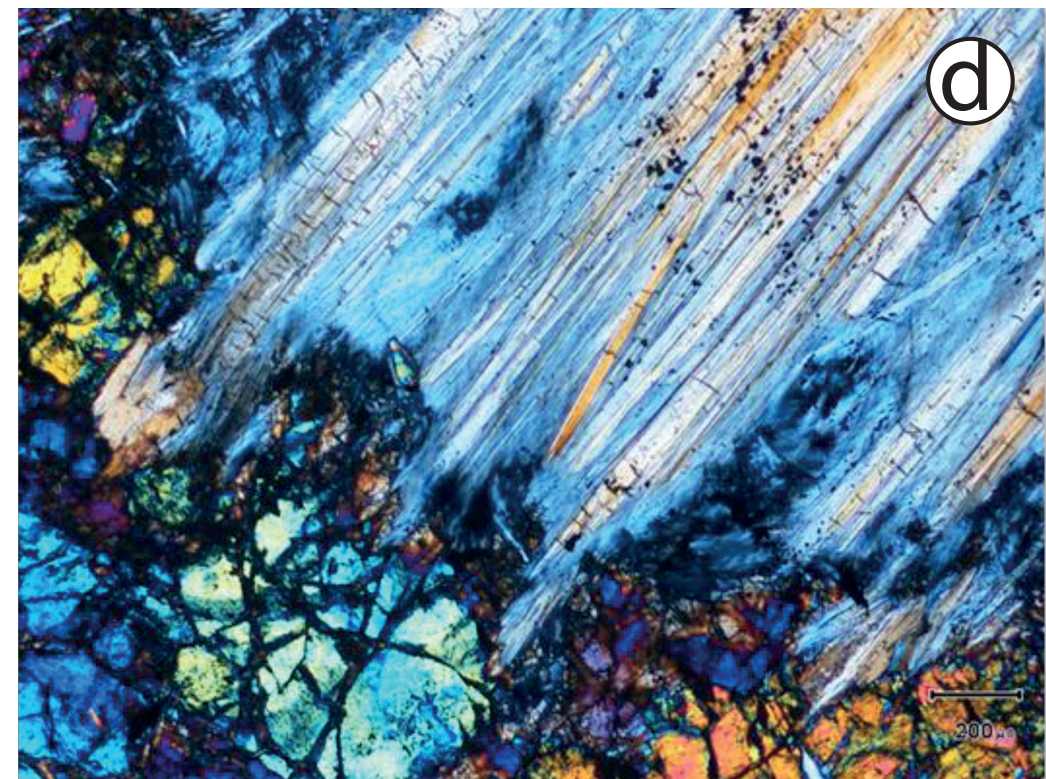
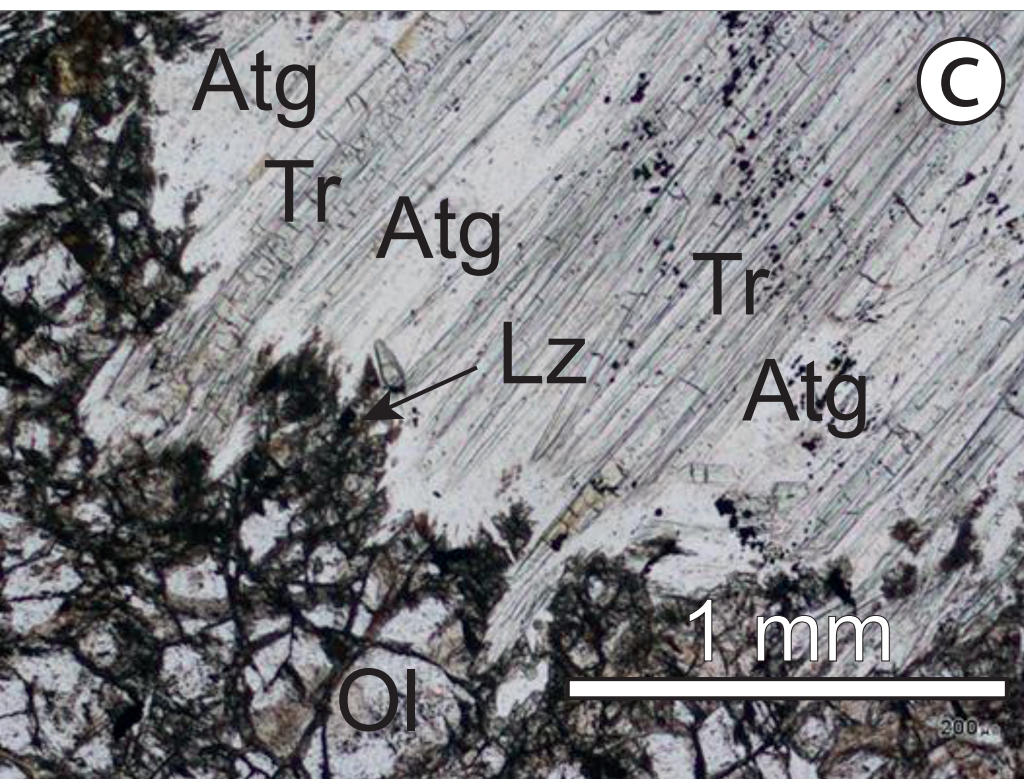
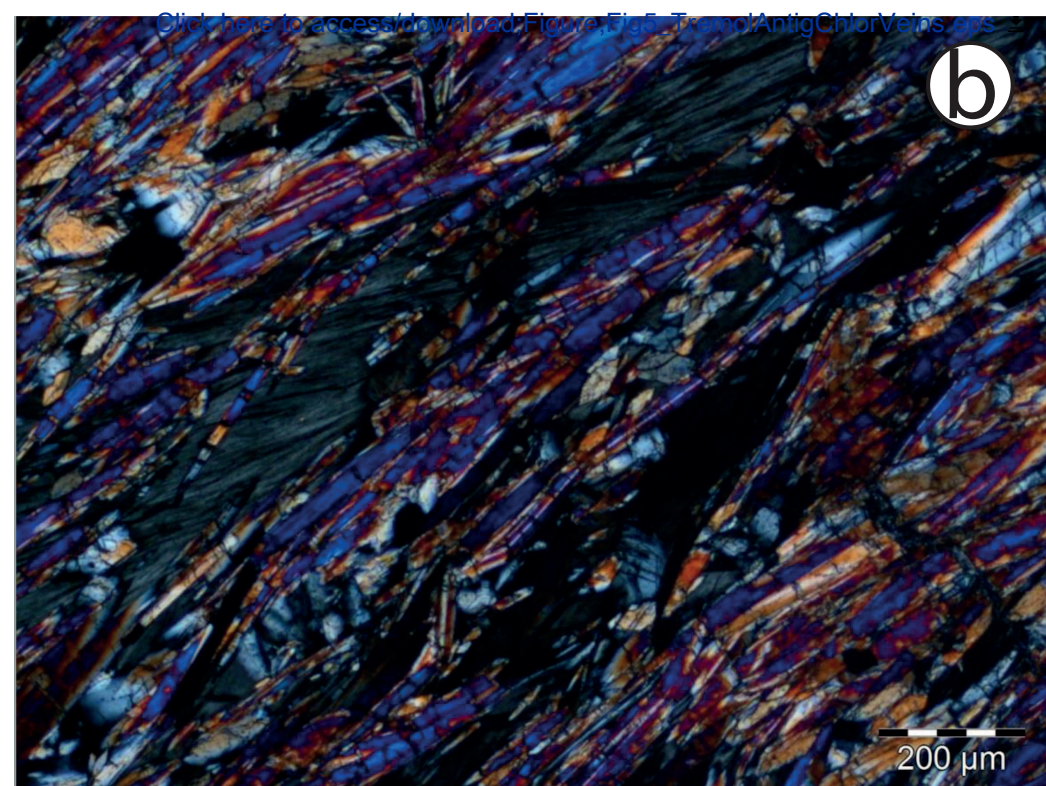
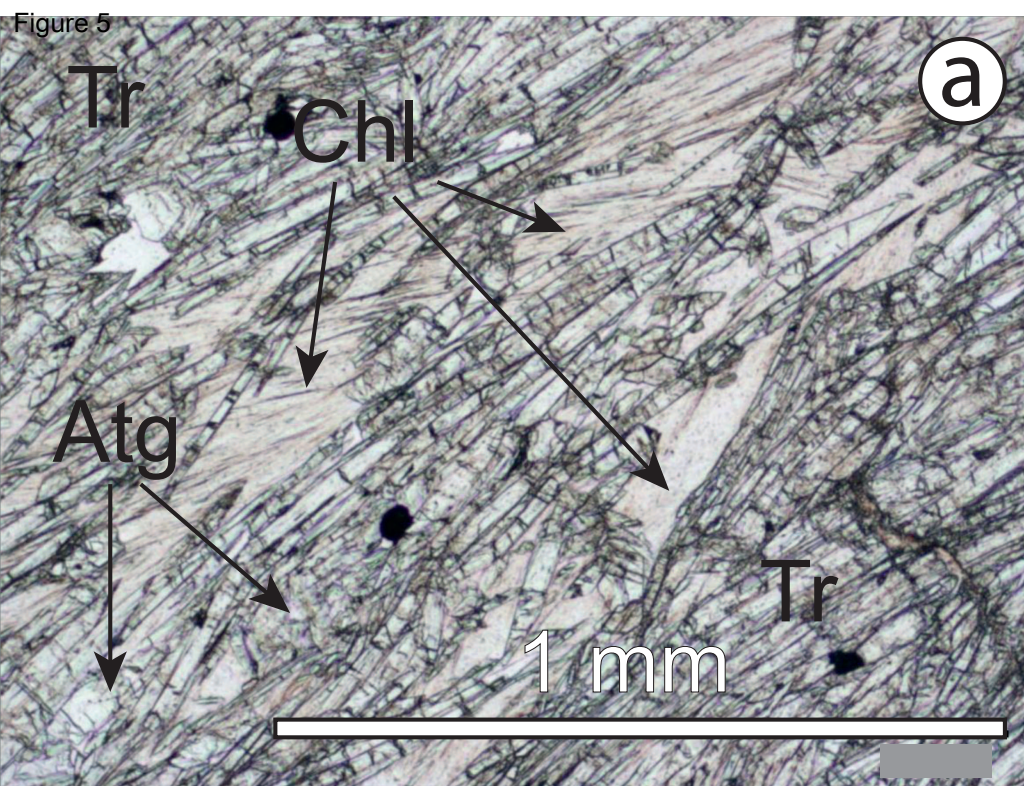
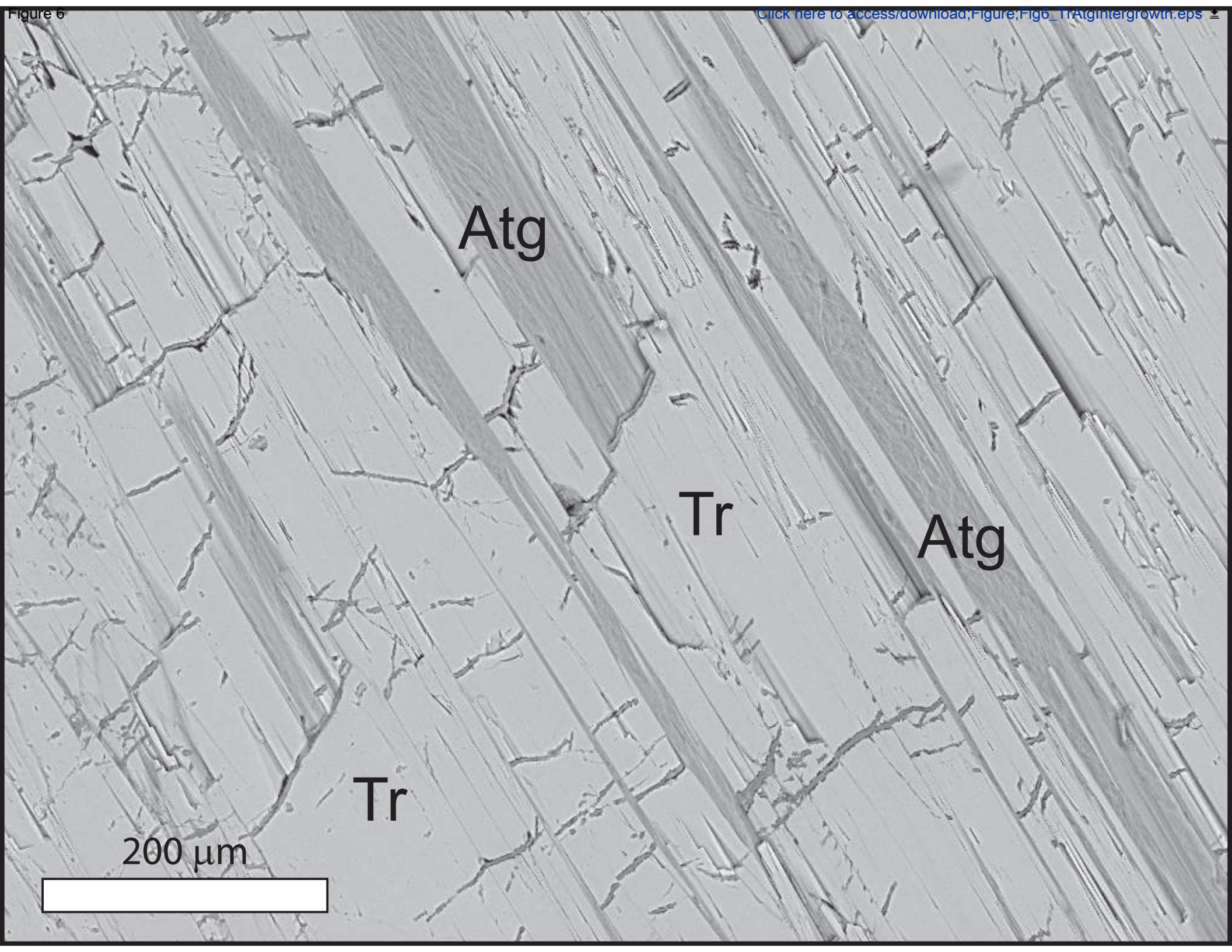


Figure 5





Atg

Tr

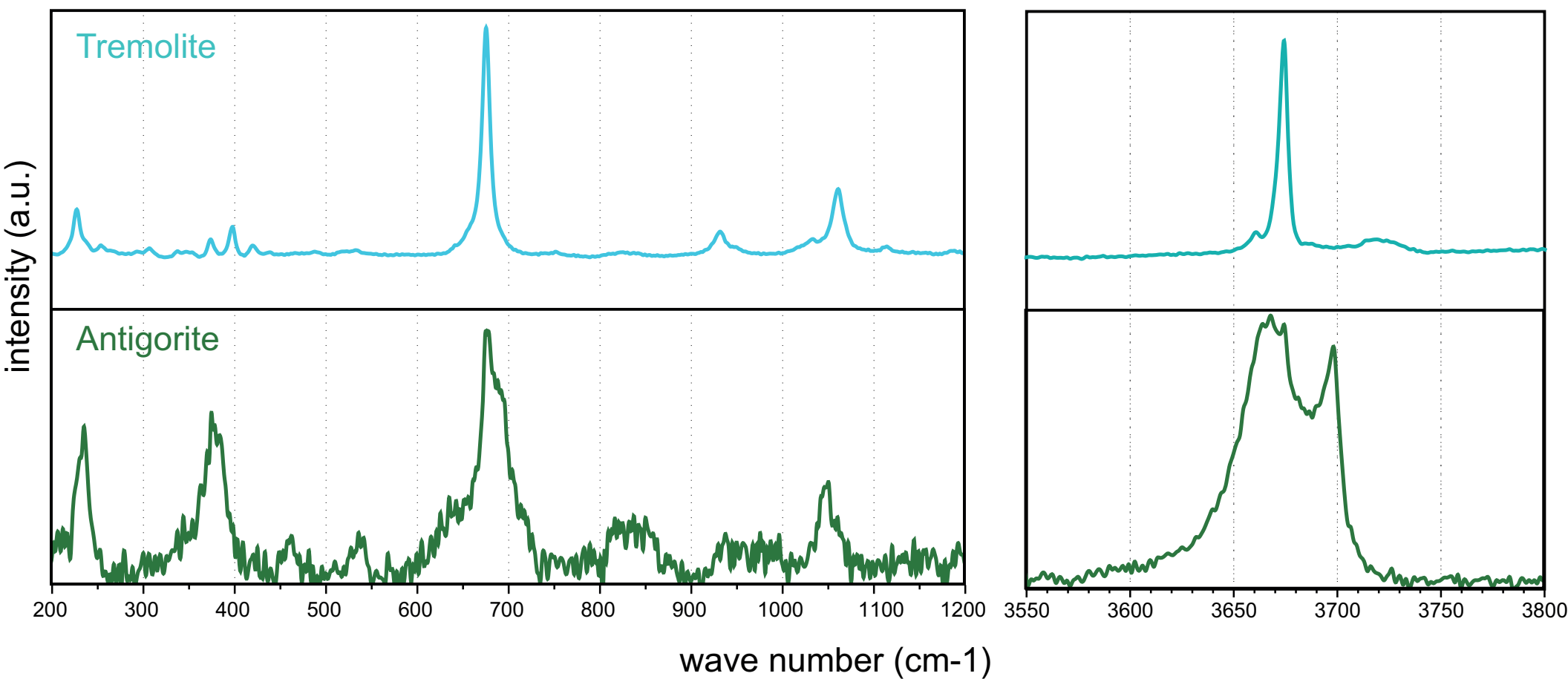
Atg

Tr

200 μm



Figure 7



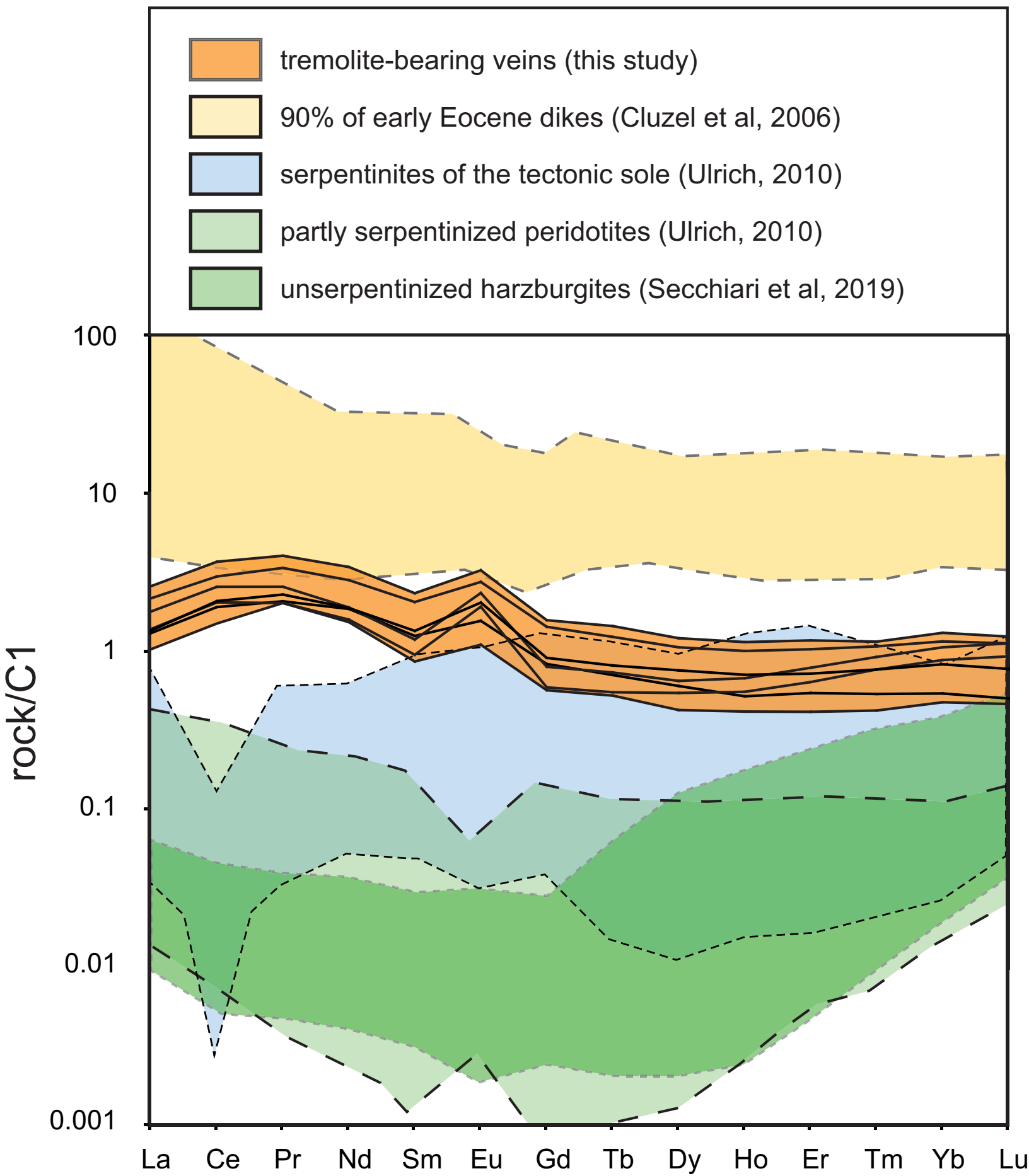
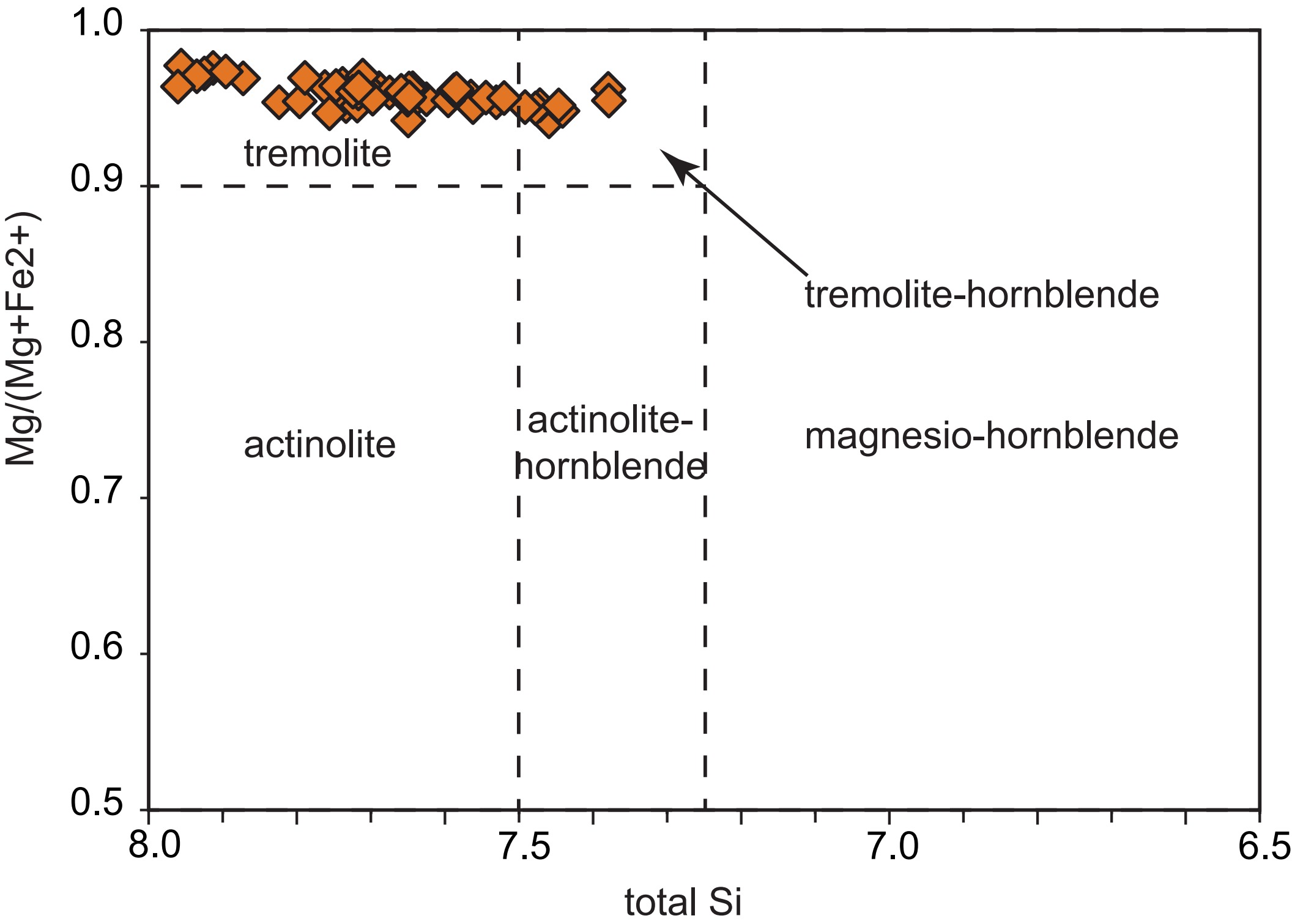
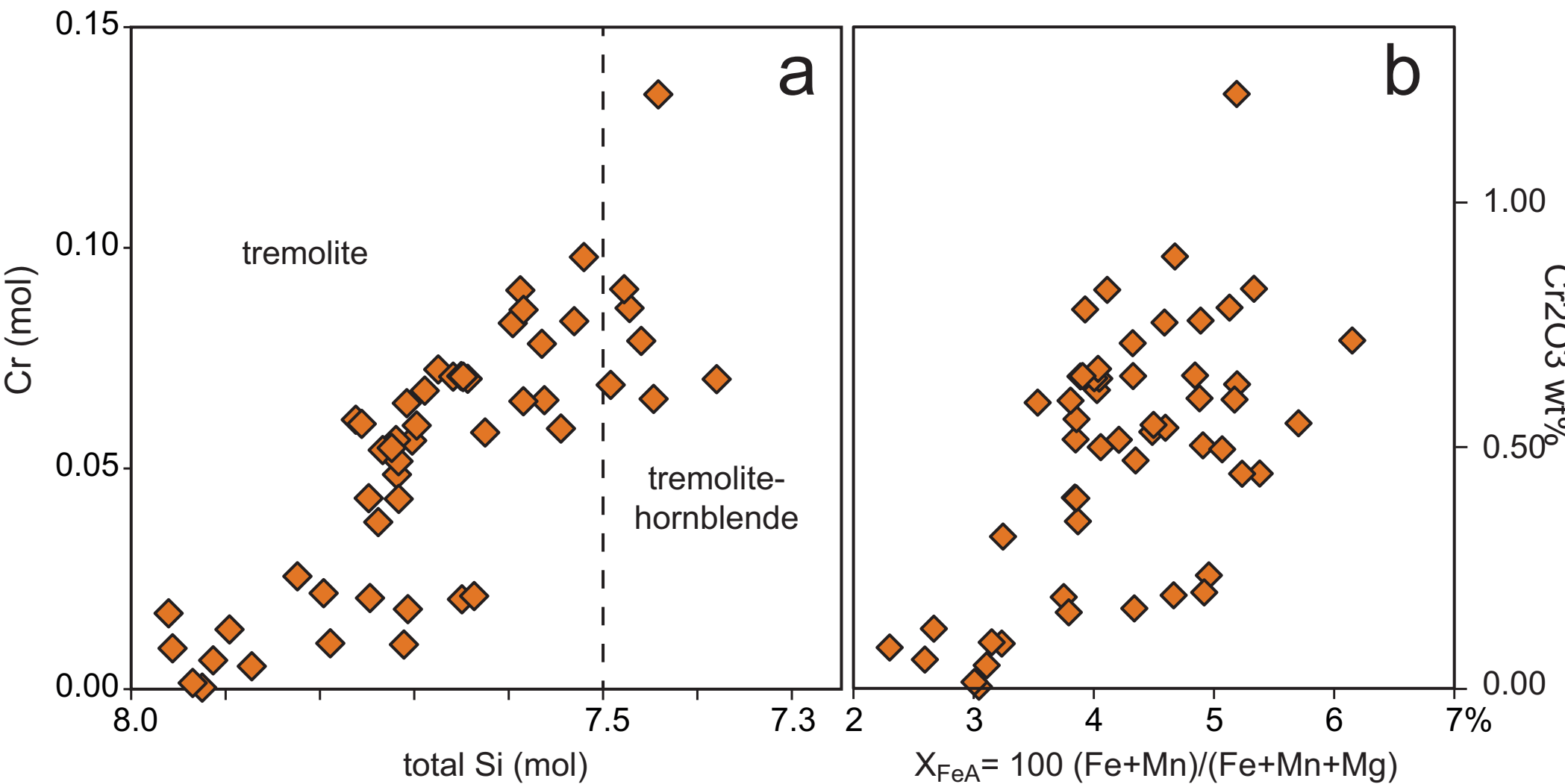


Figure 9





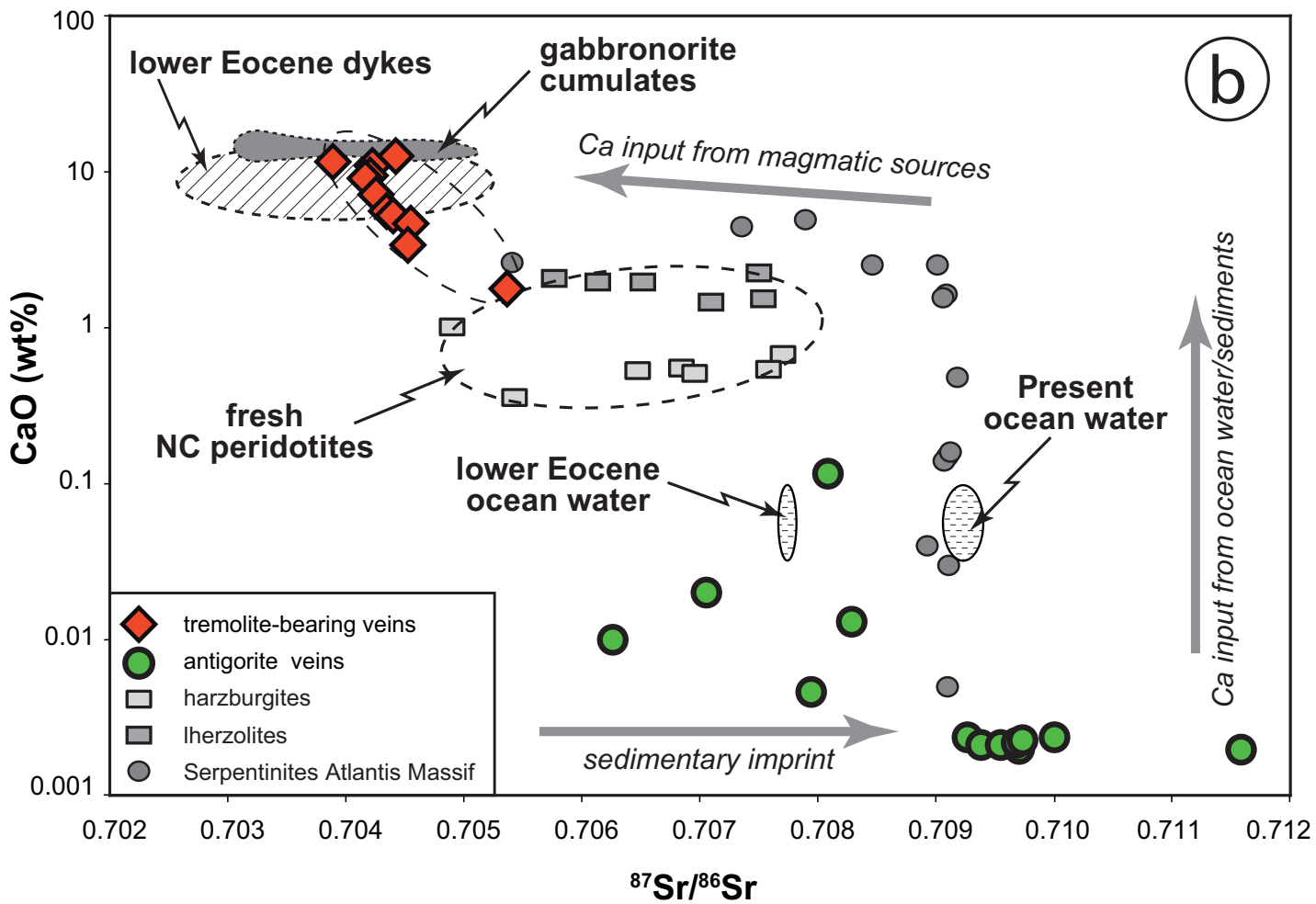
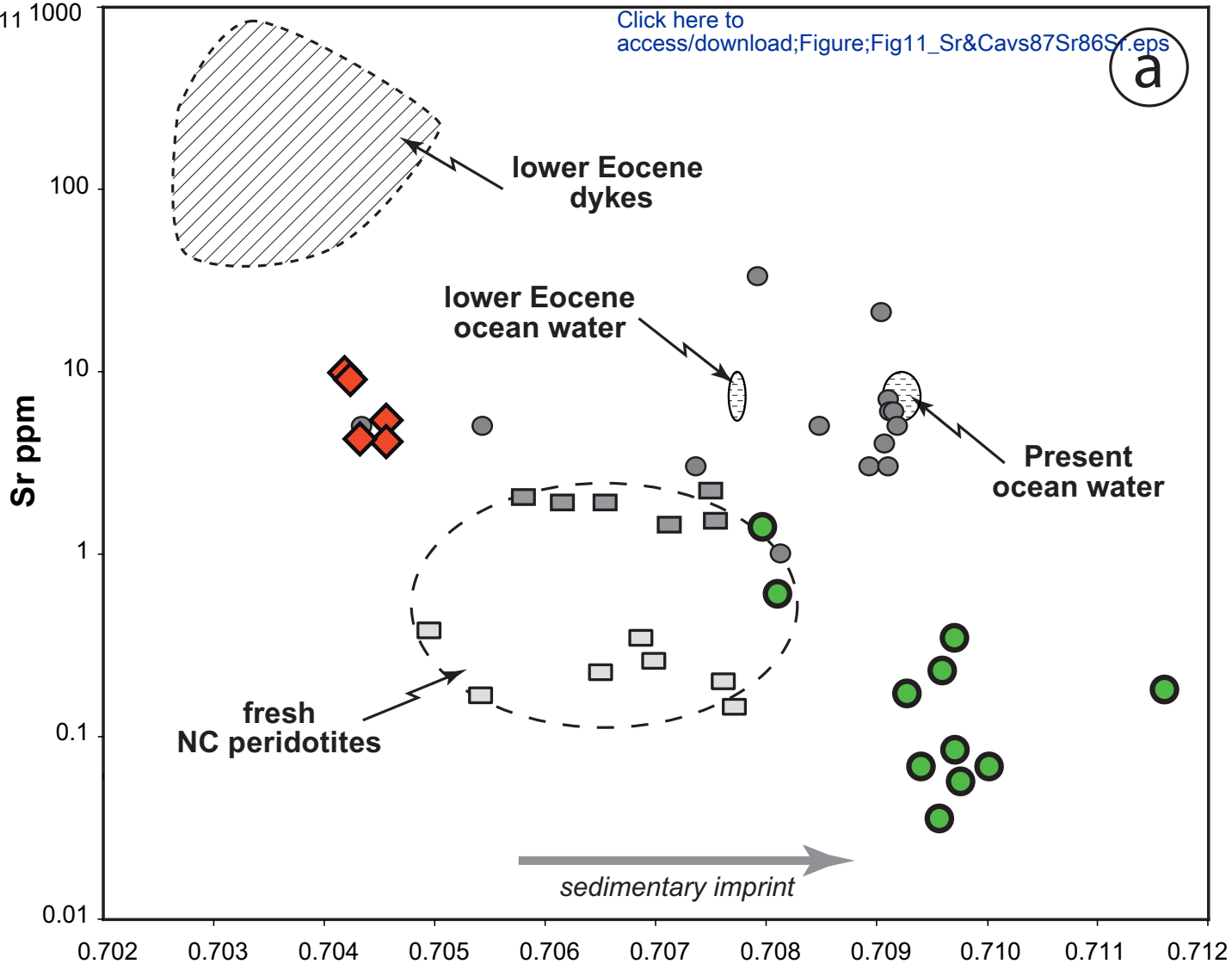


Figure 12

