

1 **Late Ordovician regional high-pressure metamorphism in Scotland:**
2 **Caledonian metamorphic climax predated the closure of Iapetus**

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24 **Abstract**

25 We document newly recognised Late Ordovician high-pressure (HP) metamorphism
26 in the Scottish Caledonides. Garnet growth at ca. 455–445 Ma within metabasic rocks
27 from the Ross of Mull is constrained to pressures >0.9 GPa and was associated with
28 formation of kyanite-bearing assemblages in meta-pelites that equilibrated at peak
29 conditions of 1.0–1.2 GPa and 700–780°C. This requires the burial of rock to ~40 km
30 depth driven by crustal shortening and thickening that *post-dated* Late Cambrian-Early
31 Ordovician Grampian arc-continent collision but *predated* the final Silurian closure of
32 the Iapetus Ocean, south of the Midland Valley. Similar garnet bearing amphibolites
33 dated at ca. 455–445 Ma on the Scottish north coast and Shetland, suggest that this
34 period of HP metamorphism was a regional feature across the Northern Highland
35 Terrane and Shetland. This was followed by Scandian nappe stacking and lower
36 pressure metamorphism at ca. 444–415 Ma, potentially forming a single protracted
37 orogenic phase prior to the final closure of Iapetus. There are several potential drivers
38 for this Late Ordovician event including: (1) subduction flip south of the Midland Valley
39 Terrane to NW-directed subduction followed by collision of cryptic outboard terranes
40 and/or Baltica, (2) continued SE directed subduction and collision of the Midland Valley
41 terrane with Laurentia, or (3) subduction-flip followed by NW-directed flat slab
42 subduction causing protracted accretionary orogenesis. Irrespective of the preferred
43 tectonic model, the climax of Caledonian orogenesis in Scotland predated terminal
44 continental collision.

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52 **Introduction**

53 Regional high-pressure (HP) metamorphic events result from deep burial and heating
54 of continental crust during orogenesis (e.g. Weller et al. 2021 and references therein).

55 Multiple HP events may occur within a single orogenic belt due to episodes of terrane
56 collision and/or changes in subduction geodynamics (e.g. Yonkee & Weil 2015 and
57 references therein). However, if accretionary orogenesis is terminated by continental
58 collision, the relative contributions of these tectonic processes to orogenic
59 development may be difficult to ascertain in ancient examples.

60

61 The Scottish Caledonides are widely regarded as having resulted from the closure of
62 the Lower Palaeozoic Iapetus Ocean and the collision of Laurentia, Baltica and
63 Avalonia (Fig. 1; Pickering et al. 1988; Soper et al. 1992). They are separated from
64 the Hebridean (Laurentian) foreland to the northwest by the ca. 430 Ma Moine Thrust
65 Zone (Fig. 2). To the southeast, the Northern Highland (NHT) and Grampian (GT)
66 terranes are dominated by Laurentian metasedimentary rocks and are separated by
67 the sinistral Great Glen Fault (GGF). The Midland Valley Terrane is a Cambrian-
68 Ordovician magmatic arc (the Grampian-Taconic arc), and immediately to the south,
69 the Southern Uplands Terrane is an accretionary prism separated from Avalonia by
70 the Iapetus Suture (Fig. 1; Chew & Strachan 2014 and references therein). The
71 Iapetus Ocean started to close in the Cambrian following development of a

72 southeastward dipping (ocean-facing) subduction zone near Laurentia which formed
73 the Grampian-Taconic arc (Fig 1a). During the early-middle Ordovician (ca. 490–465
74 Ma), the Laurentian margin was partially subducted under the arc, resulting in ophiolite
75 obduction, HP metamorphism and regional deformation and metamorphism of footwall
76 Dalradian and Moine metasedimentary successions of the NHT and GT – the
77 Grampian orogenic event (Lambert & McKerrow 1976; Dewey & Shackleton 1984;
78 Friedrich et al. 1999; Friend et al. 2000; Oliver et al. 2000; Chew & Strachan, 2014;
79 Johnson et al. 2017). This was followed by Late Ordovician metamorphism at ca. 455–
80 445 Ma (Bird et al. 2013), and Silurian metamorphism and deformation at ca. 444–415
81 Ma, both confined to the Northern Highland Terrane (NHT) (Fig. 2a). The sinistrally
82 oblique Laurentia-Avalonia collision has been considered as a ‘soft’ collision (Soper &
83 Woodcock 1990), without causing significant crustal thickening. Palaeomagnetic and
84 faunal data indicate that the northern Iapetus Ocean did not close until ca. 420 Ma
85 (Soper & Woodcock 1990), thus most of the orogenesis pre-dated final continental
86 collision.

87

88 There are, however, significant gaps in our current understanding of Caledonian
89 tectonics. The thermal evolution of GT Dalradian rocks is well understood as they were
90 not significantly overprinted after the Grampian orogenic event. In contrast, the thermal
91 evolution of NHT Moine rocks is more complex. Neoproterozoic regional metamorphic
92 assemblages were overprinted during the Caledonian orogeny *sensu lato* (Law et al.
93 2024; Strachan et al. 2024 and references therein). The pressure-temperature (PT)
94 conditions and tectonic driver(s) of the ca. 455–445 Ma event, and whether this is
95 distinct from the Grampian and Scandian events, are also uncertain (Bird et al. 2013;
96 Searle 2021; Walker et al. 2021; Law et al. 2024). Here we show that ca. 455–445 Ma

97 mineral assemblages on the Ross of Mull within the Scottish Caledonides are
98 associated with regional HP conditions. We interpret this as the early stage of a
99 protracted Late Ordovician-Silurian accretionary mountain building event which
100 predated closure of the Iapetus Ocean and terminal continental collision.

101

102 **Regional setting**

103 The Ross of Mull is located in the NHT and exposes a synformal inlier of Moine rocks
104 and associated metabasic intrusions (Figs 2 & 3). Across the NHT, the Moine rocks
105 are stacked as three nappes, collectively defining an inverted Caledonian
106 metamorphic gradient resulting from syn- to post-metamorphic thrusting (Fig. 2a;
107 Mazza et al., 2018). These nappes include:

108

109 (1) The Naver Nappe which underwent HP metamorphism during the Grampian
110 orogenic event, as indicated by garnet-clinopyroxene bearing assemblages that
111 equilibrated at P - T conditions of 1.0–1.2 GPa and 650–700°C (Friend et al. 2000). A
112 sillimanite-grade event followed at P - T conditions of ca. 0.8–0.9 GPa and 600°C with
113 decompression and heating to 0.6–0.7 GPa and 700 °C dated at ca. 444–415 Ma by
114 U–Pb monazite and xenotime (Ashley et al. 2015; Mako et al. 2019). Sm–Nd and Lu–
115 Hf Caledonian garnet dates indicate garnet growth between ca. 454–446 Ma, although
116 the metamorphic conditions are unknown.

117

118 (2) The Sgurr Beag Nappe with kyanite and sillimanite-bearing assemblages that
119 record P - T conditions of 0.7–0.9 GPa and 650–700°C, with U–Pb monazite and
120 titanite dates spanning ca. 473–464 Ma (Cutts et al. 2010; Mako et al. 2019) and Sm–
121 Nd and Lu–Hf garnet dates spanning ca. 475–463 Ma (Bird et al. 2013).

122

123 (3) The Moine Nappe which is associated with a decrease in P – T conditions westward,
124 and down-structural section, from 0.82 GPa and 680 °C in the east to 0.42 GPa and
125 550 °C in the west of the hanging-wall of the Moine Thrust (Ashley et al. 2015; Mazza
126 et al. 2018; Mako et al. 2019). U–Pb zircon ages from syn-thrusting plutons as well as
127 monazite and xenotime ages are consistent with Scandian nappe stacking and
128 metamorphism between ca. 444–415 Ma (Mako et al. 2019; Strachan et al. 2020). Lu–
129 Hf and Sm–Nd garnet geochronology suggest garnet growth mainly between ca. 455–
130 445 Ma (Bird et al. 2013).

131

132 Widespread upright ‘D3’ folding affected both the Sgurr Beag and Moine nappes after
133 they had been juxtaposed to form the Northern Highland Steep Belt. D3 folding is
134 bracketed by the 448.7 ± 2.9 Ma intrusion age of the deformed Glen Dessary syenite
135 (U–Pb zircon; Goodenough et al. 2011) and the 431.6 ± 1.3 Ma intrusion age of the
136 cross-cutting and undeformed Cluanie Pluton (U–Pb zircon; Milne et al. 2023) (Fig. 2).
137 Formation of the Northern Highland Steep Belt thus plausibly overlapped the ca. 455–
138 445 Ma metamorphic event recorded by Bird et al. (2013) in the NHT and Walker et
139 al. (2021) on Shetland.

140

141 On the Ross of Mull, the Moine rocks are exposed as a 6-km section to the east of the
142 Ross of Mull granite pluton (Fig. 3) (dated by U-Pb zircon at 418 ± 5 Ma; Oliver et al.
143 2000). Kyanite bearing mineral assemblages are regionally observed and overprinted
144 by andalusite, sillimanite and K-feldspar, associated with the metamorphic aureole of
145 the Ross of Mull pluton (Wheeler et al. 2004). On the western side of the Ross of Mull
146 pluton, on the Isle of Iona, cordierite occurs within the Iona group sediments,

147 interpreted to be part of the Neoproterozoic Torridon Group in the Caledonian foreland.
148 This suggests that the Ross of Mull pluton cuts across the projection of the Moine
149 Thrust and the cryptic Sound of Iona (SIF, Figure 3) normal fault. It is possible that this
150 structure influenced the intrusion of the Ross of Mull pluton, as inferred by top-to-east
151 normal sense shear fabrics observed in the Iona Group metasediments (Zaniewski et
152 al. 2006).

153

154 To the east of the Ross of Mull pluton, the Moine rocks are folded into a synform (the
155 Assapol synform). The structurally lowest Moine rocks correlate with the Morar Group
156 and the structurally highest with the Glenfinnan Group (Holdsworth et al. 1987), and
157 are separated by an intervening shear zone, although it remains uncertain whether
158 this is the Sgurr Beag Thrust (Holdsworth et al. 1987 vs. Krabbendam et al. 2022). In
159 the Glenfinnan Group (the structurally highest rocks), meta-sandstones are
160 intercalated with metabasic sheets (now garnet amphibolites) and metapelitic schists
161 (with garnet, biotite, muscovite and locally kyanite). Published thermobarometry is
162 restricted to the latter with P – T conditions estimated at 0.8 GPa at a temperature of
163 600–650°C (Wheeler et al. 2004). However, the timing of kyanite grade metamorphism
164 has hitherto been unknown. Bird et al. (2013) reported a 448.7 ± 5 Ma Lu–Hf garnet
165 age from a garnet amphibolite at Ardalanish Bay (Fig. 3), similar to ca. 455–445 Ma
166 Lu–Hf garnet dates recorded regionally across the Moine and Naver nappes on
167 mainland Scotland. However, it is unclear what P – T conditions these garnet dates
168 relate to. The rocks on the Ross of Mull therefore represent an opportunity to
169 determine the P – T conditions of this hitherto poorly understood Late Ordovician
170 metamorphism in the Scottish Caledonides as they are likely located at an
171 intermediate structural level within thrust sheets of the NHT (Figs. 2A, 3A).

172

173 **Field Relationships and Petrography**

174 Two samples of garnet amphibolite (19TL20, 19TL21) were collected from ~2–15 m
175 wide NNE-SSW trending mafic bodies at Ardalanish Bay, approximately 1 km east of
176 the Ross of Mull Granite (Fig. 4). A metapelite (WMAR007) was also collected from
177 ~700 m to the east of the Ross of Mull pluton. Outcrop relations and sample
178 photomicrographs are presented in Figures 4 and 5, respectively. The amphibolites
179 are characterized by garnet porphyroblasts with rotational inclusion trails (S_1), that are
180 discordant to the amphibole-bearing matrix which exhibits a continuous internal
181 foliation (S_2), (Figures 4 and 5). This foliation is parallel with the foliation of the
182 metasedimentary host rock that is deformed into westward verging minor folds (Fig.
183 5), therefore indicating the same metamorphic and deformation history. Millimetre to
184 centimetre sized segregations of leucosome are locally aligned with S_2 in the
185 amphibolite (Fig. 5) and have trondhjemitic (plagioclase-quartz) compositions,
186 indicating that conditions locally surpassed the water saturated solidus (Weinberg et
187 al., 2015; Palin et al., 2016). Metapelites contain garnet, biotite, muscovite, rutile and
188 locally K-feldspar and kyanite and have similarly trondhjemitic leucosome
189 segregations that are ptymatically folded and also display a westward fold vergence
190 direction. A penetrative spaced foliation (S_2) is axial planar to these folds and often
191 defined by alignment of muscovite (Figs 5 and 6). These observations suggest that
192 westward directed shearing occurred during or shortly following anatexis.

193

194 Within the metabasic bodies, a bimodal distribution of garnet sizes occurs in discrete
195 30 cm to metre sized domains that are oriented parallel with intrusion margins (Fig.
196 4B). Sample 19TL20 has abundant small garnets and 19TL21 contains fewer but

197 larger cm-sized garnets, although both samples have similar proportions of garnet
198 (15%), ilmenite (7%), quartz (10%), rutile (0.5%) and titanite (0.7%) with leucocratic
199 plagioclase (9%). Domains with smaller garnets (19TL20) contain no biotite or epidote
200 and more amphibole (56%) whereas domains with larger garnets (19TL21) have
201 greater abundances of hydrous phases (13% biotite and 3% epidote) and although
202 less amphibole (42%) and are associated or in contact with trondjhemitic leucosomes
203 (Fig. 5).

204

205 In contrast, metapelite sample WMAR007 preserves compositional layering (S_0) that
206 is characterized by mica-rich domains with muscovite and biotite up to several cm
207 across. Fine grained quartzo-felspathic domains up to 1 cm wide containing
208 plagioclase quartz, garnet, and K-feldspar that define S_1 . S_1 is subsequently folded on
209 the cm-metre scale forming crenulations, with a penetrative spaced foliation (S_2)
210 aligned axial planar to the folds. Garnets often show atoll-like textures (Fig. 5), with
211 the core replaced by biotite, plagioclase and muscovite, and the outer rims resorbed
212 by retrograde biotite. However, in some garnets (e.g., garnet 1), the core is partially
213 preserved. Rutile, ilmenite and titanite also occur as secondary phases throughout the
214 matrix, consistent with a HP assemblage variably overprinted by retrogression.
215 Although kyanite was seen in micaceous layers at outcrop, the thin section used for
216 thermobarometry from sample WMAR007 does not contain kyanite.

217

218 **Mineral Chemistry**

219 The compositions of phases in samples 19TL20, 19TL21 and WMAR007 were derived
220 from a CAMECA SX5-FE (field emission) electron microprobe at the Department of
221 Earth Sciences, University of Oxford. Operating conditions involved an accelerating

222 voltage of 15.0 keV corresponding to a current of 20 nA, a range of primary and
223 secondary standards were used including andradite (Fe, Mg, Ca), TiO₂ (Ti), Mn metal
224 (Mn), labradorite (Na, Al, Si) and sanidine (K) for major elements and synthetic
225 standards for (from the University of Edinburgh) for the rare earth elements (REE's).
226 Garnet line profiles were collected using a 10- μ m step size across all garnets. Mineral
227 abbreviations follow the guidelines of Whitney and Evans (2010). Anhydrous phase
228 compositions were calculated to standard numbers of oxygen per formula unit (Deer
229 et al., 1992), micas were recalculated to 11 oxygens, and chlorite to 28 oxygens.
230 Where present, H₂O content was assumed to occur in stoichiometric amounts. The
231 proportion XFe³⁺ (Fe³⁺/Fe_{total}) was calculated using AX (Holland, 2009) and amphibole
232 exchange vectors were calculated using the method of Droop, (1987), the complete
233 electron probe microanalyses are presented in Supplementary Table S1.

234

235 In garnet amphibolite samples 19TL20 and 19TL21, chemical traverses across
236 garnets show overlapping, flat and homogenised profiles [$X_{Alm}(0.56-0.63)$ $X_{Prp}(0.12-$
237 $0.14)$, $X_{Grs}(0.24-0.27)$ and $X_{Spss}(0.02-0.04)$] (Fig. 5). These flat profiles are observed
238 irrespective of garnet size. Matrix amphiboles across these two samples have similar
239 Tschermakitic-Ferro-tschermakitic compositions (Supplementary Material). The
240 similarity in garnet and amphibole chemistry and modal abundances between the two
241 samples is consistent with almost identical bulk rock chemical compositions derived
242 from X-ray fluorescence. However, subtle differences do occur between samples.
243 Domains containing cm-sized, fewer garnets (sample 19TL21), have greater
244 abundances of hydrous phases (biotite and epidote) and are closely associated or in
245 contact with trondhjemitic leucosomes (Fig. 4). This suggests that domains with larger
246 garnets were more hydrated and that garnet growth in the amphibolites was facilitated

247 by the presence of fluid or melt. This is also apparent from the increased stability of
248 garnet with increasing Molar H₂O at a given pressure and temperature in sample
249 19TL20 (see below and in Fig. 7). Whereas in sample WMAR007 garnets show an
250 increase in spessartine (Mn) content from core to outer rim, indicative of garnet
251 resorption by biotite and retrograde net transfer reactions (Kohn and Spear, 2000). In
252 contrast, pyrope, grossular and almandine decrease from the garnet cores to outer
253 rims [X_{Alm} (0.72-0.60) X_{Prp} (0.08-0.06), X_{Grs} (0.20-0.10) and X_{Spss} (0.02-0.10)] (Fig. 5).
254 These features suggest peak conditions are most likely to be preserved in the garnet
255 cores or interior, whereas the garnet rims likely constrain retrogression during
256 exhumation.

257

258 **Thermobarometry**

259 Thermobarometric calculations were performed using the following methods,
260 Amphibole-plagioclase thermometry (Holland and Blundy, 1994) was used for the
261 amphibolites and Ti-in-biotite thermometry (Henry et al., 2005) was used for biotite
262 and graphite bearing samples (19TL21 and WMAR007). Garnet-biotite thermometry
263 (Bhattacharya et al., 1992; Holdaway, 2000) and garnet-aluminosilicate-plagioclase
264 and garnet-plagioclase-muscovite-biotite barometry (Spear, 1993) was used for
265 metapelite sample WMAR007. THERMOCALC Average PT mode and dataset 62
266 (Holland and Powell, 2011) was used for all samples. The full results are presented in
267 Figs. 6-8 and Table 1.

268

269 Amphibole-plagioclase thermometry (Holland and Blundy, 1994), predicts a mean
270 temperature for sample 19TL21 of $671 \pm 15^\circ\text{C}$ using the Edenite-Tremolite
271 thermometer and $688 \pm 21^\circ\text{C}$ for the Edenite-Richterite thermometer using the

272 average matrix plagioclase anorthite composition ($An = 0.28$). In contrast, sample
273 19TL20 records a mean temperature of $665 \pm 15^\circ\text{C}$ for the Edenite-Tremolite
274 thermometer and $717 \pm 34^\circ\text{C}$ for the Edenite-Richterite thermometer (Supplementary
275 Material). Ti-in-biotite thermometry (Henry et al. 2005) predicts peak temperatures of
276 $748 \pm 12^\circ\text{C}$ for sample WMAR007, and $723 \pm 12^\circ\text{C}$ for sample 19TL21, and with most
277 biotite analyses clustering around $660\text{--}670^\circ\text{C}$ (Supplementary Material), similar to
278 amphibole-plagioclase thermometry. These results suggest that most biotite is
279 retrograde, and peak metamorphic temperatures in all samples likely exceeded
280 720°C .

281

282 For metapelite sample WMAR007, garnet-biotite thermometry combined with garnet-
283 aluminosilicate-plagioclase (GASP) and garnet-plagioclase-muscovite-biotite
284 (GPMB_Mg) barometry suggest that the garnet core (Grt 1) attained $638^\circ\text{C} \pm 30^\circ\text{C}$
285 and 1.28 GPa (GPMB_Mg), 1.04 GPa (GPMB_Fe) and 1.07 GPa (GASP) ± 0.12 GPa
286 respectively. However, caution must be taken with the GASP result as although
287 kyanite was seen in outcrop, the thin section sample did not contain kyanite. The GMB
288 result is considered robust, although calculated Grt-Bt temperatures likely represent a
289 minimum due to retrograde net transfer reactions (Kohn and Spear, 2000). In contrast,
290 garnet rims in contact with biotite record retrograde conditions (Grt 4) of $496 \pm 30^\circ\text{C}$
291 and 0.53 GPa (GASP), 0.67 GPa (GPMB_Mg) and 0.51 GPa (GPMB_Fe) ± 0.12 GPa
292 respectively. These results suggest cooling of the sample through the kyanite stability
293 field (Fig. 8) and is consistent with the observation of andalusite pseudomorphing
294 kyanite and the lack of regional sillimanite (Wheeler et al. 2004).

295

296 Calculations using THERMOCALC Average PT mode, based on garnet core
297 compositions and average matrix compositions suggest that the assemblage Grt-
298 Amp-Fsp-Rt-Ilm-Qz-Sph-H₂O in sample 19TL20 equilibrated at 1.06 GPa ± 0.13 GPa
299 and 755 ± 96°C whereas Grt-Amp-Bt-Pl-Rt-Ilm-Ep-Qz-Sph-H₂O in sample 19TL21
300 attained 1.02 GPa ± 0.11 GPa and 777 ± 54°C (Fig. 8). Calculations on metapelite
301 sample WMAR007 using garnet cores and the observed assemblage Grt-Ms-Bt-Kfs-
302 Pl-Qz-Rt-Ilm-H₂O suggest that it attained 1.06 ± 0.14 GPa and 750 ± 41°C (Fig. 8).

303

304 **Petrological Modelling**

305 Petrological modelling was performed in Theriak-Domino (de Capatani, 2010) using
306 Dataset 62 (Holland and Powell, 2011) using the bulk compositions determined by X-
307 ray fluorescence (XRF) (Table S2). An $X_{Fe^{3+}} = 0.3$ was used in modelling amphibolites
308 based on an estimated bulk rock composition from using amphibole compositions as
309 a proxy. Temperature-M(O) phase diagram calculations for 19TL20 at a pressure of
310 1.0 GPa also best reproduce the observed assemblage (Grt-Amp-Pl-Qz-Rt-Ilm-
311 Liq±Rt) when $X_{Fe^{3+}} \sim 0.1-0.35$ which is within error of average Fe^{3+} values for
312 metabasites = 0.26 ± 0.12 (Forshaw et al. 2024) (Fig. 7). For metapelite sample
313 WMAR007, an $X_{Fe^{3+}} = 0.3$ was also used, which is within error of the global median
314 $Fe^{3+} = 0.23$ for metapelites (Forshaw and Pattison, 2023). Bulk rock H₂O contents
315 used in modelling for amphibolite samples 19TL20 and 19TL21 was 4.5%mol, which
316 was determined by minimally saturating the solidus at a pressure of 0.75 GPa (Fig.
317 6A, B), with free H₂O as a volatile phase removed from the assemblage within 10°C
318 of the solidus. In contrast, in metapelite sample WMAR007 the loss on ignition (LOI)
319 determined from XRF was used for the H₂O as primary muscovite remained stable at
320 peak conditions and therefore the measured bulk rock water content likely represents

321 the water content within the rock at peak conditions. For amphibolite samples 19TL20
322 and 198TL21, the activity-composition models for amphibole, clinopyroxene,
323 orthopyroxene, biotite, chlorite, garnet and melt (Green et al., 2016); ternary feldspar
324 (Holland et al., 2022); epidote (Holland et al., 2011); white mica (White et al., 2014)
325 and ilmenite (White et al., 2000) were used. Because Mn is negligible in both
326 amphibolites, and Mn bearing activity models for amphibole are not available,
327 calculations were performed in the 10-component system $\text{Na}_2\text{O}-\text{CaO}-\text{K}_2\text{O}-\text{FeO}-\text{MgO}-$
328 $\text{Al}_2\text{O}_3-\text{SiO}_2-\text{H}_2\text{O}-\text{TiO}_2$ (NCKFMASHTO). In contrast, for metapelite sample WMAR007,
329 Mn was included in the calculations, and the pelite models for garnet, biotite, white
330 mica, staurolite, chlorite, melt, orthopyroxene and cordierite were used (White et al.
331 2014). The phase diagrams are presented in Figs. 6-7.

332

333 Phase diagram results show that garnet-bearing fields in the amphibolites are confined
334 to pressures exceeding 0.9 GPa (Fig. 6A-B). This provides a minimum pressure
335 constraint for garnet growth, and therefore a direct link between the ca. 448 Ma Lu–Hf
336 date and high-pressure metamorphic conditions (Figs. 6-7). A phase diagram for
337 sample 19TL20 (Fig. 6A) predicts clinopyroxene stable across the pseudosection,
338 which is inconsistent with the observed assemblage. This is a well-known issue with
339 the augite activity-composition model of Green et al. (2016), but it does not affect the
340 stability of other major phases (e.g., Forshaw et al., 2018). Furthermore, the predicted
341 Cpx is negligible (<2% volume), and therefore ignoring Cpx, the peak assemblage Grt-
342 Amp-Pl-Qz-Rt-Ilm-Liq for sample 19TL20 is reproduced between 750–900°C and
343 1.05–1.40 GPa. Intersection of garnet isopleths $X_{\text{Prp}}(0.13)$, $X_{\text{Grs}}(0.26)$ and the
344 observed (15%) garnet volume place even tighter $P-T$ constraints of 1.05–1.30 GPa
345 and 790–810°C. (Fig. 6C, E).

346

347 In contrast, the phase diagram of sample 19TL21 (Fig. 6B) does not predict Cpx until
348 much higher temperatures and the peak assemblage Grt-Amp-Pl-Bt-Rt-Ilm-Qz-Liq is
349 reproduced at 1.08–1.3 GPa and 750–810°C. Intersection of the observed garnet
350 isopleths $X_{Prp}(0.13)$, $X_{Grs}(0.26)$ and the observed (15%) garnet volume place even
351 tighter P – T constraints of 1.08–1.30 GPa and 760–780°C (Fig. 6D, F). These
352 calculated P – T constraints overlap within uncertainty, which is encouraging given the
353 samples were taken less than 1 metre apart.

354

355 A calculated phase diagram for metapelite sample WMAR007 (Fig. 7C) using an $X_{Fe^{3+}}$
356 = 0.3 predicts the observed assemblage Grt-Ms-Bt-Pl-Kfs-Ilm-Rt-Qz-Liq in a small
357 stability field at 1.30–1.32 GPa and 750–760°C. Garnet core isopleths intersections
358 $X_{Prp}(0.08)$, $X_{Grs}(0.20)$ constrain conditions ~1.2–1.3 GPa and 720–750°C in the Kfs
359 absent Grt-Ms-Bt-Pl-Ilm-Rt-Qz-Liq field (Fig. 7D). This field is however directly
360 adjacent to, and within ~10°C of the predicted appearance of K-feldspar, which is
361 within uncertainty of the model results (Palin et al. 2016) and therefore is in good
362 agreement with observations. Due to the elevated pressures predicted, these results
363 suggest that peak metamorphic conditions did not surpass muscovite breakdown
364 (which has a positive reaction P – T slope). This is consistent with the presence of
365 primary muscovite in WMAR007, and evidence for trondjemitic leucosomes that are
366 heterogeneously distributed across Ardalánish Bay. Both these observations suggest
367 that anatexis occurred by the local addition of water rather than incongruent melting
368 of muscovite. However, we acknowledge that closer to the Ross of Mull Pluton,
369 muscovite is absent, and fibrolitic sillimanite and K-feldspar occur in the groundmass,

370 inferred to be a result of muscovite breakdown at lower pressure (~0.3 GPa) after the
371 HP kyanite grade event (Wheeler et al. 2004) (Fig. 7).

372

373 **Discussion**

374 Integration of the new *P–T* constraints with published Lu–Hf garnet geochronology
375 from the same amphibolite suite (Bird et al. 2013) reveals that the 448.7 ± 5 Ma garnet
376 date constrains peak HP metamorphic conditions to ca. 1.0–1.2 GPa and 700–780°C
377 (Fig. 8). This is because garnet bearing fields in the amphibolites are constrained to
378 pressures exceeding 0.9 GPa, whereas in metapelites garnet growth can occur at
379 lower pressure due to more Mn in the bulk composition. A near identical Lu–Hf garnet
380 age of 447.3 ± 1.7 Ma was obtained by Bird et al. (2013) from a similar metabasic
381 intrusion in the Moine Nappe on the north coast of Scotland (Fig. 2A). Walker et al.
382 (2021) also presented Lu–Hf garnet ages from similar garnet amphibolites on Shetland
383 of 453.6 ± 5.1 Ma and 449.2 ± 2.3 Ma, and 452.0 ± 1.4 Ma from a kyanite bearing
384 gneiss. Together, these data suggest that similar HP conditions likely extended along
385 the western NHT and Shetland between ca. 455–445 Ma, and requires a (now
386 removed) ~40 km thick overburden.

387

388 At this time, rocks in the overlying thrust sheets in the eastern NHT (Sgurr Beag and
389 Vaver Nappes) and the GT show no record of the HP metamorphism and were
390 presumably experiencing exhumation and cooling (Mako et al. 2019; Law et al. 2024),
391 following earlier burial, underplating and Ordovician metamorphism due to the
392 Grampian arc-continent collision at ca. 490–480 Ma (Friend et al. 2000; Kutts et al.
393 2011). However, we see no evidence on the Ross of Mull for a regional lower-pressure
394 sillimanite-grade metamorphism between ca. 444–415 Ma that is recorded in the

395 underlying northern Moine Nappe (Mako et al., 2019; Mazza et al., 2018; Fig. 2). The
396 Ross of Mull kyanite-bearing rocks therefore likely occur at structurally intermediate
397 levels of the orogenic wedge and were not overprinted by later high-temperature
398 regional metamorphism.

399

400 Within the contact aureole of the Ross of Mull Granite (Fig. 3B), cordierite, sillimanite,
401 and andalusite overprint earlier kyanite-bearing assemblages at 0.3 GPa and 500–
402 750°C (Wheeler et al., 2004). This requires substantial exhumation of rock associated
403 with decompression from ca. 1.1–0.3 GPa between ca. 449–418 Ma. This
404 necessitates the removal of ~30 km of crust at a rate of ~1 km/ Myr. No regionally
405 significant extensional detachments have been identified (Law et al. 2024), and so
406 exhumation and cooling are here related to erosion driven by contemporaneous, and
407 continued convergence and underplating of the Laurentian foreland, ultimately
408 resulting in the development of the Moine Thrust Zone at ca. 430 Ma.

409

410 Because the ca. 455–445 Ma HP episode is confined to the NHT and was followed by
411 metamorphic ages spanning ca. 444–415 Ma (Mako et al. 2019), it has been
412 suggested that it represents a single, protracted Late Ordovician-Silurian orogenic
413 event (Law et al. 2024) that commenced prior to closure of the Iapetus Ocean at ca.
414 420 Ma. The HP metamorphic garnet ages presented by Bird et al. (2013) and Walker
415 et al. (2021) also shortly follow the ‘Grampian’ metamorphic ages by only ~4 Myrs.
416 Assuming realistic rates of convergence and dip of thrust faults resulting in the burial
417 of rock to depths of 40 km required by our metamorphic pressures of 1.2 GPa, this
418 raises the possibility that the Caledonian Orogeny in Scotland formed semi-
419 continuously and diachronously over a protracted ca. 50 Myr period.

420

421 However, the new data could be consistent with several competing tectonic models
422 that might account for such a protracted period of Caledonian orogenesis prior to
423 continental collision. At least three models need to be considered (Fig. 9): (1) a post-
424 Grampian flip to NW-dipping subduction was followed by Late Ordovician to Silurian
425 orogenesis in the NHT due to accretion of outboard terranes (Bird et al. 2013) and/or
426 segments of southern Baltica (Law et al. 2024), followed by ca. 700 km of late Silurian-
427 Devonian strike-slip displacement along the Great Glen Fault (GGF) to juxtapose the
428 NHT and GT in their present relative positions (Dewey & Strachan 2003) (Fig. 9A).
429 This model may explain the consistent timing of ca. 455–445 Ma HP metamorphism
430 confined to the NHT as a discrete event, however, such cryptic accreted micro-
431 continental terranes are not exposed, and the timing of the Baltica collision is
432 constrained to ca. 430–420 Ma after the ca. 455–445 Ma period of crustal thickening
433 and HP metamorphism; (2) GGF displacements are minimal and continuous SE-
434 directed underthrusting of Laurentia beneath the Midland Valley arc caused the locus
435 of crustal thickening and metamorphism to migrate northwestwards from the GT
436 across the NHT over ca. 50 Myrs (Searle, 2021) (Fig 9B). This model is consistent
437 with the diachroneity of deformation and metamorphism migrating towards the
438 Laurentian foreland but cannot explain the eventual closure of the Iapetus without
439 establishing another subduction zone to the south of the Midland Valley Terrane. As
440 such, Model 2 promotes an accretionary system; alternatively (3) subduction flip
441 during/after the Grampian event was followed by NW-dipping flat-slab subduction that
442 drove the locus of contractional deformation, magmatism and metamorphism inboard
443 (NW) from the trench (Bird et al. 2013; Dewey et al. 2015) (Fig. 9C). In this model,
444 there is no need for a cryptic micro-continent as in Model 1, yet it can explain closure

445 of the Iapetus to the south of the Midland Valley. Notably, we compare this latter style
446 of convergence with the North America Cordillera, where major crustal thickening and
447 high-grade metamorphism during the Laramide Orogeny was related to flat-slab
448 subduction during the Late Cretaceous to Paleocene due to increased tractions along
449 the plate interface and end-load at the trench (Yonkee & Weil, 2015 and references
450 therein; Lamont et al. 2024). This could account for the ca. 455–445 Ma HP regional
451 metamorphism prior to terminal continental collision. In addition, later rollback of the
452 low-angle slab would also increase asthenospheric heat-flow, and cause melting of
453 the sub-continental lithosphere that was previously hydrated during the earlier phase
454 of flat-slab subduction, ultimately forming the ‘Newer Granites’ at ca. 430–400 Ma
455 (Milne et al. 2023).

456

457 Irrespective of the preferred tectonic model, our data suggest that the Caledonian
458 Orogen in Scotland was an accretionary mountain belt with the climax of orogenesis
459 occurring prior to terminal continental collision of Laurentia, Avalonia and Baltica. This
460 is very different from the East Greenland (Laurentia) and Norway (Baltica) collision
461 (Fig. 1C) along strike, where HP metamorphism was associated with the final Iapetus
462 closure and extended into the Devonian (Weller et al. 2021).

463

464 **Conclusions**

465 We conclude that Late Ordovician HP metamorphism in the Scottish Caledonides
466 shortly followed the Grampian arc-continent collision but *predated* closure of the
467 Iapetus Ocean. The newly recognized HP metamorphism, reaching conditions of 1.0–
468 1.2 GPa and 700–780 °C at ca. 455–445 Ma likely reflects a period of accretionary
469 orogenesis that was continuous with terminal continental collision at ca. 430–425 Ma.

470 These contrasting styles and timing of crustal thickening and regional HP
471 metamorphism along the Iapetus Suture of the Caledonian Orogen emphasize that
472 protracted periods of mountain building throughout earth's history need not always
473 require terminal continental collision (Dewey 1982, Cawood et al. 2011).

474

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481

482 **Figures**

483 Figure 1. Plate tectonic reconstruction for the Iapetus Ocean and Caledonian Orogeny
484 based on Chew and Strachan, (2014).

485

486 Figure 2. A) Terrane map of the Caledonian Orogeny in Scotland B) Simplified
487 geological map of Scotland and P - T - t compilation, C) Summary of geochronological
488 data from the NHT and GT.

489

490 Figure 3: Schematic cross section through the Northern Highland Terrane, showing
491 the likely structural level for the Ross of Mull in relation to the larger scale structures.

492 B-D) Simplified geological map and cross sections of the Ross of Mull showing sample
493 locations, structures and key cross-cutting relationships.

494

495 Figure 4) Field photographs showing kyanite grade metamorphic assemblages and
496 partial melting at Ardalanish Bay. A) Outcrop photographs of kyanite grade
497 assemblages and partial melting metapelites with pyritically folded trondhjemitic
498 leucosomes containing peritectic garnet. B) Garnet amphibolite with larger garnets
499 concentrated along leucosomes. C) Retrogressed kyanite schist with kyanite
500 pseudomorphed by andalusite. D) Garnet amphibolite with peritectic garnet in
501 leucosome with plagioclase and quartz. E) metapelitic migmatite with leucosomes
502 aligned parallel to S_1 which are folded, and affected by boudinage along S_2 (sub-
503 horizontal east-west trending foliation) subparallel to the fold axes. F) Mesoscale
504 trondhjemitic leucogranite sill derived from the amphibolite. G) Outcrop of sample
505 WMAR007 with micaceous and quartz-felspathic banding, likely representing
506 compositional layering (S_0 - S_1). H) Outcrop photograph of the amphibolite (metabasic
507 intrusion) on Ardalanish Bay, showing the locations of 19TL20 and 19TL21 as well as
508 the continuous S_2 foliation of the amphibolite with the surrounding metasediments.

509

510 Figure 5. Photomicrographs of samples used in the study showing locations of garnet
511 line profiles. A-B) garnet amphibolites samples 19TL20 and 19TL21 showing
512 continuous matrix foliation which largely postdates garnet growth, note discordance
513 between S_1 trapped as inclusions in garnet and the external matrix foliation S_2 . C-D)
514 Photomicrograph of sample WMAR007 showing garnet breakdown textures to biotite,
515 enclaved in primary muscovite. E-F) characteristic chemical line profiles across garnet
516 used for thermobarometry.

517

518 Figure 6. Equilibrium phase diagrams of samples 19TL20 and 19TL21, A) 19TL20
519 predicted phase fields showing the peak assemblage in red text (excluding Cpx), B)
520 Equilibrium phase diagrams of sample 19TL21 showing the peak assemblage field in
521 red text, C) 19TL20 pyrope and grossular garnet isopleths, the green polygon
522 represents the observed garnet composition $X_{Prp}(0.13)$ and $X_{Grs}(0.27)$, D) 19TL21
523 pyrope and grossular garnet isopleths, the green polygon represents the observed
524 garnet composition $X_{Prp}(0.12)$ and $X_{Grs}(0.27)$. E) Predicted garnet modal proportions
525 (vol%) for amphibolite samples 19TL20 and F) 19TL21, showing good agreement
526 between observed ~15% volume garnet and garnet isopleth intersections in both
527 samples.

528

529 Figure 7. A) Binary equilibrium phase diagram of sample 19TL20 at a pressure = 1.0
530 GPa, A) $X_{Fe^{3+}}$ varying from 0-0.6 equating to varying molar O from 0.006% to 3.690%,
531 B) Molar (H_2O) varying from 0.05-20%. Calculated assemblage fields most closely
532 match observations (red text) when $X_{Fe^{3+}} \sim 0.3$ and $M(H_2O) = 4-15\%$, although rutile
533 bearing fields require higher pressures and suggest metamorphic conditions occurred
534 at 1.1 GPa. C) Equilibrium phase diagrams of sample WMAR007, predicted
535 assemblage fields and the peak assemblage in red text, D) pyrope and grossular
536 garnet composition isopleths, the green polygon represents the observed garnet
537 composition $X_{Prp}(0.075)$ and $X_{Grs}(0.20)$.

538

539 Figure 8. Summary $P-T-t$ path for the Ross of Mull. $P-T-t$ data for the Naver Nappe
540 (Friend et al. 2000), Sgurr Beag Nappe (Cutts et al. 2010) and Moine Nappe (Ashley
541 et al. 2015; Mazza et al. 2019) are shown for comparison.

542

543

544 Figure 9. Competing tectonic models for the Scottish Caledonides; A) Discrete
545 orogenic events model of Bird et al. (2013) related to i) collision of the Midland Valley
546 Arc resulting in the 'Grampian Orogeny', ii) collision of a cryptic microcontinent
547 resulting in the 'Grampian II Orogeny', and iii) collision of Baltica resulting in the
548 'Scandian Orogeny' before ca. 700km sinistral motion on the Great Glen Fault. B)
549 Continuous SW-directed underthrusting of Laurentia model of Searle, (2021), resulting
550 in diachronous metamorphism and deformation that migrated towards the NW with
551 time with minimal off-set on the Great Glen Fault. C) Flat-slab subduction model after
552 a post-Grampian 'subduction flip', resulting in diachronous deformation and
553 metamorphism, without necessarily requiring large offsets on the Great-Glen Fault.
554 Subsequent slab-rollback after ca. 430 Ma would cause melting of the previously
555 hydrated lithospheric mantle that was previously hydrated during flat-slab subduction,
556 resulting in the 'Newer Granites'.

557

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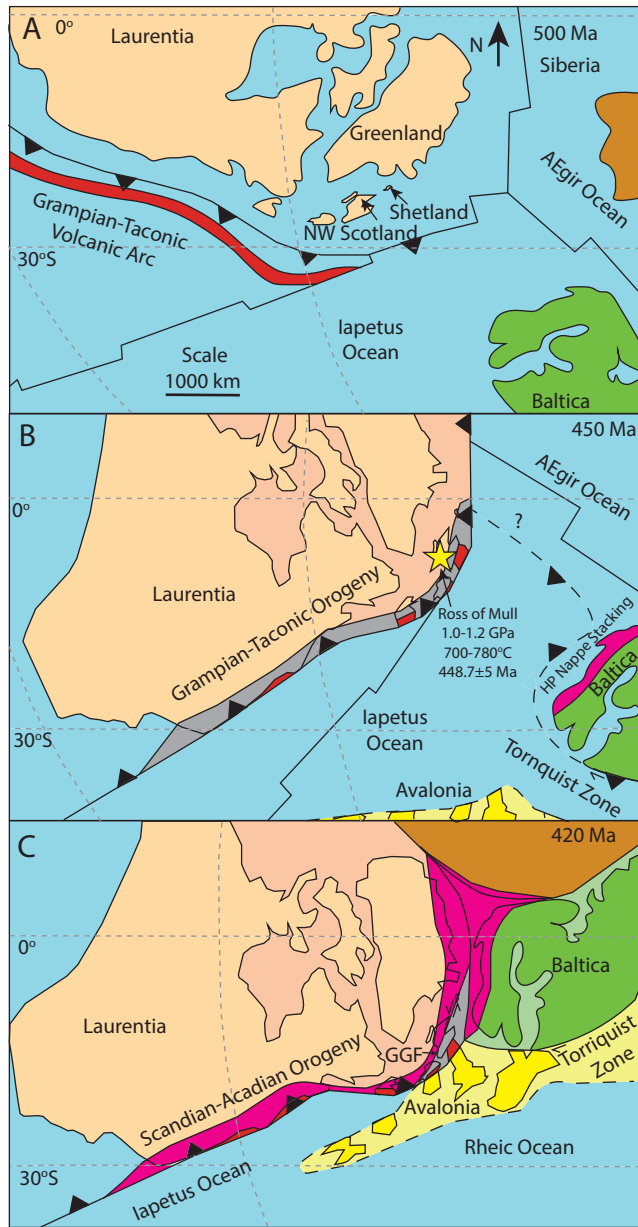
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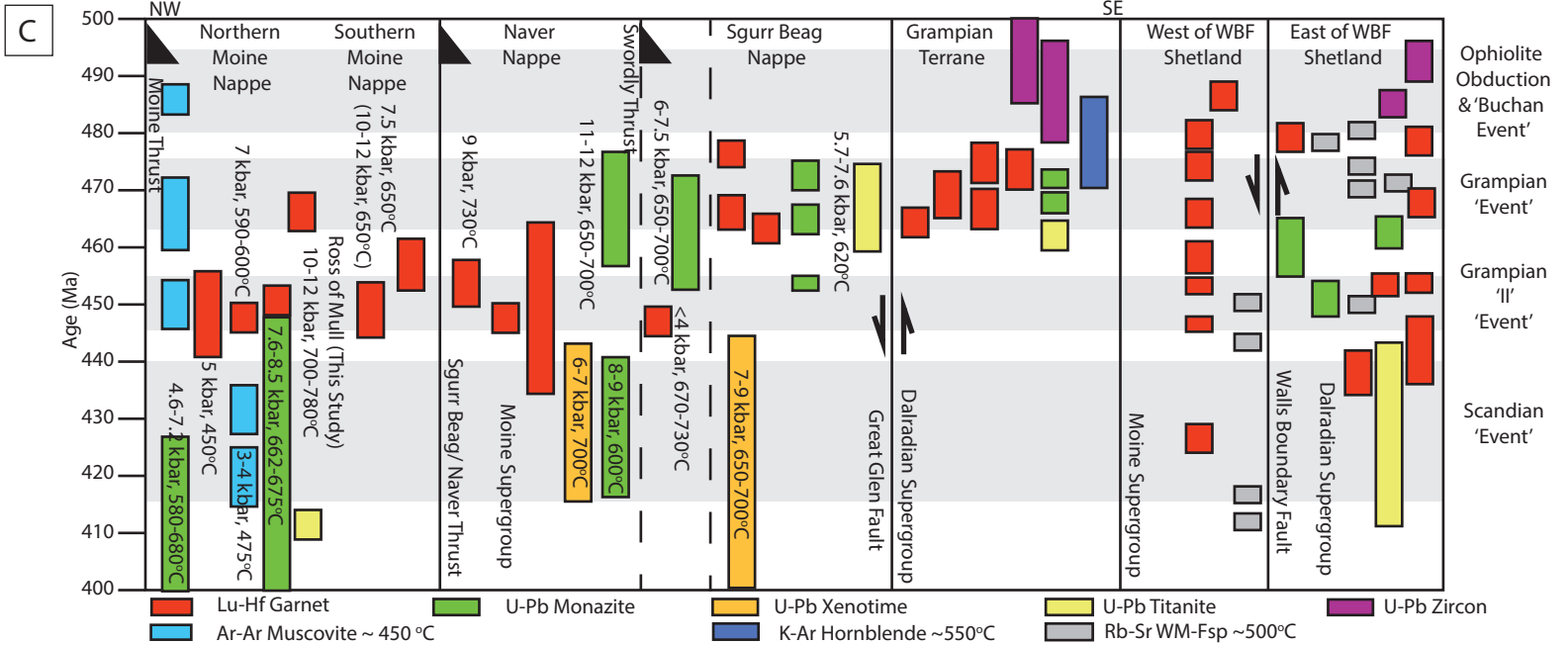
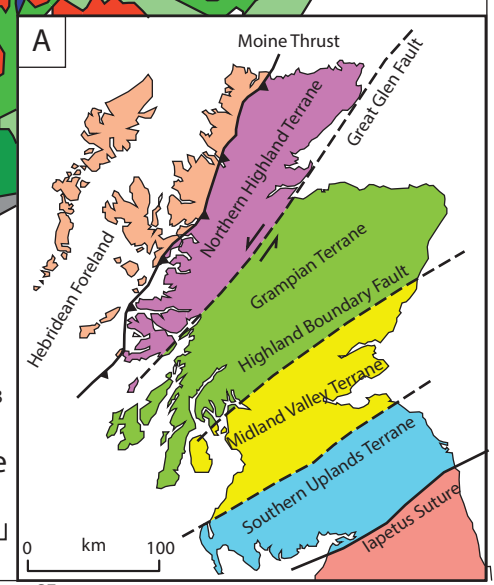
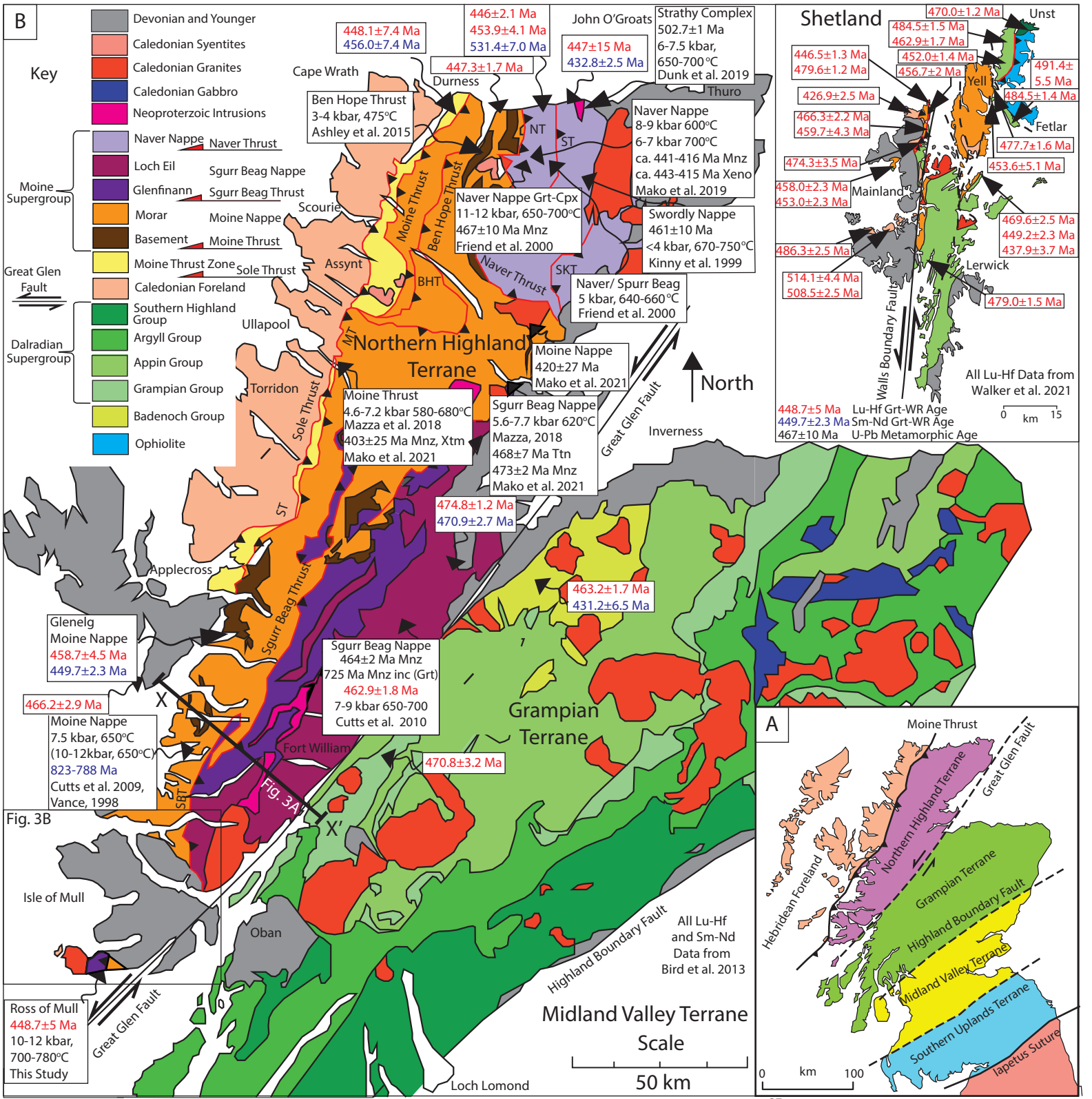
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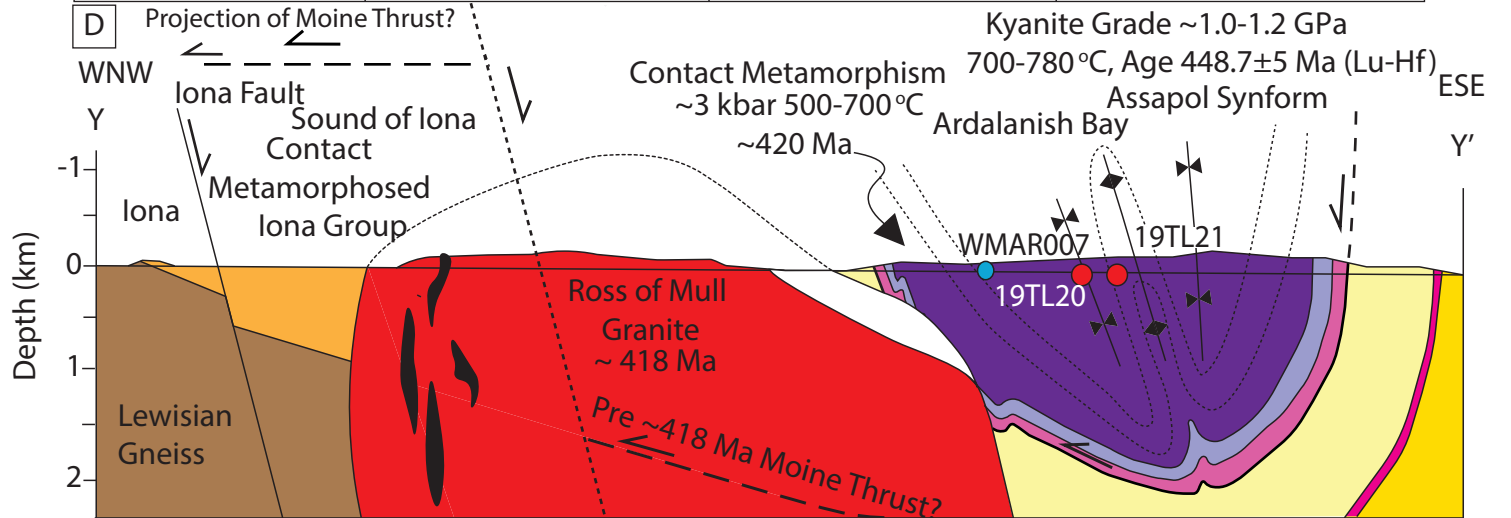
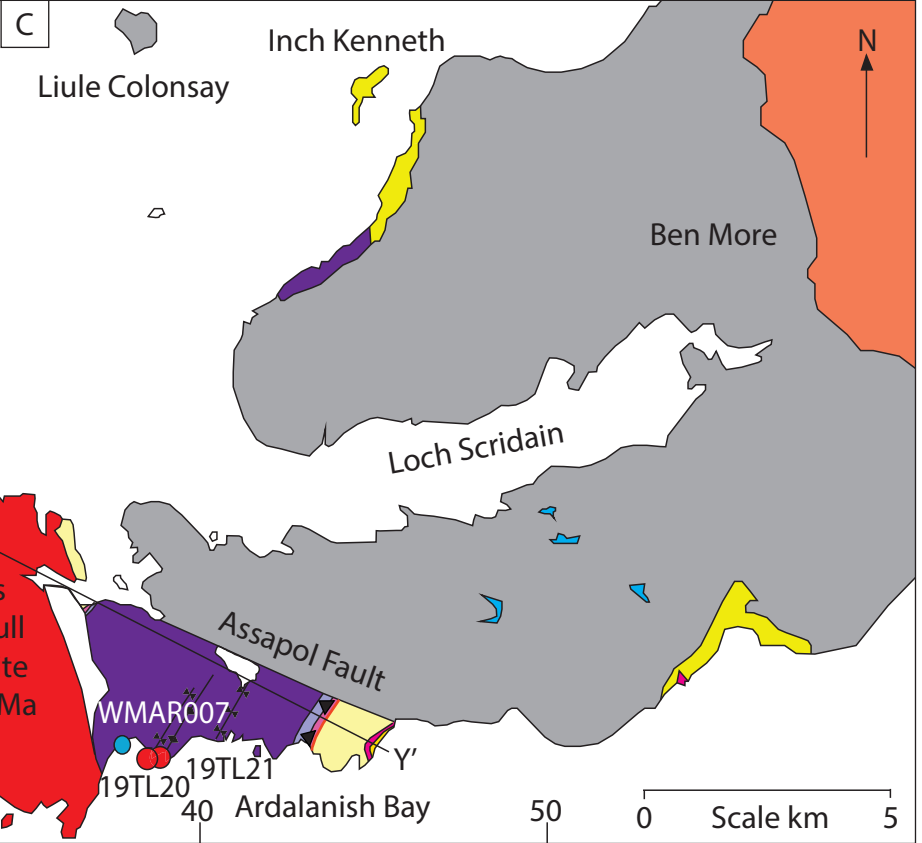
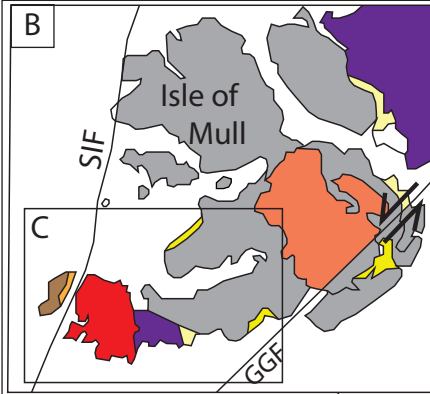
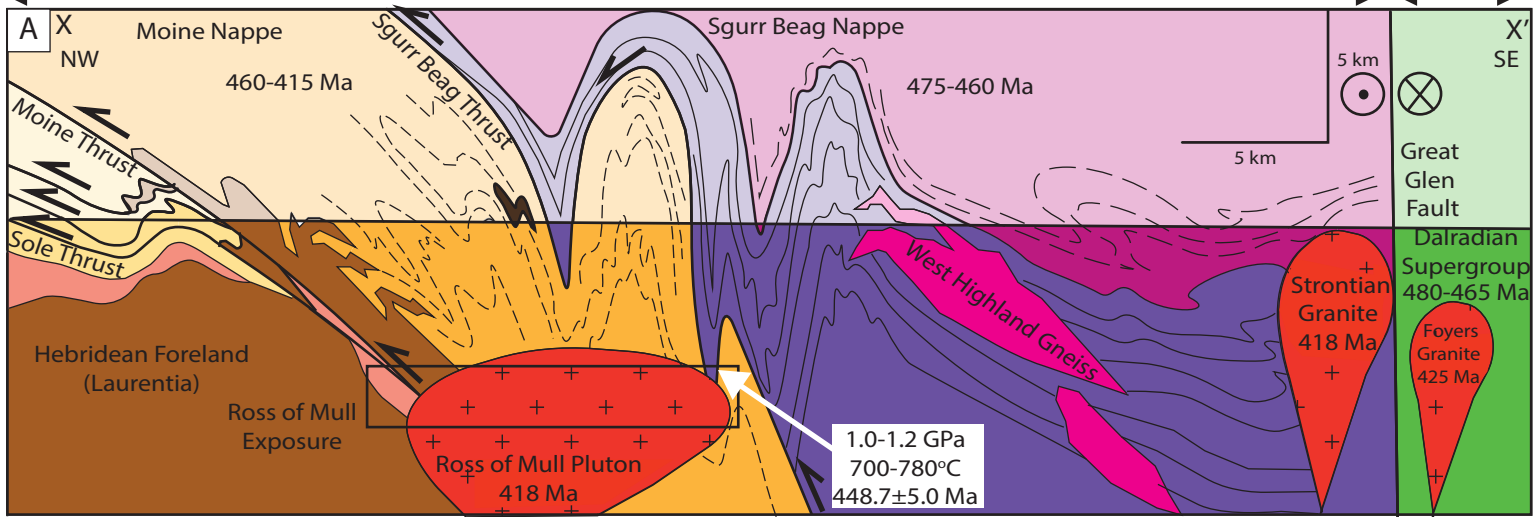
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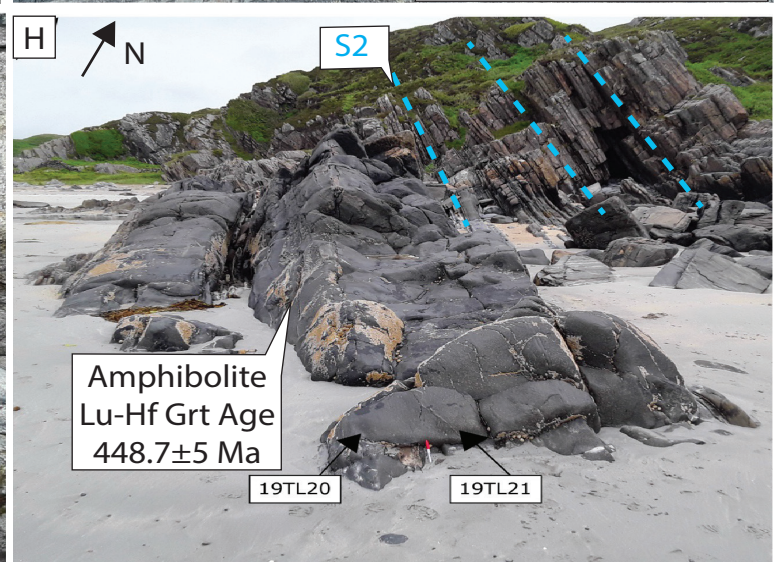
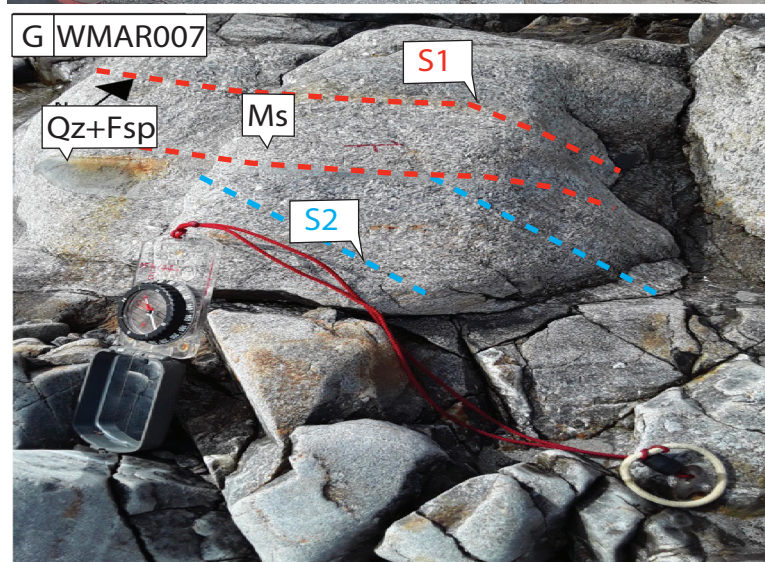
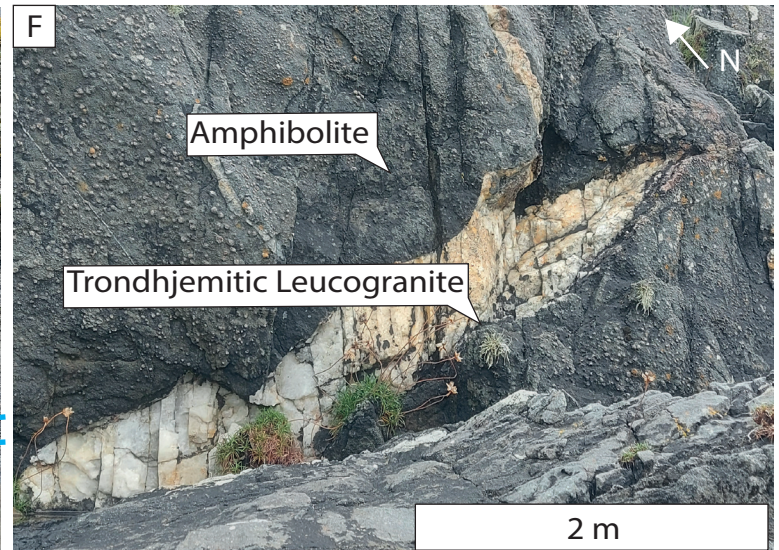
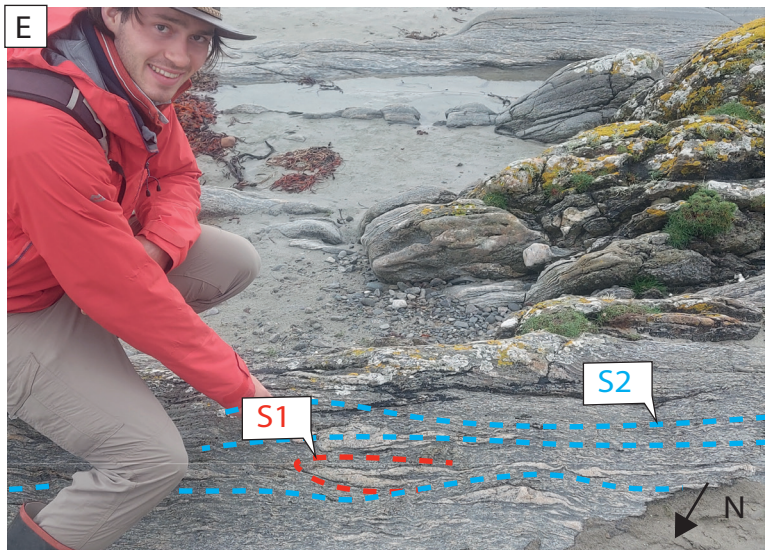
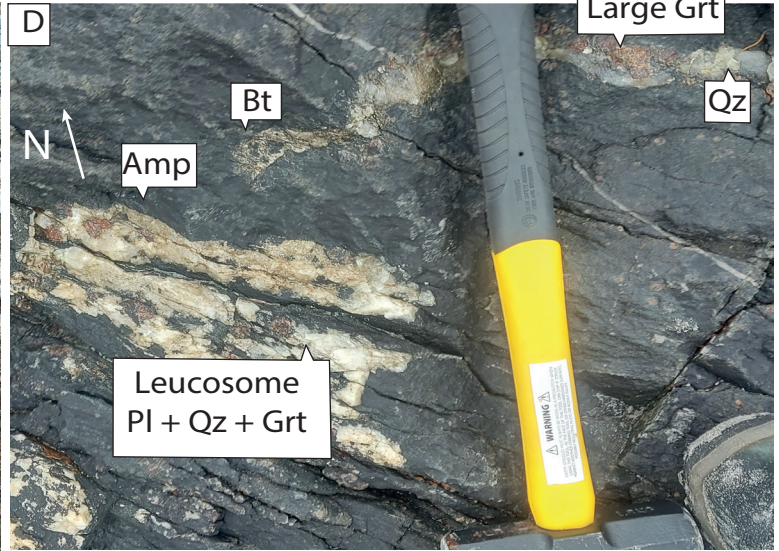
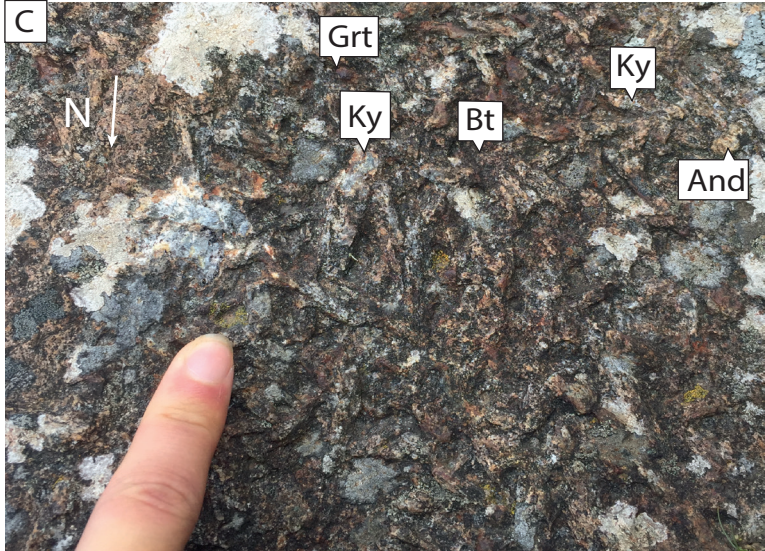
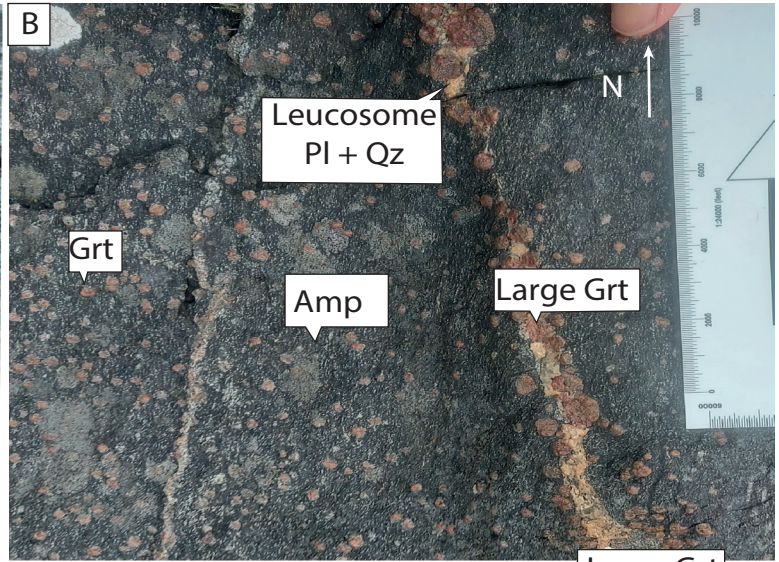
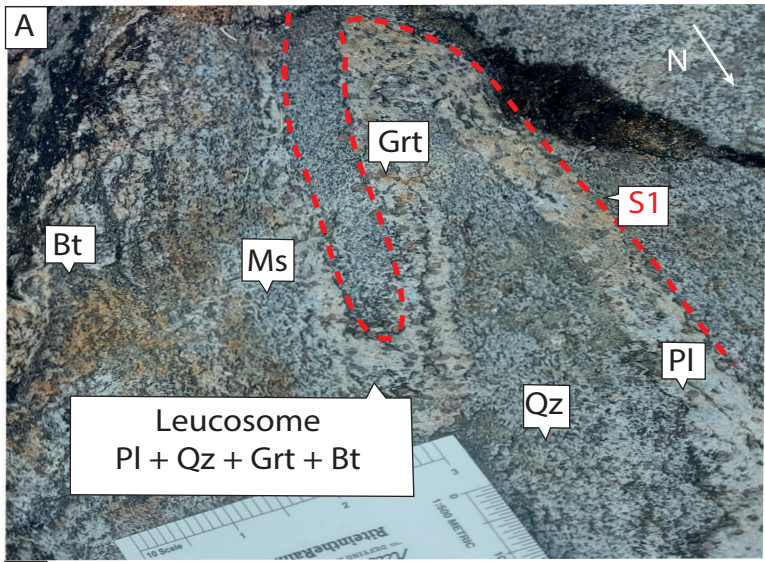
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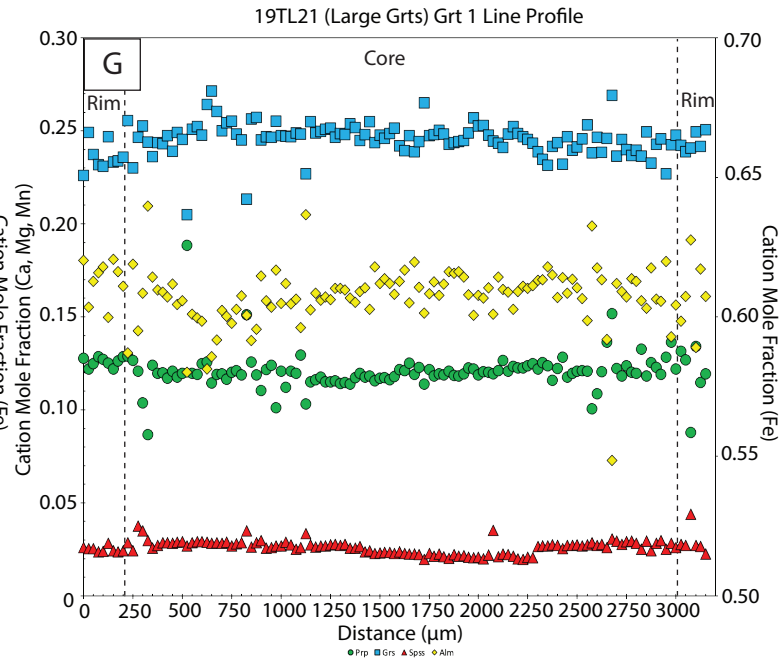
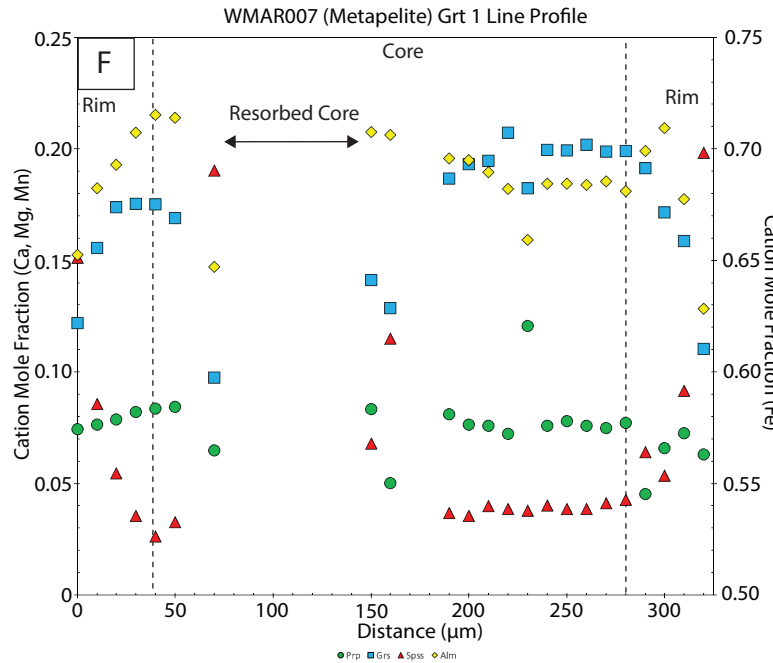
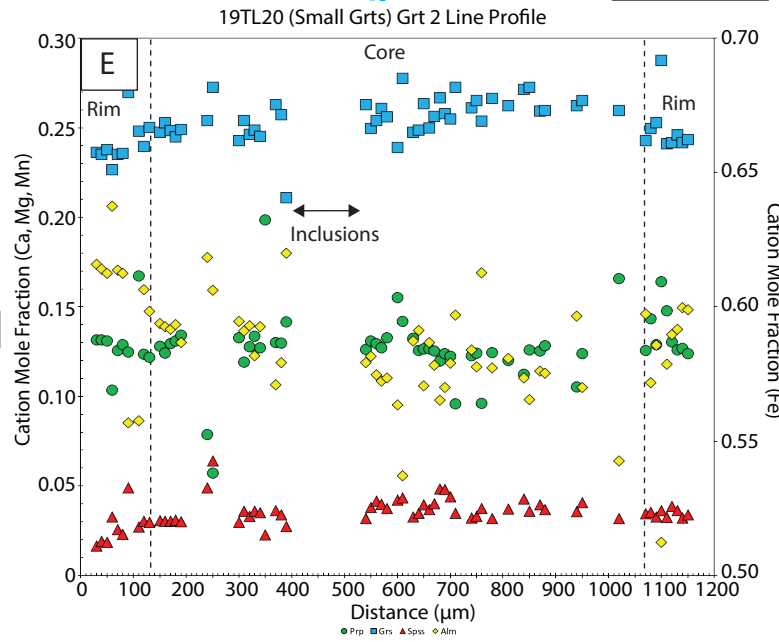
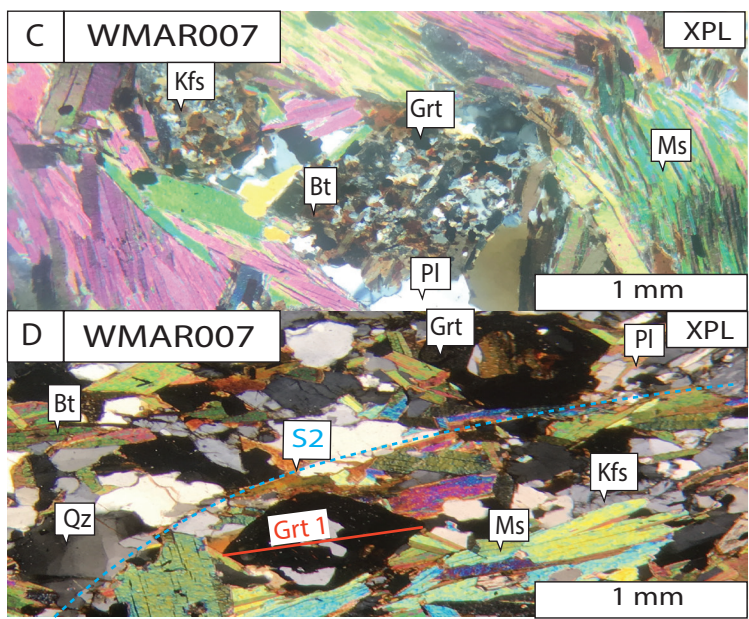
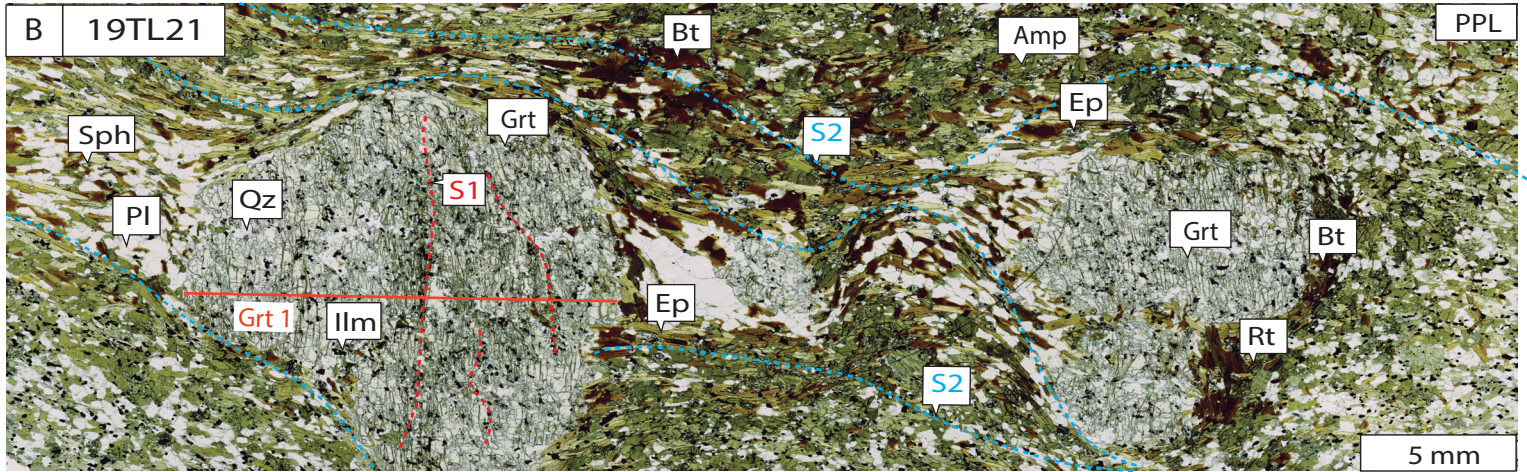
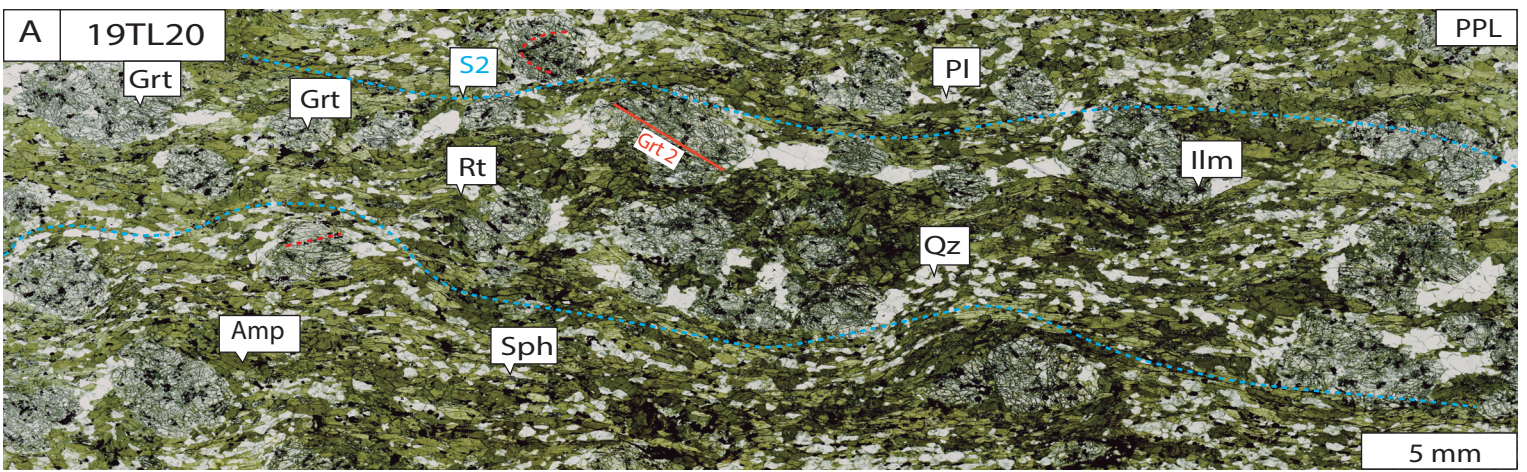
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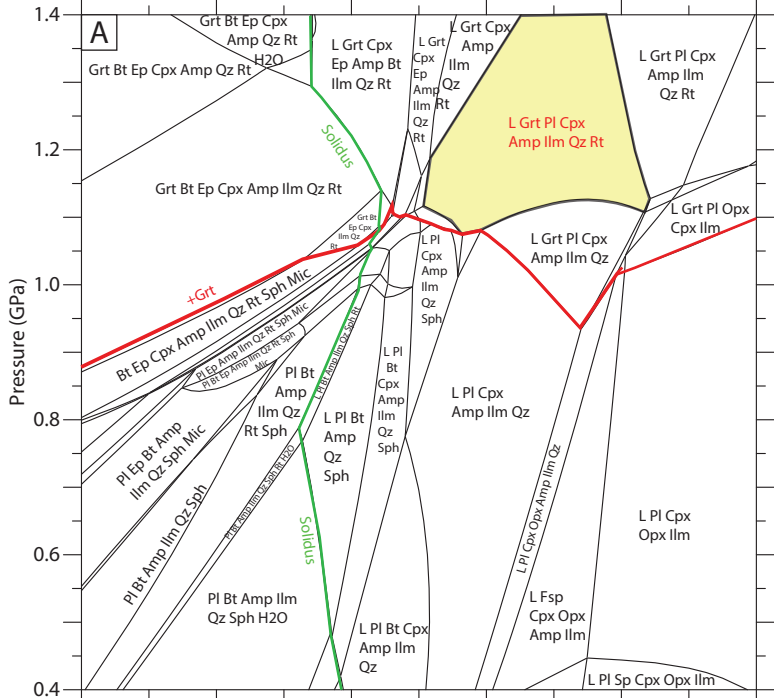




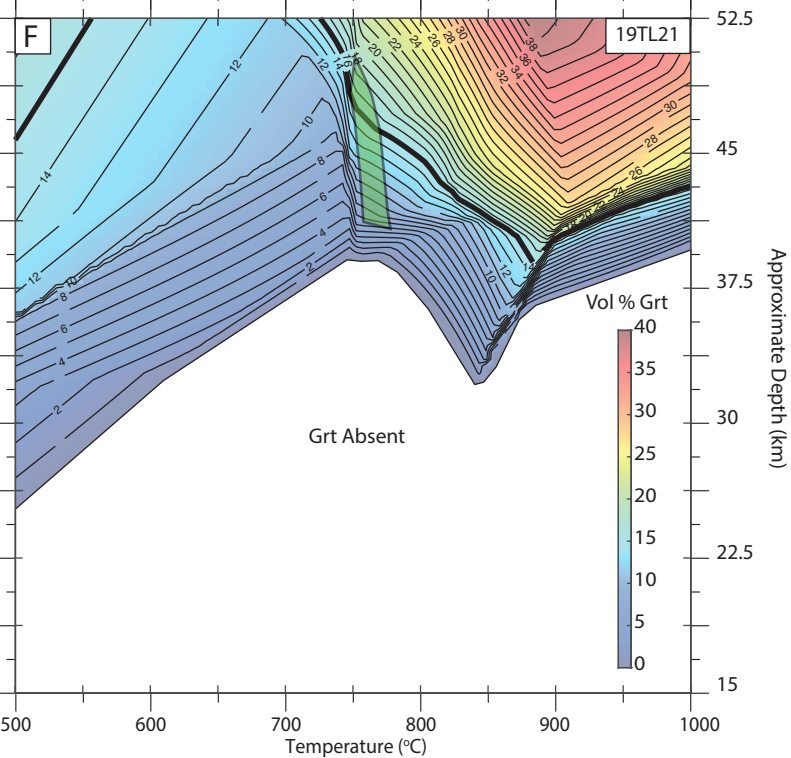
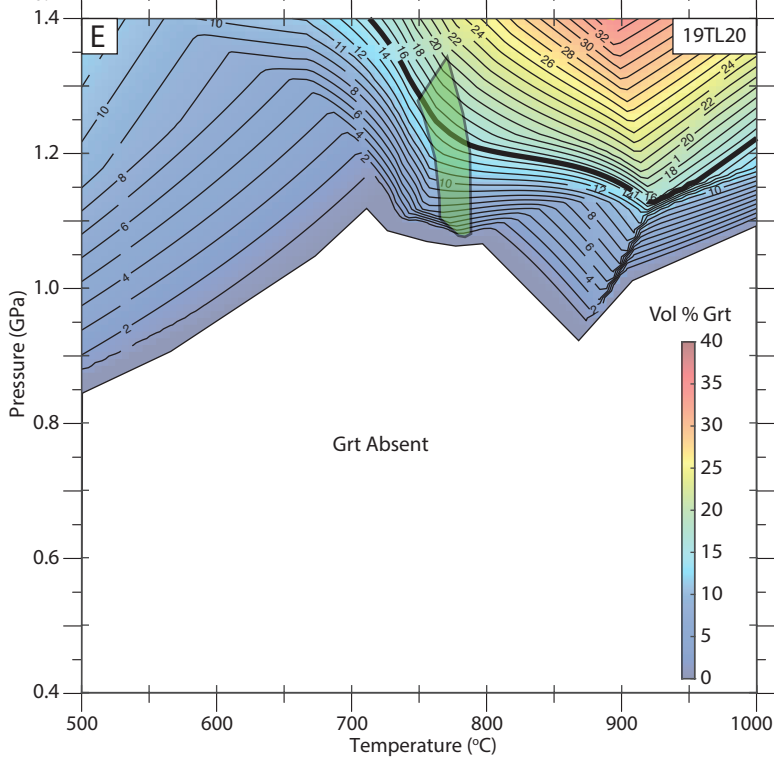
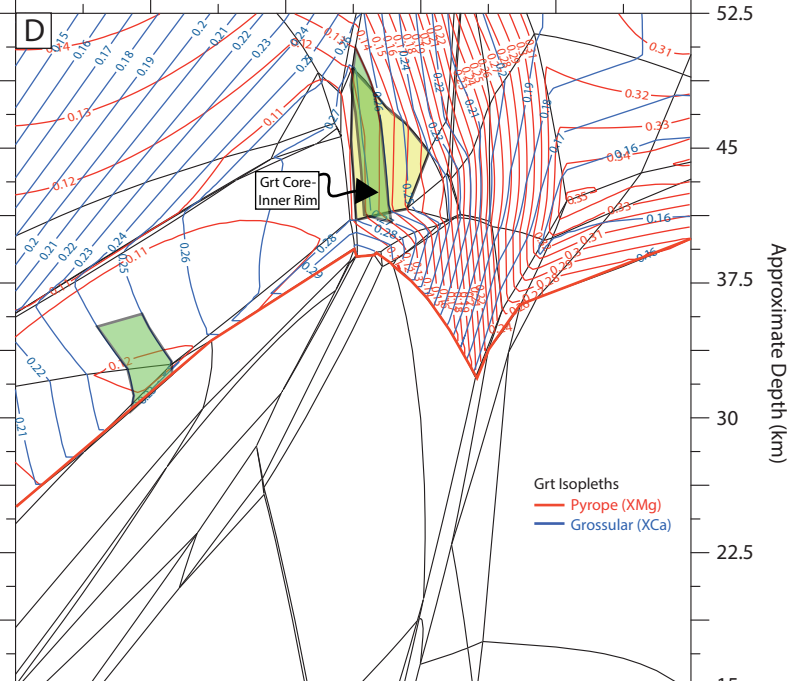
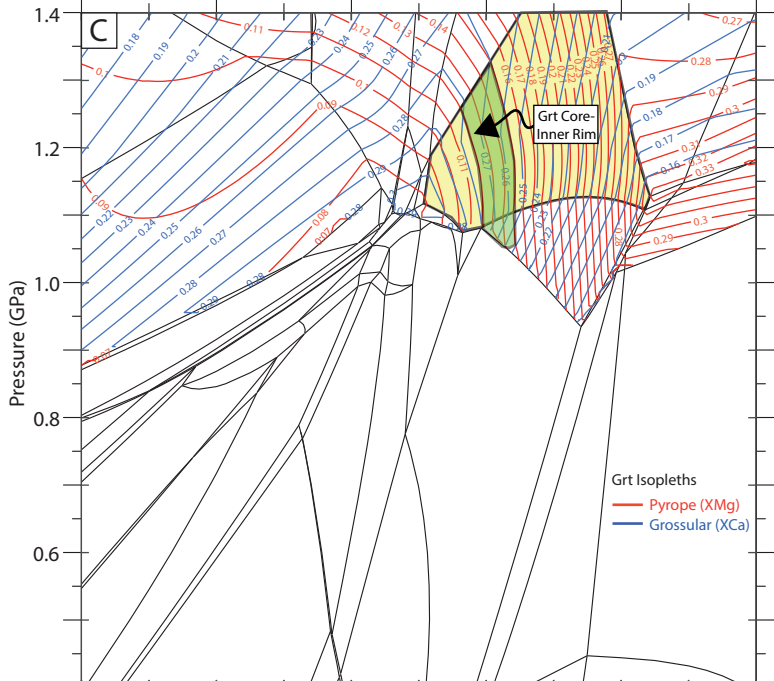
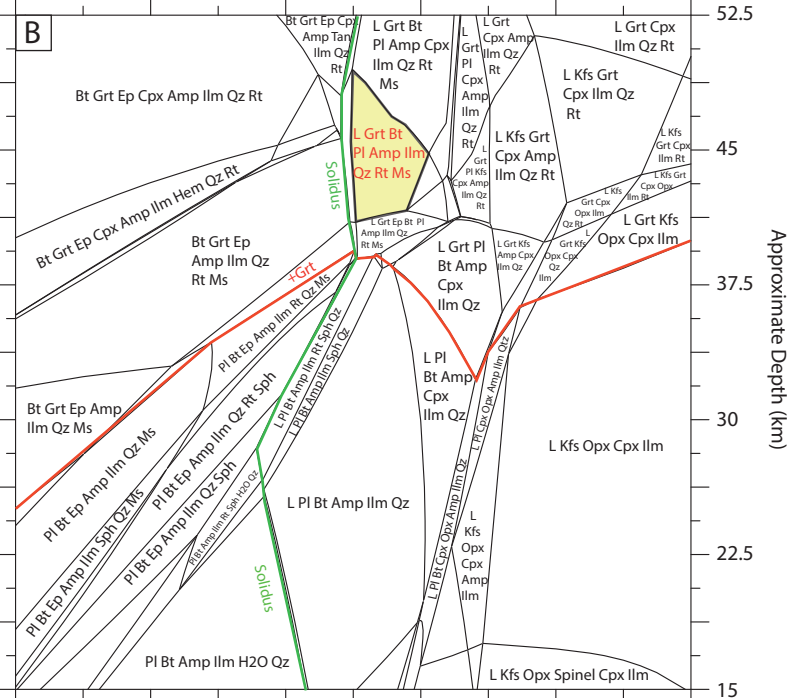




19TL20 Small Grt Amphibolite NCKFMSHTO
 SiO₂ TiO₂ Al₂O₃ FeO MgO CaO Na₂O K₂O O H₂O
 48.477 3.758 7.963 12.513 8.473 10.052 1.780 0.379 1.877 4.500

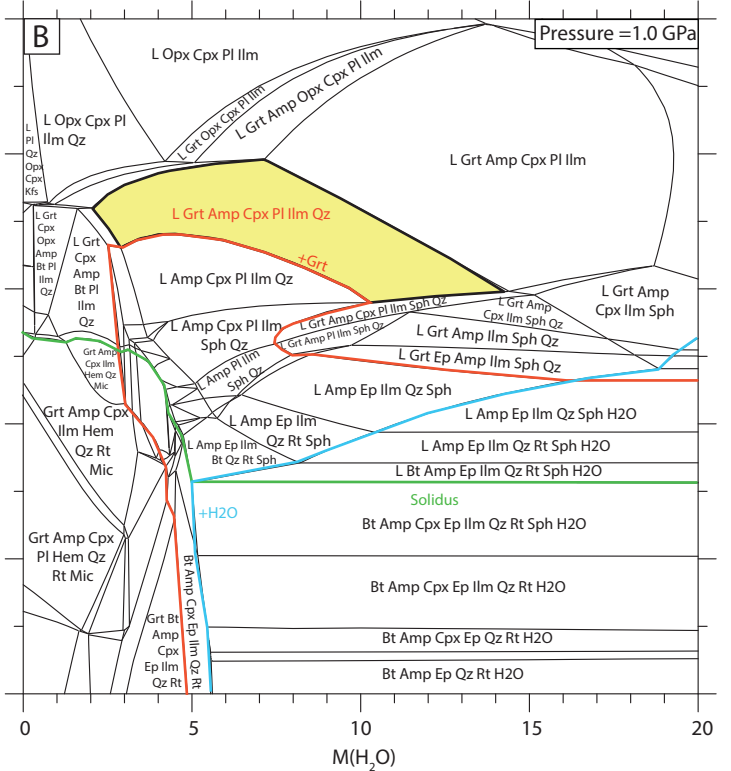
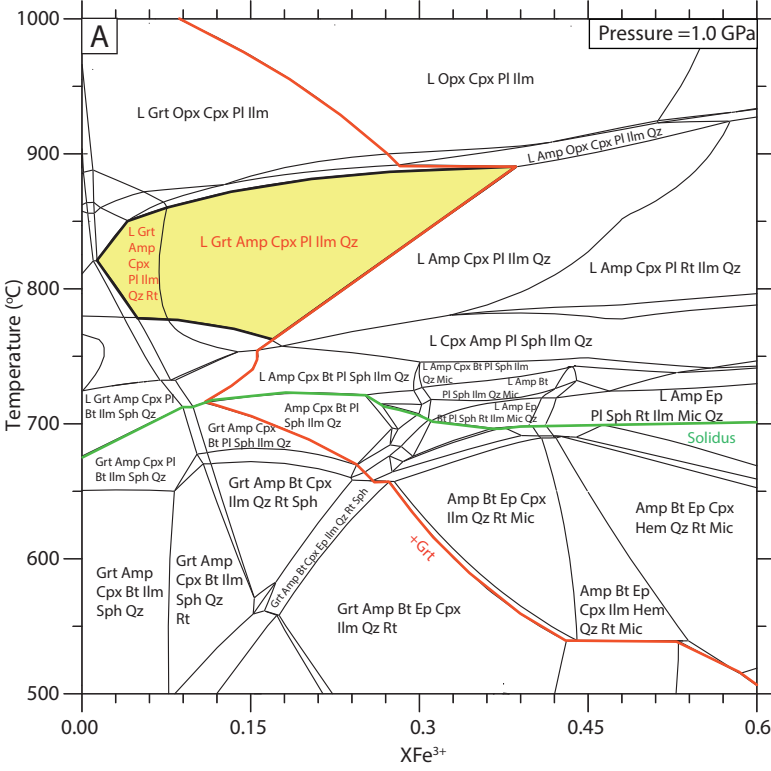


19TL21 Large Grt Amphibolite NCKFMSHTO
 SiO₂ TiO₂ Al₂O₃ FeO MgO CaO Na₂O K₂O O H₂O
 48.828 3.909 8.230 13.602 8.828 7.928 1.183 0.953 2.040 4.500



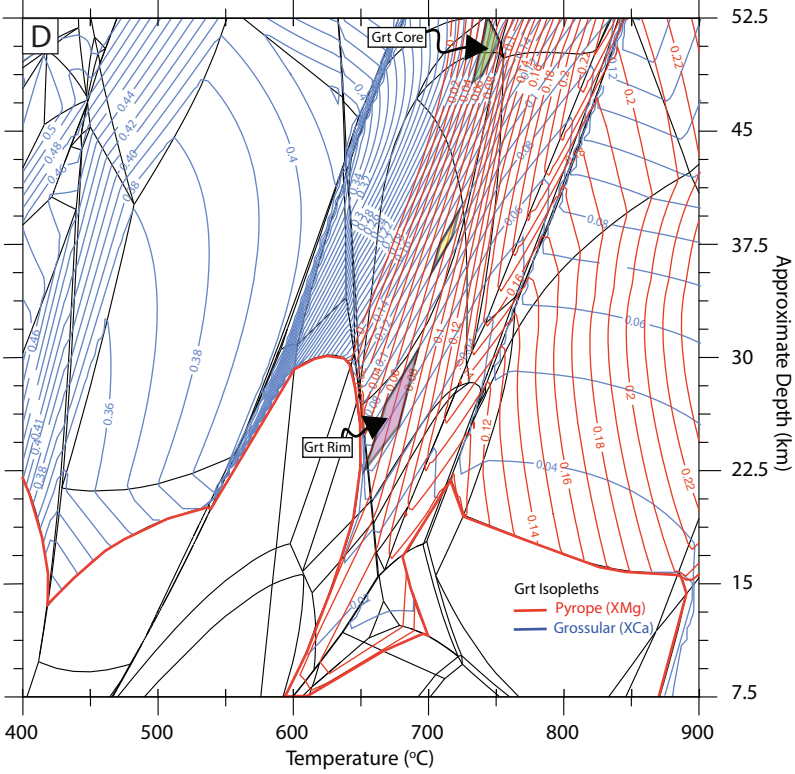
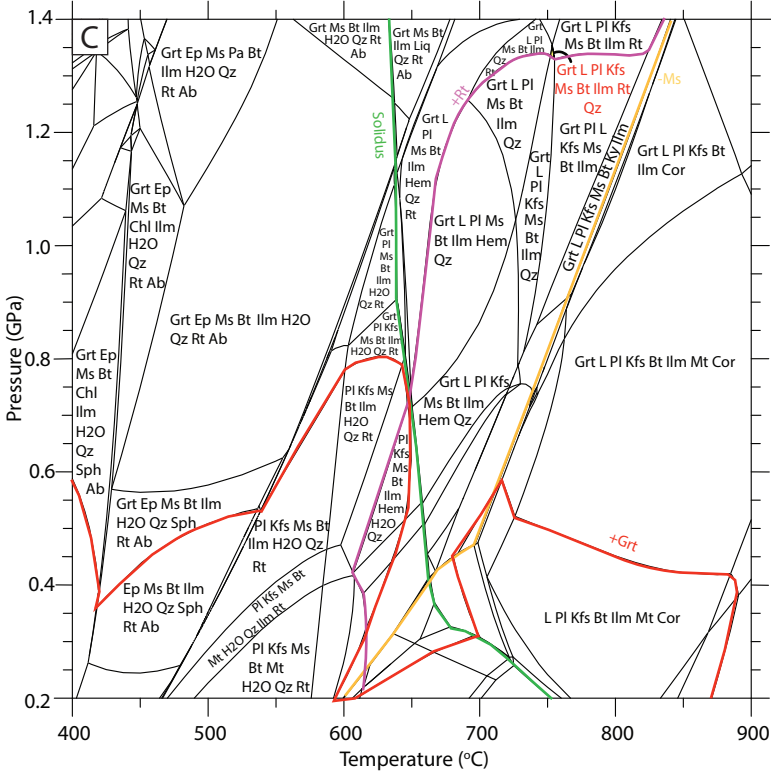
SiO₂ TiO₂ Al₂O₃ FeO MgO CaO Na₂O K₂O O H₂O
 49.566 3.842 8.140 12.795 8.663 10.278 1.820 0.388 0.006 4.500
 47.654 3.694 7.830 12.301 8.329 9.881 1.750 0.373 3.690 4.500

SiO₂ TiO₂ Al₂O₃ FeO MgO CaO Na₂O K₂O O H₂O
 50.857 3.942 8.354 13.128 8.889 10.545 1.867 0.398 1.969 0.050
 40.706 3.156 6.687 10.507 7.115 8.441 1.495 0.318 1.576 20.000

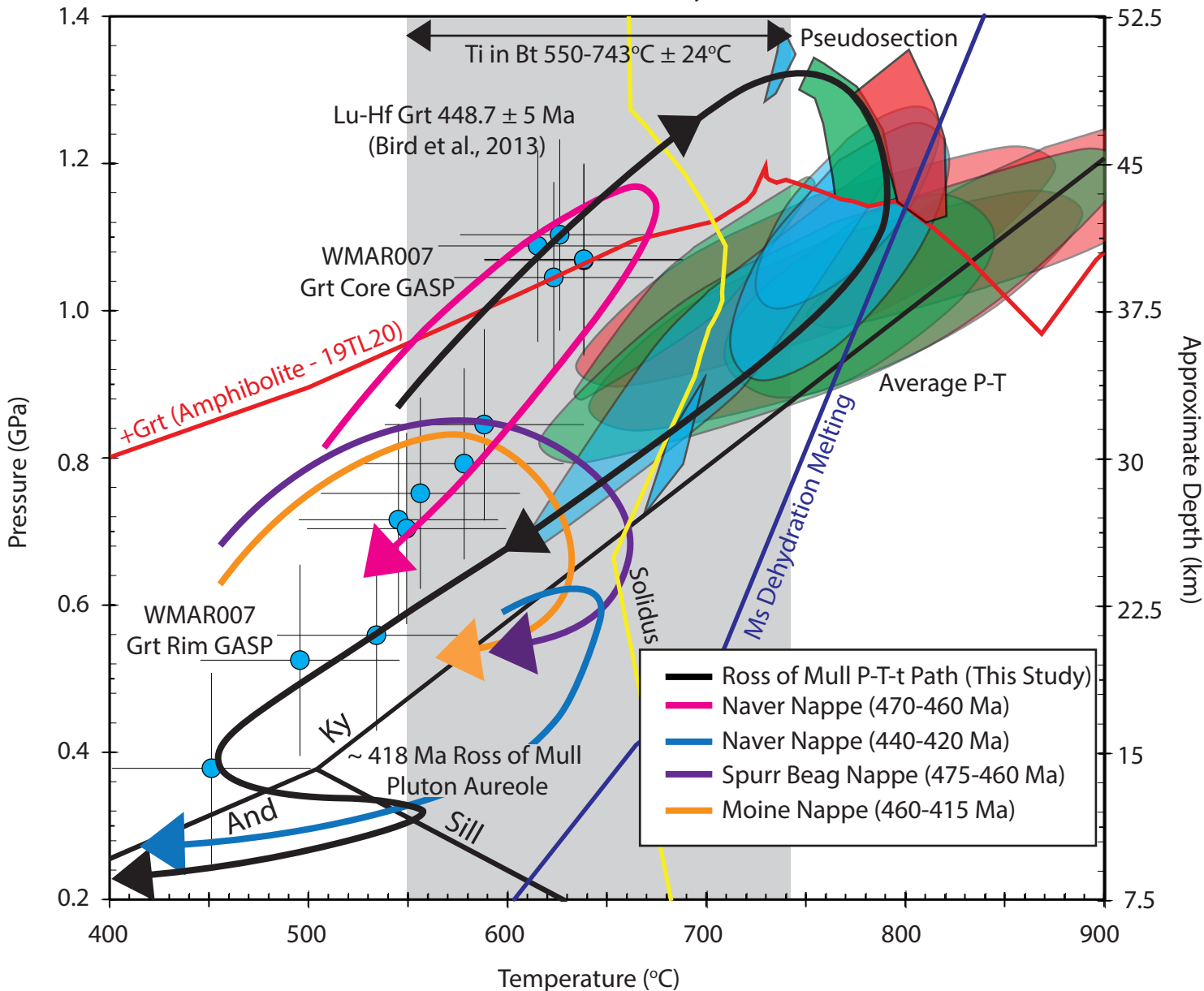


WMAR007 Grt-Ms-Kfs-Rt Schist MnNCKFMASHTO

SiO₂ TiO₂ Al₂O₃ FeO MnO MgO CaO Na₂O K₂O O H₂O
 54.619 0.945 14.885 4.894 0.098 2.391 0.999 3.797 4.269 0.734 12.370



Ross of Mull Moine Summary P-T-t Path



- Ross of Mull P-T-t Path (This Study)
- Naver Nappe (470-460 Ma)
- Naver Nappe (440-420 Ma)
- Spurr Beag Nappe (475-460 Ma)
- Moine Nappe (460-415 Ma)

— 19TL20 (Small Grt Amphibolite)
 — 19TL21 (Large Grt Amphibolite)
 — WMAR007 (Grt-Ky Schist)

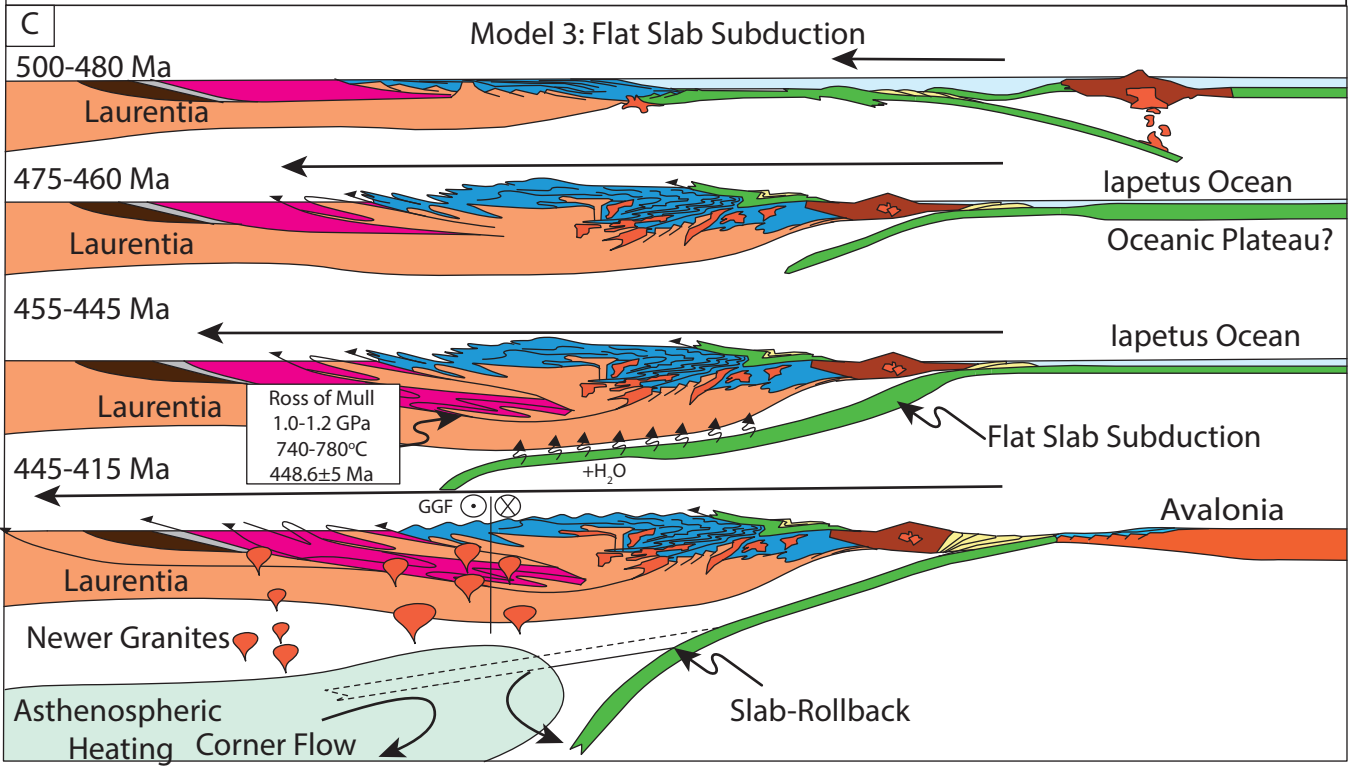
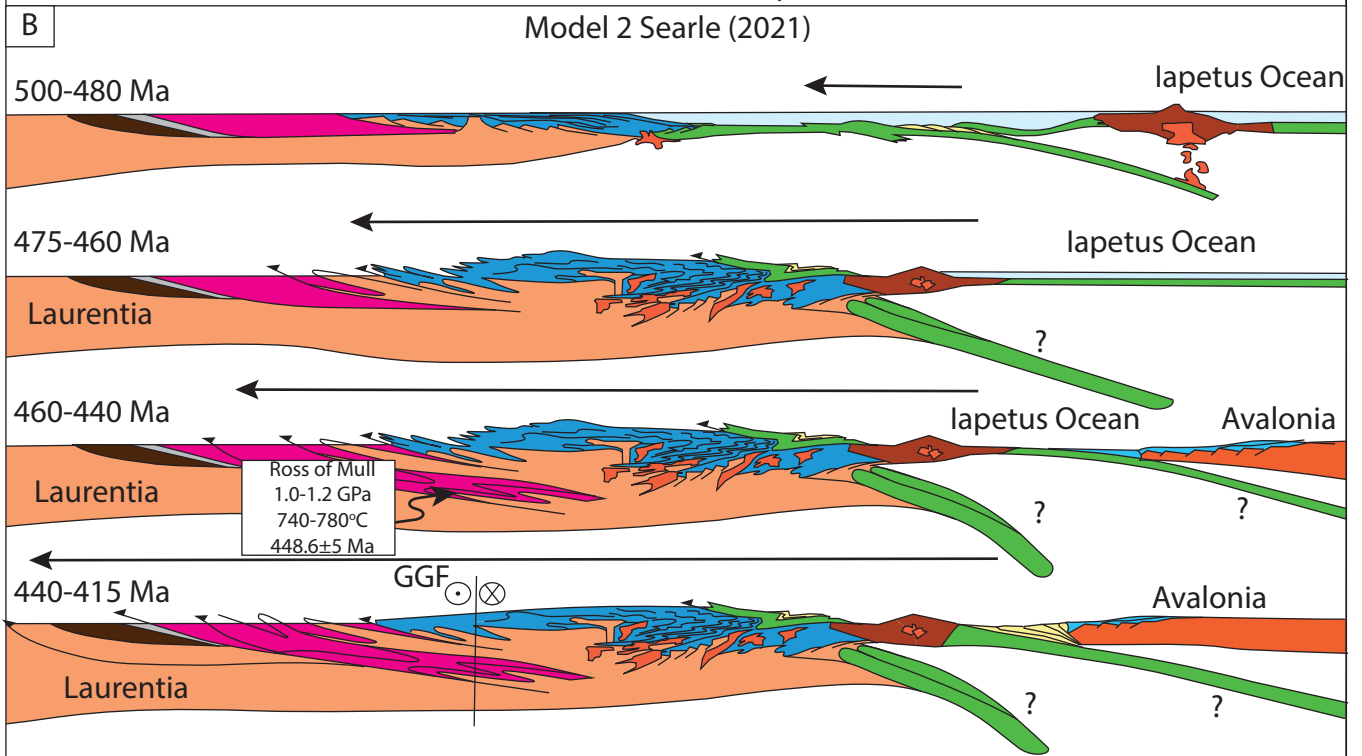
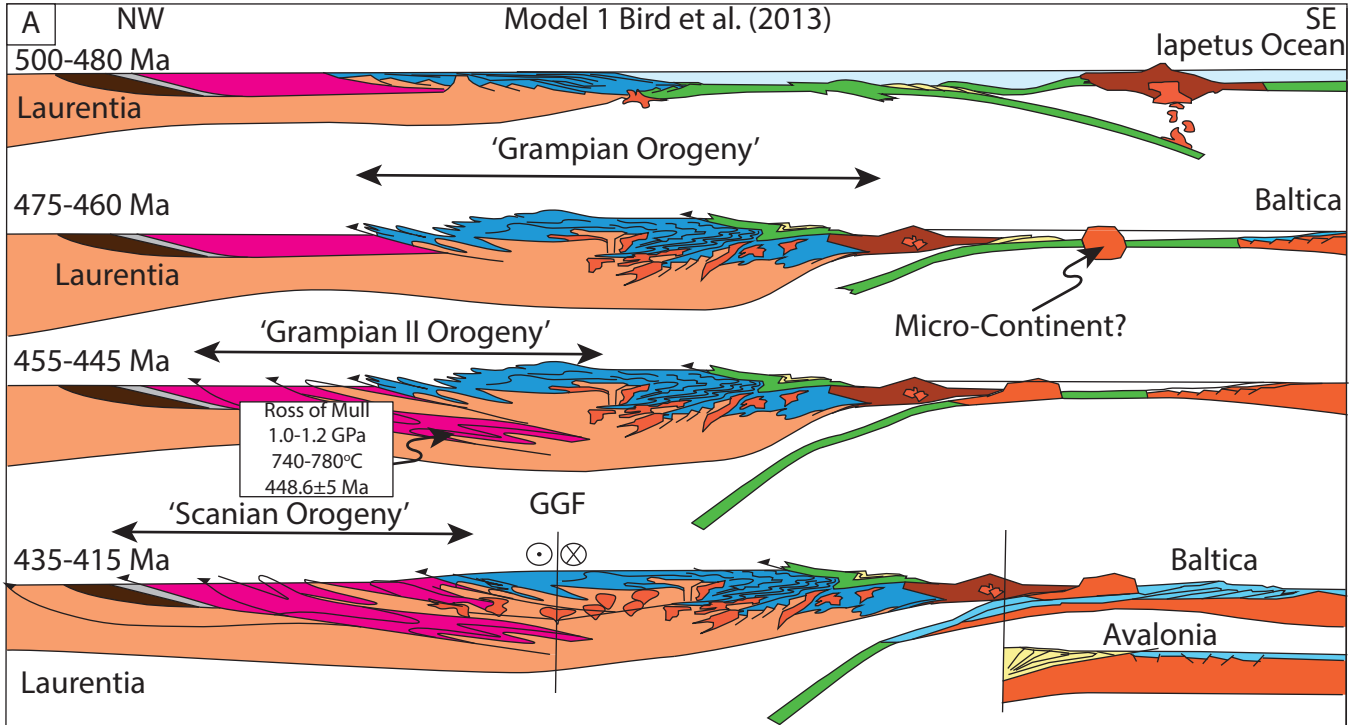


Table 1

Sample	Method	Temperature (°C)	Pressure (GPa)	Notes
19TL21	Amphibole-Plagioclase (H&B, 1994)	671 ± 15 (Edenite-Tremolite)	-	Based on matrix plagioclase (An = 0.28)
	Amphibole-Plagioclase (H&B, 1994)	688 ± 21 (Edenite-Richterite)	-	
	Ti-in-Biotite (Henry et al., 2005)	723 ± 12	-	Most biotite clusters 660-670°C, retrograde Based on Grt-Amp-Bt-Fsp-Rt-Ilm-Ep-Qz-Sph-H ₂ O assemblage Garnet isopleths (XPrp 0.13, XGrs 0.26), 15% garnet volume
	THERMOCALC (H&P, 2011)	777 ± 54	1.02 ± 0.11	
	Phase Diagram (Pseudosection)	760–780	1.08–1.30	
19TL20	Amphibole-Plagioclase (H&B, 1994)	665 ± 15 (Edenite-Tremolite)	-	Based on Grt-Amp-Fsp-Rt-Ilm-Qz-Sph-H ₂ O assemblage Garnet isopleths (XPrp 0.13, XGrs 0.26), 15% garnet volume
	Amphibole-Plagioclase (H&B, 1994)	717 ± 34 (Edenite-Richterite)	-	
	THERMOCALC (H&P, 2011)	755 ± 96	1.06 ± 0.13	
	Phase Diagram (Pseudosection)	790–810	1.05–1.30	
WMAR007	Ti-in-Biotite (Henry et al., 2005)	748 ± 12	-	Garnet core conditions Garnet in contact with biotite Based on Grt-Ms-Bt-Kfs-Fsp-Qz-Rt-Ilm-H ₂ O assemblage Garnet core isopleths (XPrp 0.08, XGrs 0.20)
	Garnet-Biotite (B, 1992) + GASP/GPMB Barometry (S, 1993)	638 ± 30	1.28 (GPMB_Mg), 1.04 (GPMB_Fe), 1.07 (GASP) ± 0.12	
	Garnet Rim (Retrograde)	496 ± 30	0.53 (GASP), 0.67 (GPMB_Mg), 0.51 (GPMB_Fe) ± 0.12	
	THERMOCALC (H&P, 2011)	750 ± 41	1.06 ± 0.14	
	Phase Diagram (Pseudosection)	720–750	1.2–1.3	