

1 ***Silurian-Devonian magmatism, mineralization, regional exhumation and brittle***
2 ***strike-slip deformation along the Loch Shin Line, NW Scotland.***

3
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15
16 ***ABSTRACT***

17 The Loch Shin Line (LSL) is a geological-geophysical lineament associated with a
18 zone of mantle-derived appinites, granites and strike-slip faulting that runs NW-SE
19 across the Moine Nappe, N Scotland. U-Pb zircon and Re-Os molybdenite dating of
20 the Loch Shin and Grudie plutons that lie immediately southwest of the NW-SE Loch
21 Shin-Strath Fleet fault system yield ca. 427-430Ma ages that overlap within error.
22 They also coincide with previously obtained U-Pb zircon ages for the Rogart pluton
23 which lies along strike to the southeast. Field and microstructural observations
24 confirm the similarity and contemporaneous nature of the plutons and associated
25 sulphide mineralisation. Fluid inclusion analyses place further constraints on the P-
26 T-X conditions during regional late Caledonian exhumation of the Moine Nappe.
27 Synchronous to slightly younger brittle dextral strike slip faulting along the WNW-
28 ESE Loch Shin-Strath Fleet Fault System was likely antithetic to sinistral movements
29 along the nearby Great Glen Fault Zone. Our findings support the hypothesis that the
30 LSL acted as a deep crustal channelway controlling the ascent and emplacement of
31 Silurian magmas into the overlying Moine Nappe. We propose that this deep

32 structure corresponds to the southeastern continuation of the Precambrian-age
33 Laxford Front shear zone in the buried Lewisian autochthon.

34

35 Supplementary material: [Field photographs, photomicrographs and fluid inclusion
36 information] is available at www.geolsoc.org.uk/SUP0000

37

38 ***INTRODUCTION***

39 Orogenic belts worldwide are characterized by interlinked systems of thrust, strike
40 slip and extensional faults and, at deeper crustal levels, by shear zones that
41 collectively accommodate crustal deformation in broad continental deformation
42 zones during plate collision (i.e. 'block and flake tectonics'; Dewey *et al.* 1986). The
43 location, geometry and persistence of faults and shear zones in such regions are
44 known to be influenced by the reactivation of crustal-scale pre-existing structures
45 (Sutton & Watson 1986; Holdsworth *et al.* 1997, 2001). These same structures are
46 also known to act as channelways that control the upward migration and
47 emplacement of hydrous mineralizing fluids and magmas (e.g. O'Driscoll 1986;
48 Hutton 1988a; Jacques & Reavy 1994; Richards 2013). This coincidence of
49 geological processes has greatly assisted in the analysis of orogenic deformation
50 histories worldwide since dating of igneous intrusions and/or mineralization events
51 using geochronology can also be used to constrain the absolute ages of associated
52 deformation events in the adjacent wall rocks (e.g. Paterson & Tobisch 1988;
53 Schofield & D'Lemos 1998; Rosenberg 2004).

54 Integrated structural and geochronological studies of deformed igneous
55 intrusions have played a key role in constraining the timing of events within the
56 Early Palaeozoic Caledonian orogeny in Scotland (Fig. 1a). Following Ordovician arc
57 continent collision (the Grampian event), the final closure of Iapetus involved the
58 oblique collisions of three palaeo-continents: Laurentia, Baltic and Avalonia during
59 the mid- to late Silurian (e.g. Soper *et al.* 1992; Torsvik *et al.* 1996). In NW Scotland,
60 regional deformation occurred due to the sinistral oblique Scandian collision of
61 Baltica with Laurentia. Crustal thickening here was overlapped and followed by
62 major sinistral displacements along orogen-parallel strike-slip faults such as the
63 Great Glen Fault Zone (GGFZ; Fig 1a) heralding a transition from a regime of
64 sinistral transpression to transtension (Dewey & Strachan 2003 and references
65 therein). Igneous activity and associated mineralization related to slab breakoff was
66 associated with this transition so that earlier granites were syn-tectonically
67 emplaced along Scandian thrusts (e.g. Naver Thrust, see Holdsworth & Strachan
68 1988; Kinny *et al.* 2003; Goodenough *et al.* 2011; Kocks *et al.* 2013), whilst later,
69 volumetrically larger volumes of melt were emplaced along steeply-dipping strike-
70 slip or normal faults (e.g. GGFZ; Hutton 1988b; Hutton & McErlean 1991; Jacques &
71 Reavy, 1994; Stewart *et al.* 2001). In many cases the controlling faults or shear
72 zones are exposed at the present-day surface, but others are more enigmatic
73 features. As illustrated by Jacques & Reavy (1994) they are commonly inferred
74 'buried' structures based on geological, geophysical or geochemical alignments that
75 define regional scale transverse lineaments that run generally at high angles to the
76 orogenic strike. One of these NW-SE features, the Loch Shin Line (LSL) – first

77 defined by Watson (1984) – is associated with an anomalous zone of mantle-derived
78 appinites, granites and brittle faulting in the Moine Nappe SE of the Moine Thrust on
79 the N side of the Assynt Culmination (Fig. 1a, b). The LSL follows a strong NW-SE
80 gravity gradient that defines the NE margin of a strong negative anomaly centred on
81 the Grudie Granite (Figs 1b, see Leslie *et al.* 2010 and references therein). Watson
82 (1984) suggested that the LSL corresponds to the presence of a Precambrian shear
83 zone in the Lewisian autochthon underlying the Moine Nappe and that this shear
84 zone has controlled the siting and ascent of magmas and associated mineralization
85 during the Silurian. The dextral faulting that follows the trend of the LSL defines the
86 Loch Shin, Strath Fleet and Dornoch Firth fault systems (Fig. 2a; Strachan &
87 Holdsworth 1988) which are thought to be part of a regional fault set antithetic to
88 the regional sinistral movements along the GGFZ (see Johnson & Frost 1977; Watson
89 1984). The Rogart igneous complex (Fig. 1a; Soper 1963), a large composite
90 igneous intrusion of mantle derivation that lies on the NE margin of the LSL, is
91 bounded to the SW by the Strath Fleet Fault. Kocks *et al.* (2013) have shown that
92 emplacement of the central pluton – dated at 425 ± 1.5 Ma using U-Pb (TIMS) zircon -
93 was likely controlled by dextral motions along the LSL. These authors used this
94 evidence to date the switch from sinistral transpression with thrusting to
95 transtension with regional strike slip faulting at ca. 425 Ma.

96 The present paper re-examines this hypothesis in the region of Loch Shin
97 where two plutons hosted in Moine and Lewisian country rocks are notably
98 associated with molybdenite mineralization (Gallagher & Smith 1975): the Loch
99 Shin and Grudie granites (Figs 1 & 2). Field observations and microstructural

100 studies are used to constrain the geometry, kinematics and relative ages of
101 deformation in the plutons and country rocks, whilst U-Pb zircon and Re-Os
102 molybdenite geochronology are used to date both pluton emplacement and the
103 spatially associated mineralization. Fluid inclusion studies are used to further
104 constrain the P-T-X conditions during deformation and igneous emplacement and
105 assess the relationships between regional structures and fluid flow.

106

107 ***GEOLOGICAL SETTING***

108 The Loch Shin area is underlain by variably deformed metasedimentary rocks of the
109 Morar Group, part of the Neoproterozoic Moine Supergroup in NW Scotland (Figs 1,
110 2; Holdsworth *et al.* 1994; Strachan *et al.* 2010). To the northwest, the Moine Nappe
111 is bounded by the underlying Moine Thrust and Moine Thrust Zone, whilst to the
112 north and east it is overlain by the Naver Thrust which carries the Loch Coire
113 Migmatite Complex (Fig. 1a; Kocks *et al.* 2013). Zircon U-Pb geochronology shows
114 that the migmatite complex formed during the Ordovician Grampian event ca 470-
115 460Ma (Kinny *et al.* 1999). This was followed by generally top-to-the-NW Scandian
116 ductile thrusting with early displacement along the Naver Thrust, then later thrusts
117 propagating progressively towards the Caledonian foreland ending with the
118 development of the Moine Thrust Zone (Barr *et al.* 1986; Johnson & Strachan 2006;
119 Alsop *et al.* 2010; Leslie *et al.* 2010). Zircon U-Pb dating of various syn-kinematic
120 igneous intrusions constrains thrust movements to ca. 435-425Ma (Kinny *et al.*
121 2003; Kocks *et al.* 2006; Goodenough *et al.* 2011). The broad arcuate swing of the
122 regional foliation and ductile thrusts within the Moine and Naver nappes (Fig. 1a,

123 2a) is attributed to the development of the Cassley structural culmination and
124 regional-scale flexuring in the rocks overlying the Assynt Culmination (Elliott &
125 Johnson 1980; Butler & Coward 1984; Leslie *et al.* 2010).

126 The Loch Shin and Grudie granites are hosted in Morar Group rocks locally
127 interleaved with antiformal isoclinal infolds of their underlying Lewisianoid
128 basement (Read *et al.* 1926; Gallagher & Smith 1975; Strachan & Holdsworth 1988;
129 Leslie *et al.* 2010). The Moine rocks are unmigmatized psammities interlayered with
130 subordinate semipelitic and pelitic horizons preserving rare sedimentary structures
131 such as cross-lamination and grading in areas of low tectonic strain. The
132 Lewisianoid rocks are lithologically diverse and include hornblendic and
133 quartzofeldspathic gneisses, amphibolites and subordinate units of ultramafic
134 hornblendite, together with thin strips of metasedimentary schist and marble (e.g.
135 Airde of Shin, Fig. 2a; see Strachan & Holdsworth 1988 and references therein).
136 Individual Moine-Lewisianoid boundaries – where exposed - are marked either by
137 the development of local basement conglomerates or by the development of mica-
138 rich ‘tectonic schists’ (Peacock 1975; Strachan & Holdsworth 1988).

139 The dominant structures in the Moine and Lewisianoid rocks are tight to
140 isoclinal D2 folds that carry an axial planar S2 crenulation fabric of an earlier
141 bedding parallel schistosity (S1). The main foliation is therefore a composite
142 S0/S1/S2 fabric which carries an ESE- to SE-plunging mineral extension lineation L2
143 (Strachan & Holdsworth 1988). This lineation is interpreted to lie parallel to the
144 regional direction of top-to-the-NW tectonic transport during Scandian thrusting
145 (e.g. Barr *et al.* 1986; Strachan *et al.* 2010). Associated regional metamorphism

146 during D2 in the Loch Shin area was within the low to mid-amphibolite facies (Soper
147 & Brown 1971; Strachan & Holdsworth 1988).

148 The Moine and Lewisianoid rocks around Lairg and Loch Shin are cut by a
149 number of granitic bodies, which include (from largest to smallest): the Grudie,
150 Claonel and Loch Shin intrusions (Fig. 2; Gallagher & Smith 1975), together with
151 numerous small associated sheets and plugs of similar composition. These fall into
152 two distinct groups: early foliated granodiorites (e.g. Claonel), thought to be directly
153 equivalent to parts of the Rogart igneous complex, and supposedly later, generally
154 unfoliated intrusions of pink adamellite including the Grudie and Loch Shin bodies.
155 The trace of the LSL is also marked by a concentration of small plugs and pipe-like
156 bodies of intermediate to ultramafic appinites known as the Ach'uaire hybrids (Fig.
157 1b; Read *et al.* 1925; Watson 1984). These also occur as comagmatic enclaves within
158 the ca. 425Ma central granodiorite of the Rogart igneous complex (Fowler *et al.*
159 2001; Kocks *et al.* 2013). Appinites are widely associated with late Caledonian
160 plutons throughout the Scottish Highlands and point to a significant mantle
161 contribution to this magmatism (e.g. see Fowler & Henney 1996; Fowler *et al.* 2008).

162 Regional mapping, stream sediment sampling and analysis of shallow
163 borehole cores in the Loch Shin-Grudie area has shown that low grade molybdenite
164 mineralization is associated with pyrite in thin post-foliation quartz veins cutting
165 both country rock and granites; subordinate chalcopyrite, fluorite, galena, barite and
166 sphalerite also occur (Gallagher & Smith 1975). This mineralization is spatially
167 associated with the granites, but Gallagher & Smith (*op cit*) suggest that it may also

168 have been significantly influenced by regional structures in the surrounding wall
169 rocks.

170 Between Loch Shin and the Moray Firth to the east, the Moine and Lewisian
171 rocks are cut by at least three major, sub-vertical brittle faults: the Loch Shin, Strath
172 Fleet and Dornoch Firth fault zones (Fig. 1a; Read *et al.* 1925, 1926; Strachan &
173 Holdsworth 1988; Kocks *et al.* 2013). Exposure of these fault zones is generally very
174 poor with only the Strath Fleet Fault previously studied in any detail (Soper 1963).
175 A series of NW-SE-trending steeply dipping crush zones were recognized that
176 overprint Moine country rocks, the Rogart igneous complex and unconformably
177 overlying Devonian basal conglomerates (middle Old Red Sandstone). There is
178 evidence for multiple fault movements, with cataclastic fault rocks included as clasts
179 within overlying Devonian conglomerates and minor intrusions that cut brittle fault
180 rocks whilst also being overprinted by later faulting (Soper 1963). However, there is
181 little published evidence to support the dextral shear sense inferred by many
182 authors along these NW-SE faults (e.g. Johnson & Frost 1977; Watson 1984),
183 although apparent regional offsets of regional boundaries in the Moine Nappe are
184 consistent with right-lateral movements along the Strath Fleet and Dornoch Firth
185 Faults (Fig 1a; Soper 1963; Strachan & Holdsworth 1988). A presumably late
186 (?Devonian) NE-side-down movement is also inferred for the Strath Fleet Fault
187 based on the preservation of Devonian conglomerates in an elongate NW-SE-
188 trending outlier that follows the Strath Fleet Valley (e.g. see Kocks *et al.* 2013).

189 There are no published structural studies of any of the igneous bodies that
190 occur close to Loch Shin due to the poor levels of exposure (<1%). The Grudie

191 pluton is inferred to cross-cut all ductile fabrics and geological boundaries in the
192 Moine and Lewisian rocks based on the obviously discordant nature of the mapped
193 boundaries and the absence of an internal foliation (Fig. 2b; Gallagher & Smith
194 1975).

195 The present study focusses on two key areas of exposure: a ca 1 km long
196 sporadically continuous section through Moine rocks and part of the Loch Shin
197 Granite on the southwest shore of Loch Shin; and isolated exposures of Grudie
198 Granite exposed in road cuts related to the Meall a' Gruididh wind farm
199 development (Fig. 2b).

200

201 ***LOCH SHIN GRANITE***

202 Good quality water-washed exposures of Moine country rocks, the Loch Shin
203 Granite and associated mineral veins occur along the SW shore of Loch Shin
204 between NC 5650 0590 and NC 5625 0668 (Fig. 2b; see also Appendix A,
205 Supplementary Material). Isolated poor quality exposures also occur in inland areas
206 and stream sections, notably along the Allt a' Chlaonaidh (see Gallagher & Smith
207 1975, fig. 3).

208 Moine country rocks are exposed south of the Loch Shin granite between NC
209 5650 0590 and NC 0623 5638 and, north of the granite, between NC 5625 0668 and
210 NC 5587 0766. They are mostly fine to medium grained grey mica psammities with a
211 flaggy foliation and mm-scale compositional banding. Isolated layers of grey-brown
212 weathering semipelite-pelite are sparsely developed in layers up to 20 cm thick. In
213 thin section, the psammities comprise quartz, plagioclase, K feldspar, green biotite

214 and accessory phases (mineralization, garnet, epidote). Quartz and feldspar
215 uniformly display sub-equant polygonal to cusped-lobate textures typical of
216 amphibolite facies conditions (e.g. see Holdsworth & Grant 1990), with the main
217 banding parallel fabric (S0/S1/S2) being defined primarily by aligned biotite grains.
218 The foliation and associated mineral lineations are locally variable in orientation –
219 possibly due to the local effects of late brittle folding and faulting (see below) - but
220 the majority strike NE-SW with moderate SE dips (Figs 2b, 3a). The associated fine
221 mineral lineations, interpreted here as L2, plunge mainly ESE (Fig. 3a) typical of this
222 part of the Moine Nappe in Sutherland (e.g. Strachan & Holdsworth 1988).

223 The ductile foliation in the Moine rocks is cross cut at low angles by generally
224 NE-SW trending, moderately SE dipping pink granite and granite pegmatite sheets
225 up to 1 m thick (e.g. Fig. 4b). These are unfoliated and are compositionally very
226 similar to the Loch Shin granite.

227 The contacts of the Loch Shin granite are not exposed but are inferred to
228 trend NE-SW and dip to the SE based on the orientation of the exposed granite-
229 pegmatite veins (Figs 2b, 3b). The pink granite is typically fine to medium grained
230 and is unfoliated, lacking both magmatic and solid-state ductile fabrics. In thin
231 section it typically comprises weakly sericitised plagioclase, perthitic K-feldspar
232 (occasionally as phenocrysts), quartz, biotite (often altered to secondary chlorite)
233 and iron oxide (?magnetite). The granite is homogeneous in terms of both
234 composition and grain size and no internal contacts were seen. No magmatic-state
235 fabric is present, nor is there any evidence of crystal plasticity other than low-
236 temperature features spatially associated with fractures.

237 The granite is cut by irregular sets of quartz-pyrite-chalcopyrite veins with
238 rare molybdenite. These veins have no dominant orientation. However, at NC 5630
239 0660, a large sub-vertical SSE-NNW trending quartz-pyrite-sphalerite-chalcopyrite-
240 galena vein up to 1 m thick can be traced for over 10 metres along strike. All veins
241 lack ductile deformation fabrics, but are cross-cut by brittle faults and low
242 temperature cataclasis (e.g. Fig. 4a). Rice & Cope (1973) and Gallagher & Smith
243 (1975) give further details of veins and mineralization found in the surrounding
244 country rocks and report the additional presence of minor covellite, barytes and
245 fluorspar. Rare, late veins of zeolite <1 mm thick were observed cross-cutting fault-
246 related breccias in Moine host rocks (e.g. NC 5625 0668).

247 Widespread brittle deformation cuts Moine country rocks, the Loch Shin
248 Granite and associated granite-pegmatite veins alike (Figs 4a-f). The Loch Shin
249 Granite is cut by steeply-dipping, several metre long planar dextral faults trending
250 WNW-ESE with shallowly plunging slickenlines (Figs 3c, 4c). The total offsets are
251 unknown. Dextral faults are everywhere associated with shorter length, steeply-
252 dipping N-S to NE-SW sinistral faults with cm-scale offsets (Figs 3c, 4a) that either
253 abut against, or are cross-cut by dextral faults (Fig. 4d) suggesting that they are
254 contemporaneous. Irregularly oriented, mainly shallowly-dipping reverse faults
255 with prominent NNW- to SSE-plunging grooves & slickenlines are locally present in
256 the granite (e.g. around NC 5635 0630; Figs 3d, 4e). The fault planes are curvilinear
257 & lineated, with a series of ramp-flat configurations. Offsets are mostly small (mm-
258 cm scale). Once again these faults show mutually cross-cutting relationships with
259 the steeply dipping strike slip faults suggesting that they are broadly

260 contemporaneous. A stress inversion analysis of all fault slickenline data suggests a
261 normal faulting to transtensional stress regime with a component of N-S shortening
262 and E-W extension, consistent with regional-scale dextral shear along the Loch Shin
263 Fault (Fig 3f).

264 In addition to brittle faults, both Moine rocks and granite are locally cut by
265 metre-scale zones of brecciation and cataclasis, some of which appear to be
266 associated with specific faults whilst others are diffuse and irregular. The banded
267 Moine rocks locally preserve brittle-ductile box folds with generally moderate to
268 steep easterly plunges (e.g. Figs 3e, 4f). These structures refold the ductile foliation
269 (S2) and lineation (L2). The age of these folds relative to granite emplacement is
270 uncertain, but one example appears to detach along a NE-SW sinistral fault
271 suggesting that the folds are also post-granite features related to the regional brittle
272 deformation. Such folds have not been observed within the granite, but this may
273 reflect the lack of a pre-existing mechanical layering in these rocks.

274 In thin section, the effects of brittle deformation and cataclasis are
275 widespread in all samples from the Loch Shin shore section (e.g. Figs 5a-f). Irregular
276 networks of small-offset shear and hybrid fractures host variable amounts of
277 mineralization and secondary alteration features including sericite and other clay
278 minerals, quartz, chlorite, hematite, pyrite, chalcopyrite, limonite, fluorite and
279 zeolite (e.g. Figs 5c, e, f). This suggests that the fractures have hosted significant
280 volumes of fluid, an assertion supported by the widespread preservation of multiple
281 sets of healed microfractures (Tuttle lamellae) in quartz in a wide range of
282 orientations (Figs 5d, e). The presence of both pyrite and chalcopyrite in these

283 fracture fills suggest that at least some of the widely observed base metal
284 mineralization was synchronous with brittle deformation. In several cases, sericite-
285 filled fractures cutting feldspars are seen to pass laterally into well-defined Tuttle
286 lamellae in adjacent quartz grains (Fig. 5e). Isolated veins of zeolite <1 mm thick
287 cross cut all other brittle structures (Fig. 5f) and appear to represent the final phase
288 of mineralization.

289

290 ***GRUDIE GRANITE***

291 The Grudie Granite is poorly exposed and none of its contacts have been observed.
292 In surface exposures, the granite is unfoliated, medium to fine grained, with sparse
293 large phenocrysts of perthitic K-feldspar up to 1 cm across and large rounded
294 xenocrysts of polycrystalline quartz up to 1 cm across (see Appendix B,
295 Supplementary Material). These are set in a matrix of lightly to moderately
296 sericitized plagioclase and quartz, with sparse K-feldspar, biotite and iron oxide.
297 Little internal variation in grain size or mineralogy has been observed and internal
298 contacts were not found.

299 In the field, well-developed joints carry epidote, chlorite, zeolite, iron and
300 manganese oxides with slickenlines locally developed in a variety of orientations,
301 mainly dip-slip or oblique slip. In thin section, the effects of brittle deformation are
302 limited with small fractures filled mainly with epidote, white mica, chlorite and
303 limonite. The overall level of fracturing is less intense than in the Loch Shin Granite.

304

305 **ZIRCON U-Pb ISOTOPE ANALYSIS**

306 *Sample, mineral separation and analytical protocols*

307 A representative sample of Loch Shin granite from the SW west shore of Loch Shin
308 (DS1-11; Fig. 2b, NC 5635 0625) was selected for Zircon U-Pb LA-ICP-MS
309 geochronology. The analytical detail for the U-Pb analysis, including zircon
310 reference materials, is presented in Appendix C (see also Darling *et al.* 2012). In
311 brief, zircons were separated from sample DS1-11 using traditional methods and
312 mounted in epoxy resin. Prior to Laser ablation (LA)-ICP-MS U-Pb isotope analyses
313 they were imaged via cathodoluminescence. Laser ablation (LA)-ICP-MS U-Pb isotope
314 analyses were undertaken at the University of Portsmouth, using a New Wave 213
315 nm Nd:YAG laser coupled with an Agilent 7500cs quadrupole ICP-MS.

316

317 *Results*

318 The zircons separated from sample DS1-11 are generally small (<120 μm in length).
319 The majority of the zircons possess euhedral to sub-euhedral prismatic forms, with
320 oscillatory or banded zonation textures as revealed by CL imaging (Fig. 6).
321 Approximately 15 percent of grains are significantly different, and have variable
322 habit from equant to elongate with sub-euhedral to anhedral forms. The CL textures
323 of these grains are also variable, including sector zonation, broad banding and
324 oscillatory zonation with spongy overgrowths. A total of 19 zircon grains were
325 analysed by LA-ICP-MS, including a range of textural types (Table I). Three analyses
326 were rejected due to high levels of ^{204}Pb (common Pb), which was not corrected for
327 during data reduction.

328 The majority of the analyzed grains yield Silurian ages, although there is one
329 concordant analysis with a $^{207}\text{Pb}/^{206}\text{Pb}$ age of 1284 ± 19 Ma and three slightly
330 discordant analyses with $^{207}\text{Pb}/^{206}\text{Pb}$ ages ranging from 1725 to 1771 Ma (Table I,
331 Fig 7a; all age uncertainties given to two standard deviations). These older grains
332 are of the equant, anhedral group and have Th/U ratios (0.4-0.6) that are
333 significantly lower than the Silurian grains (Th/U = 0.9 to 1.5). Ten of the prismatic,
334 more euhedral grains with oscillatory zonation textures yield $^{206}\text{Pb}/^{238}\text{U}$ ages
335 ranging from 416 to 436 Ma (Fig. 7b). In combination, these grains yield a concordia
336 age of 427.3 ± 3.7 Ma. Two additional analyses yielded discordant U-Pb isotope data,
337 and fall on a discordia line between the younger concordant population and ca.
338 1700 Ma. These are interpreted as mixed analyses, which is supported by the
339 observation of variable isotopic ratios in the time resolved signals. The 427.3 ± 3.7
340 Ma concordia age of the younger group of prismatic zircons, with CL textures
341 (oscillatory or fine-banded) and Th/U ratios (0.9-1.5) typical of igneous zircon, is
342 taken as the best estimate of intrusion age of the Loch Shin Granite (Fig. 8).

343

344 **RHENIUM-OSMIUM MOLYBDENITE GEOCHRONOLOGY**

345 *Samples*

346 Four molybdenite samples were collected for rhenium-osmium (Re-Os)
347 geochronology to constrain the timing of sulphide mineralization associated with
348 the Loch Shin and Gruide granite intrusions. Although molybdenite mineralization
349 was noted in several places within the Loch Shin intrusion by Gallagher & Smith
350 (1975) only one *in-situ* quartz-molybdenite vein was observed in the field (AF33-10;

351 NC 5614 0650; Fig. 2b). The ~1 cm quartz vein hosts minor fine grained (~1 mm)
352 rosettes and disseminated molybdenite grains. No appreciable alteration selvage is
353 present, with the exception of minor silicification, and chloritization of magmatic
354 biotite.

355 Three additional samples were selected from the area around the Grudie
356 granite. Molybdenite±pyrite mineralization sufficient for geochronological analysis
357 was only observed in the neighboring Moine rocks adjacent to the intrusion (Fig. 2b).
358 The mineralization post-dates all ductile Moine fabrics. Molybdenite mineralization
359 is associated with and without quartz veins and, similar to the Loch Shin granite,
360 wallrock alteration is limited to silicification, and chloritization of biotite in the
361 Moine rocks. Molybdenite within quartz veins is fine grained (0.5 to 1mm) and
362 occurs as disseminations and parallel to the boundary between the quartz vein and
363 wallrock (AF01-11; AF02-11). Molybdenite also occurs as coatings along fractures
364 (AF36-10).

365

366 *Mineral separation and analytical protocols*

367 Molybdenite separation and Re-Os analytical protocols follow the methodology
368 described by Selby and Creaser (2004), and Lawley and Selby (2012). These are
369 summarized in Appendix D.

370

371 *Results*

372 The four molybdenite samples from the Loch Shin (n = 1) and Grudie granites (n =
373 3) possess between ~1.6 and 8 ppm Re and 7.5 and 36 ppb ¹⁸⁷Os. All four

374 molybdenite samples yield ages identical within uncertainty (Table II; Figure 8),
375 indicating that mineralization associated with the Loch Shin and the Grudie granite
376 intrusions occurred during the upper mid-Silurian (ca. 428 – 430Ma).

377

378 **FLUID INCLUSION ANALYSIS**

379 *Analytical protocols*

380 Three molybdenite-bearing quartz veins from the Loch Shin Granite and wall rocks
381 of the Grudie granite were studied in the Geofluids Research Laboratory at the
382 National University of Ireland Galway (see Appendix E for analytical details). A
383 petrographic classification scheme for the quartz-hosted fluid inclusions was
384 developed using transmitted polarised light microscopy (Table III; see also
385 Appendix F, Supplementary Material).

386

387 *Fluid Inclusion Petrography*

388 Molybdenite-bearing quartz veins were investigated from the Loch Shin Granite
389 (one sample: AF33-10) and from the Moine wall rocks of the Grudie Granite (two
390 samples: AF35-10 and AF02-11). The fluid inclusion petrographic study adopted the
391 concept of fluid inclusion assemblages (FIA) described by Goldstein (2003), an
392 approach that places fluid inclusions into assemblages interpreted to represent
393 contemporaneous fluid trapping. Fluid inclusions (FIs) in all samples display
394 ellipsoidal to irregular morphologies. Inclusions are commonly ~10 µm in longest
395 dimension and show low degrees of fill ($F=0.7-0.95$). The degree of fill [$F=\text{vol. liquid}$
396 / (vol. liquid + vol. vapour)] was measured by estimating the proportions of liquid

397 and vapour at 25°C and comparing to published reference charts (Shepherd *et al.*,
398 1985). Four inclusion types (*Type 1*, *Type 2*, *Type 3* and *Type 4*) have been identified
399 hosted in vein quartz and their petrological characteristics are presented in Table III.
400 The classification scheme is based on phase relations in fluid inclusions at room
401 temperature.

- 402 • *Type 1* are two-phase liquid-rich (L>V) aqueous inclusions. They are
403 abundant in all three samples, occurring in trails and in clusters and they
404 commonly display subrounded to irregular shapes. They range from 9 µm to
405 25 µm in length and their degree of fill is ~0.70 to 0.95.
- 406 • *Type 2* are monophasic aqueous fluid inclusions (L only), and are present in
407 all samples. They occur in trails alongside *Type 1* FIs and range in longest
408 dimension from 1 µm to 5 µm in length. These are interpreted as being
409 metastable and indicate fluid trapping temperatures of < 50°C (Goldstein and
410 Reynolds, 1994).
- 411 • *Type 3* are three-phase (L+L+V) aqueous-carbonic fluid inclusions. They are
412 aligned within annealed fractures and occur as clusters or as isolated
413 individuals. They exhibit subrounded to subangular morphologies that range
414 between 4 and 17 µm in the longest dimension.
- 415 • *Type 4* are monophasic (L) carbonic fluid inclusions. They are aligned within
416 annealed fractures and also occur in clusters associated with *Type 3*
417 aqueous-carbonic inclusions. They range between 5 and 10 µm in longest
418 dimension and possess rounded to sub-rounded morphologies. They are rare

419 and have been observed in samples AF33-10 (Loch Shin Granite) and AF02-
420 11 (Grudie Granite).

421

422 *Fluid Inclusion Microthermometry*

423 In sample AF33-10 from the Loch Shin Granite, T_{FM} values for Type 1 range from -
424 50.5° to -45.5°C. This temperature interval indicates the probable presence of NaCl
425 and $CaCl_2$ (Shepherd *et al.*, 1985). T_{LM} values are from -13.5 to -1.1°C yielding
426 salinities ranging from ~ 1.9 to 17.3 eq. wt. % NaCl (mean 9.7 eq. wt. % NaCl). Fluid
427 inclusions homogenise to the liquid state between 119°-170°C (Table III, Fig. 9a).

428 T_{FM} values for Type 1 in sample AF02-11 from the Grudie Granite wall rocks
429 range between -23° and -22.5°C corresponding to the eutectic point of the H_2O -
430 $NaCl \pm KCl$ system. T_{LM} values range from -3.60 to -0.70°C yielding salinities of ~3.7
431 to 6.9 eq. wt. % NaCl (mean 5.4 eq. wt. % NaCl). Homogenization to the liquid state
432 occurs between 214° and 279°C. In sample AF35-10 T_{LM} values for Type 1 range
433 from -4.3° to -2.2°C yielding salinities ranging from ~1.2 to 5.9 eq. wt. % NaCl (mean
434 4.4 eq. wt. % NaCl) Type 1 FIs homogenise to the liquid state between 151° and
435 244°C (Table III, Fig. 9a).

436 Type 3 aqueous-carbonic inclusions have been identified in all three samples
437 but only microthermometry on Grudie Granite samples (AF02-11 and AF35-10) are
438 reported here, because of the size (<3 microns) of these inclusions in the Loch Shin
439 sample. CO_2 homogenisation (to the liquid state, and by meniscus fading at 31.10°C)
440 occurs between 28° and 30.9°C yielding CO_2 densities that range between 0.47 and
441 0.65 gm/cc. CO_2 melting temperatures range from -56.6°C (the triple point for pure

442 CO₂) to -57.2°C, the latter indicates the presence of additional species (*e.g.* H₂S +H₂ –
443 see LRM results). Clathrate (CO₂ 5.75 HO₂) melting takes place between +5.6° and
444 +9.9°C yielding aqueous phase salinities between ~0.2 and 8.1 eq. wt. % NaCl. Total
445 homogenization to the liquid state occurred between 214.2° and 279.5°C in sample
446 AF35-10, and between 262° and 308.2°C in sample AF02-11. Homogenization to the
447 vapour phase occurred in three inclusions in sample AF02-11 at ~332.7°C (Table III,
448 Fig. 9a).

449

450 *Laser Raman Microspectroscopy*

451 Laser Raman Microspectroscopy (LRM) was used to identify the phases present in
452 all fluid inclusion types observed in the three samples (Appendix G, Supplementary
453 Material) and revealed the presence of CO₂, N₂ and H₂S. LRM of Type 1 fluid
454 inclusions in all samples indicates the presence of CO₂. Type 3 FIs from the Grudie
455 granite wall rock samples have in addition to CO₂, trace amounts of H₂S and H₂. LRM
456 of Type 4 FIs from both granites indicates that they are composed of pure CO₂ with
457 trace amounts of H₂S.

458

459 *Interpretation*

460 The Mo-bearing veins from each of the granites contain a similar range of fluid
461 inclusion types, *i.e.* Types 1-4. Type 1 in the Grudie Granite wall rock veins display
462 similar fluid salinities that range between ~1 and 7 eq. wt. % NaCl. However, Type 1
463 from the Loch Shin Granite, display a significantly wider range of salinities *i.e.* ~2-18
464 eq. wt. % NaCl. This difference is coupled with T_H values for the Loch Shin sample

465 that are generally $<180^{\circ}\text{C}$ which contrasts markedly with the range recorded for
466 Type 1 and 3 from the Grudie Granite wall rock veins ($\sim 180^{\circ}\text{-}350^{\circ}\text{C}$). T_{H} histograms
467 (Fig. 9a) for Type 1 and 3 fluid inclusions indicate a decrease in homogenization
468 temperatures from Type 3 ($\sim 340^{\circ}\text{C}$) through Type 1 ($\sim 260^{\circ}\text{C}$) in the Grudie Granite
469 wall rock veins to Type 1 ($<180^{\circ}\text{C}$) fluid inclusions in the Loch Shin Granite vein.
470 Bivariate plots of T_{H} and salinity show no obvious correlations, however, Type 1
471 inclusions from the Loch Shin Granite vein display an essentially isobaric variation
472 in salinity (Fig. 9b). This low T isobaric trend displayed by the Loch Shin Type 1
473 inclusions is directly comparable to that displayed by high salinity fluids (Type 3)
474 recorded in the Galway, Donegal, Newry and Leinster Granites in Ireland. Here, they
475 are interpreted to represent basinal brines, sourced in overlying sedimentary basins,
476 which circulated through the crystalline basement during a period of post-
477 Caledonian crustal extension or transtension (see Conliffe *et al.* 2010 and references
478 therein). It is arguable, therefore, that the Type 1 fluids recorded in the Loch Shin
479 vein may post-date and be unrelated to Mo-mineralisation. Consequently P-T
480 modelling using the fluid inclusion data is only performed for the Grudie Granite
481 veins.

482

483 *P-T Modelling*

484 Grudie Granite wall rock veins: The molybdenite Re-Os chronometry shows that the
485 mineralisation in both veins is contemporaneous and occurred ca. 428Ma.
486 Accordingly, the timing of fluid trapping in AF02-11 and AF35-10 is considered to
487 be broadly contemporaneous. Bulk fluid inclusion parameters were calculated using

488 the LRM results in combination with the microthermometric data, using the
489 computer programs CLATHRATES (Bakker, 1997) and FLUIDS (Bakker, 2003).

490 Isochores for the high and lower temperature Type 1 aqueous fluids and for
491 the Type 3 aqueous carbonic fluids in the two vein samples are presented in the P-T
492 diagram (Fig. 10). The field for Type 3 inclusions is defined by two isochores that
493 reflect their range of microthermometric data. Isochores for the lower and higher
494 temperature Type 1 aqueous fluids were constructed for salinities of ~4.5 and 5
495 eq.wt% NaCl matched with T_H values of ~176 and ~251°C, respectively
496 corresponding to their range of salinities and T_H values. The veins are spatially and
497 genetically related to the Grudie Granite which places constraints on the pressure
498 regime active during mineralisation. Ferguson and Al-Ameen (1985) calculated
499 pressures of 2.50 ± 0.25 kb for the aureole of the Omey Granite, Connemara which has
500 Mo mineralisation of a similar age and setting to the Grudie Granite (Feely *et al.*,
501 2007). These pressure constraints are used in Figure 10 to estimate trapping
502 temperatures for Type 3 fluids of ~340 to 410°C. Furthermore, Gallagher *et al.*,
503 (1992) used fluid inclusion microthermometry and stable isotope data to generate a
504 P-T model for Mo- mineralisation at the western end of the Galway Granite which
505 yielded pressures of 1.2 to 2.0 kb and a temperature range of 360 to 450°C (see
506 Figure 10). A higher pressure and lower temperature regime prevailed during
507 Grudie Granite mineralisation indeed similar to that modelled for the Omey Granite
508 (Feely *et al.*, 2007). No evidence for fluid immiscibility was recorded in Type 1
509 inclusions and therefore they could have been trapped anywhere along their
510 respective isochores. Type 1 fluids are considered to be meteoric and trapped after ,

511 and at lower pressures than, the earlier magmatic aqueous carbonic Type 3
512 inclusions considered to be responsible for the Mo-mineralisation. The P-T history
513 of fluids in the Grudie Granite wall rock veins may have followed the path shown in
514 Figure 10 (black arrow).

515

516 **DISCUSSION**

517 *The relative and absolute ages of plutonism, mineralisation and deformation*

518 The U-Pb zircon and Re-Os molybdenite ages for the Loch Shin Granite and sulphide
519 mineralization associated with both plutons are all coincident and overlap almost
520 exactly within error (Fig. 8). These ages therefore confirm the geological
521 observations which suggest that the plutons and associated mineralisation are
522 contemporaneous and genetically related. The Loch Shin-Grudie granite ages
523 overlap within error with the U-Pb zircon (TIMS) age of 425 ± 1.5 Ma reported by
524 Kocks *et al.* (2013) for the central granodiorite of the Rogart pluton (Fig. 1a) which
525 was, according to these authors also emplaced contemporaneously with dextral
526 movements along the Strath Fleet Fault, the along strike southeastern continuation
527 of the Loch Shin Fault and the LSL (Fig. 1a).

528 The field and thin section observations suggest that the Loch Shin and Grudie
529 granites are petrologically similar – as suggested by previous authors (e.g. Gallagher
530 & Smith 1975). Both plutons post-date the ductile deformation fabrics in the
531 surrounding Moine and Lewisian rocks, including the main Scandian-age D2
532 structures. Both plutons are associated with a variety of ore mineralization,
533 including molybdenite and other base metal sulphides, and both are post-dated by

534 the effects of brittle deformation consistent with dextral transtensional movements
535 along the WNW-ESE-trending Loch Shin Fault. Unsurprisingly the intensity of this
536 brittle overprint is greater in the Loch Shin pluton, which lies closer to the main
537 fault trace.

538 The relative ages of the brittle faulting and mineralization are more complex.
539 Field and thin section observations of fracture-hosted sulphides (pyrite,
540 chalcopyrite) show that at least some of the base metal mineralization is
541 contemporaneous with the brittle deformation. The observations lend support to
542 the long-postulated proposal that the dextral movements along NW-SE faults such
543 as the Loch Shin, Strath Fleet and Dornoch Firth fault systems are contemporaneous
544 with, and antithetic to, regional sinistral movements along the GGFZ ca 425 Ma
545 (Johnson & Frost 1977; Watson 1984; Stewart *et al.* 2001). It also strengthens the
546 arguments of Dewey & Strachan (2003) and Kocks *et al.* (2013) that the switch from
547 regional sinistral transpression with thrusting to transtension with regional strike
548 slip faulting occurred at this time.

549 However, many brittle fractures also cross-cut mineral veins. Furthermore,
550 the Type 1 fluid inclusions seen as Tuttle lamellae in the Loch Shin granite are
551 clearly distinct from the fluid inclusion sets seen in the Grudie granite. Their
552 presence points to a somewhat later, near surface phase of fluid flow associated
553 with brittle dextral movements along the Loch Shin-Strath Fleet Fault system. Given
554 this specific association, it seems most likely that at least some dextral faulting and
555 fluid flow occurred over a protracted period into the Devonian (?Emsian, ca 410 Ma)

556 where it was associated with basin development and the very final stages of late
557 Caledonian strike-slip faulting/transension (cf. Dewey & Strachan 2003).

558

559 *Pluton relationships at depth and the magnitude of dextral strike-slip faulting*

560 The very poor levels of exposure in the Loch Shin-Lairg region make it difficult to
561 ascertain how the various plutonic bodies in this area may be related in 3
562 dimensions. Gravity modelling by Hipkin & Hussain (1983) has ruled out the
563 possibility that the large regional gravity low seemingly centred on the surface
564 outcrop of the Grudie pluton (Fig. 1b) is due to the presence of a very large pluton at
565 depth. More recent work by Leslie *et al.* (2010) suggests that the low occurs mainly
566 due to the presence of a thick thrust culmination of Moine rocks (the Cassley
567 Culmination, Fig. 2a) sitting structurally above and to the SE of the Assynt
568 Culmination. Nevertheless, their gravity models suggest the presence of a shallowly
569 buried pluton with horizontal dimensions of 7 x 11 km, with an average thickness of
570 up to 3 km (see Leslie *et al.* 2010, fig. 10). Even allowing for significant errors in the
571 calculations, these models indicate that the granites exposed in the Loch Shin-Lairg
572 region (including the Grudie, Loch Shin, Claonel bodies) are likely to be underlain by
573 a larger, possibly composite plutonic body located mainly to the SW of Loch Shin
574 (Fig. 11a). It is tempting to suggest that this buried granite and the similarly
575 composite Rogart body are part of a single pluton offset by dextral strike-slip
576 faulting. However, this would require right lateral displacement of at least 10 km
577 which seems at odds with other regional evidence. For example, the observed
578 offsets of regional markers such as the nearby Loch Shin Lewisian inlier (Fig 2a)

579 suggest displacements of no more than a few hundred metres, as does the
580 observation that the Loch Shin Fault does not appear to continue very far to the NW
581 beyond the end of Loch Shin (Leslie *et al.* 2010). It seems more likely therefore that
582 the two plutons are separate, composite bodies located either side of the Loch Shin-
583 Strath Fleet fault system in a manner rather similar to other Caledonian plutons that
584 are associated with regional strike-slip fault zones in NW Scotland, most notably the
585 GGFZ (e.g. Hutton 1988b; Jacques & Reavy 1994; Stewart *et al.* 2001).

586

587 *Implications for the nature and significance of the Loch Shin Line*

588 The present study lends support to the suggestion of Watson (1984) that the NW-SE
589 trending Loch Shin Line (LSL) is associated with an anomalous zone of broadly
590 contemporaneous mantle-derived appinites, granites (Rogart, Grudie, Loch Shin and
591 many smaller satellite bodies) intruded ca. 425-428 Ma. These are postdated by
592 slightly younger (perhaps as young as ca 410 Ma) brittle dextral faulting in the
593 Moine Nappe SE of the Moine Thrust (Loch Shin-Strath Fleet and Dornoch Firth
594 faults, Fig. 1b). Watson (1984) suggested that the LSL corresponds to the location of
595 a Precambrian shear zone in the Lewisian autochthon underlying the Moine Nappe
596 which acted as a deep crustal channelway controlling the ascent of magmas and
597 mineralization during the later stages of the Caledonian orogeny (see also the leaky
598 lower crustal fault block model of Jacques & Reavy 1994). The most obvious
599 candidate structure seen in the Lewisian Complex west of the Moine Thrust Zone is
600 the steeply S-dipping Laxford Front, the major shear zone that separates the

601 Rhiconich and Assynt terranes; this lies almost parallel to and along strike from the
602 trace of the LSL (Figs 1, 11b).

603

604 *Constraints on regional exhumation rates at the end of the Caledonian orogeny*

605 The PT estimates derived from the fluid inclusion study reported here (Fig. 10) can
606 be compared with those for peak metamorphism in the central part of the foreland-
607 propagating Scandian thrust wedge in Sutherland in order to provide constraints on
608 the rate of regional exhumation. Integrated metamorphic and isotopic studies and
609 thermal modelling suggest that peak metamorphic conditions in the vicinity of the
610 Naver Thrust of ca. 650°C and 5.5 kbar (Friend *et al.* (2000) were attained at c. 440-
611 435 Ma (Johnson & Strachan 2006; Thigpen *et al.* 2013). In contrast, this study has
612 established temperature-pressure conditions at the time (ca. 425 Ma) of Grudie
613 Granite mineralisation of c. 375°C and 2.5 kb. The contrasting pressure estimates
614 suggest that around 10 km thickness of crust was removed in c. 10-15 myr, easily
615 achieved at an erosion rate of less than or equal to 1mm a⁻¹. Essentially the same
616 erosion rate was derived by Johnson & Strachan (2006) from consideration of
617 isotopic data and the likely (Emsian) age of the oldest Old Red Sandstone strata to
618 rest unconformably on the Moine rocks.

619

620 *The regional significance of Caledonian molybdenite mineralization*

621 Intrusion-related molybdenite mineralization is documented throughout the Scottish
622 and Irish Caledonian-Appalachian Orogen (Figure 1a inset). The broad timing and
623 fluid characteristics of intrusion-related Mo-mineralisation in the Loch Shin and

624 Grudie Granite veins (ca. 428 Ma) is temporally similar to that of the Ballachulish
625 and Kilmelford igneous complexes, including the Lagaloachan porphyry Cu-Mo
626 system (ca. 433-426 Ma; Appendix H; Conliffe *et al.*, 2010), pre-dates that of the
627 Etive Igneous Complex (ca. 415 Ma; Porter and Selby, 2010), Shap granite (ca. 405
628 Ma; Selby *et al.*, 2008) and the earliest granite related Mo-mineralisation in the Irish
629 sector of the Caledonian-Appalachian Orogen (ca. 423Ma, Feely *et al.*, 2010). Fluid
630 inclusion data for these systems indicate that Mo-mineralization is ultimately
631 associated with aqueous-carbonic fluids, which has also been shown to be common
632 among Cu+Mo mineralization associated with late Caledonian magmatism (Kay
633 1985; Gallagher *et al.* 1992; Feely *et al.* 2007; Selby *et al.* 2008; this study; Feely &
634 Selby, unpub data; see Appendix I, Supplementary Material).

635 Gold mineralisation in Dalradian metamorphic rocks at Curraghinalt,
636 Northern Ireland (Parnell *et al.* 2000; Rice *et al.*, 2012) and Tyndrum, Scotland
637 (Patrick *et al.* 1988; Curtis *et al.* 1993) has also been linked to aqueous-carbonic
638 magmatic fluids that may have been derived from an underlying Caledonian
639 intrusive. Although CO₂ has only an indirect role on gold mineralization
640 (Lowenstern 2001), it may play a significant role in magmatic fluid exsolution and
641 evolution, and may lead to concentrations of Au, Cu and Mo into the vapour phase
642 (Heinrich *et al.* 1999; Ulrich *et al.* 2001). As such, intrusion-related Mo (+Cu)
643 mineralization may warrant attention during future mineral exploration,
644 particularly for porphyry Cu–Mo–Au mineralization and additionally for
645 structurally-controlled Au-mineralisation distal from the intrusion. In this regard
646 combined fluid inclusion data, U-Pb and Re-Os geochronometry have shown that

647 prolonged granite-related molybdenite mineralisation in the Connemara region was
648 accompanied by aqueous-carbonic fluids in the Omev Granite at ca. 423 Ma and later
649 in the Galway Granite at ca. 410Ma (Murvey), ca. 407Ma (Mace Head) and ca. 380Ma
650 (Costelloe; Feely *et al.*, 2007, 2010). Moreover, the earliest granite related Mo-
651 mineralisation of the Omev Granite was also initiated while major orogen parallel
652 structures, *e.g.* Great Glen and Southern Upland Fault systems (Dewey and Strachan,
653 2003) were active.

654

655 **CONCLUSIONS**

656 Using detailed field observations, microstructural studies, U-Pb zircon and Re-Os
657 molybdenite geochronology and fluid inclusion analyses, we have shown that a suite
658 of mid-Silurian (ca. 425-430 Ma) granite plutons (Grudie, Loch Shin, Rogart and
659 many smaller associated bodies) are contemporaneous with base metal sulphide
660 mineralization, including molybdenite. Synchronous to slightly younger (ca. 427-
661 410Ma) brittle dextral strike slip faulting along the WNW-ESE Loch Shin-Strath
662 Fleet Fault System was antithetic to regional sinistral strike-slip movements along
663 the NE-SW trending GGFZ (Fig. 11a). More generally, the associated plutonism,
664 mineralization and strike-slip faulting confirms the transition from regional-scale
665 transpression to transtension during the mid-Silurian to early Devonian in NW
666 Scotland as postulated by Dewey & Strachan (2003).

667 Our findings also lend support to the existence of the NW-SE trending Loch
668 Shin Line and to the hypothesis of Watson (1984) that it has acted as a deep crustal
669 channelway controlling the ascent and emplacement of Silurian granitic and

670 appinitic magmas into the overlying Moine Nappe (Fig. 11b). It seems very likely
671 that this deep structure corresponds to the southeastern continuation of the
672 Precambrian-age Laxford Front shear zone in the buried Lewisian autochthon. This
673 further illustrates how pre-existing crustal structures can be persistently
674 reactivated even when buried beneath much younger thrust nappes and influence
675 directly the migration and emplacement of hydrous mineralizing fluids and magmas
676 (e.g. Jacques & Reavy 1994; Richards 2013).

677

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682

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904

905 **Figure captions**

906 **Figure 1a)** Regional geology map of the northern Scottish Highlands. Inset map
907 shows the relative positions of Laurentia, Baltica, Avalonia and Gondwana following
908 the closure of the Iapetus Ocean (Caledonide-Appalachian belt in black).
909 Abbreviations as follows: AC = Assynt Culmination; DFF = Dornoch Firth Fault; GGFZ
910 = Great Glen Fault Zone; LCM = Loch Coire Migmatite complex; LSSFF = Loch Shin -
911 Strath Fleet Fault ; MF = Moray Firth; MT = Moine Thrust; NT = Naver Thrust; ORS =
912 Old Red Sandstone; R = Rogart igneous complex.

913 **b)** Gravity map of the Lairg-Loch Shin area, with locations of appinitic intrusions
914 (Achnuie hybrids, yellow dots), Laxford front and surface trace of Loch Shin Line
915 shown (after Watson 1984 and Leslie *et al.* 2010).

916

917 **Figure 2a)** Overview geological map of the Loch Shin area after Strachan and
918 Holdsworth (1988) & Leslie *et al.* (2010). Box shows location of map shown in
919 Figure 2b. G= Grudie, C = Claonel, LS = Loch Shin granites. L = Lairg; LSF = Loch Shin

920 Fault; AS = Aird of Shin. **b)** Simplified version of geology in the Loch Shin – Grudie
921 area (after Gallagher & Smith 1975). Geochronology sample locations are shown. GB
922 = Grudie Burn; CCB = Cnoc na Cloich-bhuaile; MG = Meall a' Ghruididh; AC = Allt a'
923 Chlaonaidh.

924

925 **Figure 3)** Equal area stereoplots of structural data collected from the Loch Shin
926 shore section. **a)** Ductile foliation (Sn/S2; great circles) and L2 mineral lineations
927 (dots). **b)** Granite veins (solid great circles) and quartz veins (dashed great circles)
928 and lineation on quartz vein (dot). **c)** Steep faults (great circles) and slickenlines
929 (dots). **d)** Shallow faults (great circles) and slickenlines (dots). **e)** Box fold hinges
930 (dots) and axial surfaces (great circles). **f)** Stress inversion analysis and Mohr plot of
931 combined fault slickenline data with weighting added to include fault sizes. LSF =
932 inferred local orientation of Loch Shin Fault.

933

934 **Figure 4)** Brittle structures cutting the Loch Shin granite and its Moine country
935 rocks. **a)** Plan view of NE-SW sinistral fault offsetting granite and quartz vein (NC
936 5635 0631). **b)** Plan view of NW-SE dextral fault offsetting granite pegmatite vein in
937 Moine psammities (NC 5639 0613). **c)** Oblique sectional view of long NW-SE
938 trending dextral fault scarp in Loch Shin granite; inset shows sub-horizontal
939 orientation of slickenlines on fault surface consistent with strike-slip fault
940 movement (NC 5631 0650). **d)** NE-SW sinistral fault offsetting and being offset by
941 NNW-SSE dextral faults in Loch Shin granite (NC 5635 0631). **e)** Shallowly NW-
942 dipping flats and shorter SE-dipping ramps ('r') in exposed small displacement, top-

943 to-the-NW faults; inset shows plan view of corrugated, lineated fault surface with
944 NW-SE slickenlines (NC 5635 0632). **f)** Plan view of steeply plunging conjugate box
945 folds detaching along sub-vertical NE-SW sinistral fault in Moine psammites (NC
946 5638 0621).

947

948 **Figure 5)** Thin sections of brittle structures and mineralization cutting the Loch
949 Shin granite and its country rocks. **a)** Small offset (<0.5mm) domino style reverse
950 (top-to-the-NW) shear fractures (arrowed) cutting Loch Shin granite viewed in ppl
951 (NC 5635 0632). **b)** Typical zone of cataclasis cross cutting Loch Shin Granite
952 viewed in crossed polars (NC 5635 0632). **c)** Irregular region of quartz iron oxide-
953 ilmenite (black) -pyrite (black, Py) -fluorite (Fl) mineralization in Moine psammites
954 immediately to the northwest of the Loch Shin granite viewed in ppl (NC 5625
955 0666). **d)** Multiple sets of fluid inclusions following healed microcracks/Tuttle
956 lamellae in quartz from the Loch Shin granite viewed in ppl (NC 5635 0632). **e)**
957 Microfractures lined with sericite where they cross-cut feldspar (Fsp) passing
958 laterally into healed microcracks/Tuttle lamellae in quartz (Qtz), in granite
959 pegmatite vein, viewed in crossed polars (NC 5639 0613). **f)** Late zeolite vein (Z)
960 cutting brecciated Moine psammite viewed in cross-polars (NC 5625 0666).

961

962 **Figure 6)** Cathodoluminescence images and SHRIMP II analysis positions for
963 representative grains from grains selected for geochronology from the Loch Shin
964 Granite sample. Also shown are the grain numbers, and $^{207}\text{Pb}/^{206}\text{Pb}$ ages for each
965 analysis pit (uncertainties are two standard deviations; percentage discordance

966 shown in brackets).

967

968 **Figure 7 a, b)** Zircon U-Pb concordia plots from the Loch Shin granite.

969

970 **Figure 8)** Plot of the U-Pb zircon and Re-Os molybdenite dates including 2 sigma
971 uncertainty with decay constant uncertainty for the Loch Shin and Gruidie granites.

972 Also given is the weighted average for the Re-Os molybdenite dates for the Gruidie
973 granite. For sample locations, see Figure 2.

974

975 **Figure 9) a)** Histogram of TH values and **b)** bivariate plot of TH vs. salinity for Type
976 1 and Type 3 inclusions in samples AF02-11 and AF35-10 from the Gruidie granite
977 and for Type 1 in sample AF33-10 from the Loch Shin granite.

978

979 **Figure 10)** Pressure-temperature space showing isochores for Type 1 and Type 3
980 fluid inclusions. Shaded area represents the field for Type 3 fluids defined by two
981 isochores. Isochores for the lower and higher temperature Type 1 aqueous fluids
982 are also shown and the parameters used for their construction are shown on the
983 isochores. Proposed P - T path for cooling history of fluids in Gruidie Granite is shown
984 by the arrow. P - T field for aqueous carbonic fluids associated with the Mo
985 mineralisation at the western end of the Galway Granite is shown for comparison
986 after Gallagher *et al.*, (1992).

987

988 **Figure 11) a)** 3-D summary of the spatial relationships between the Rogart, Loch

989 Shin, Lairg and Grudie plutons (red) and brittle strike slip faults (grey) in the Loch
990 Shin-Strath Fleet-Dornoch Firth area. **b)** Highly simplified conceptual model
991 showing how the buried Laxford front shear zone below the Moine nappe gives rise
992 to the Loch Shin Line of focussed Silurian magmas and overlapping Silurian-
993 Devonian dextral strike-slip faults.

994

995 **Tables**

996 **Table I)** U-Pb data for Loch Shin granite.

997

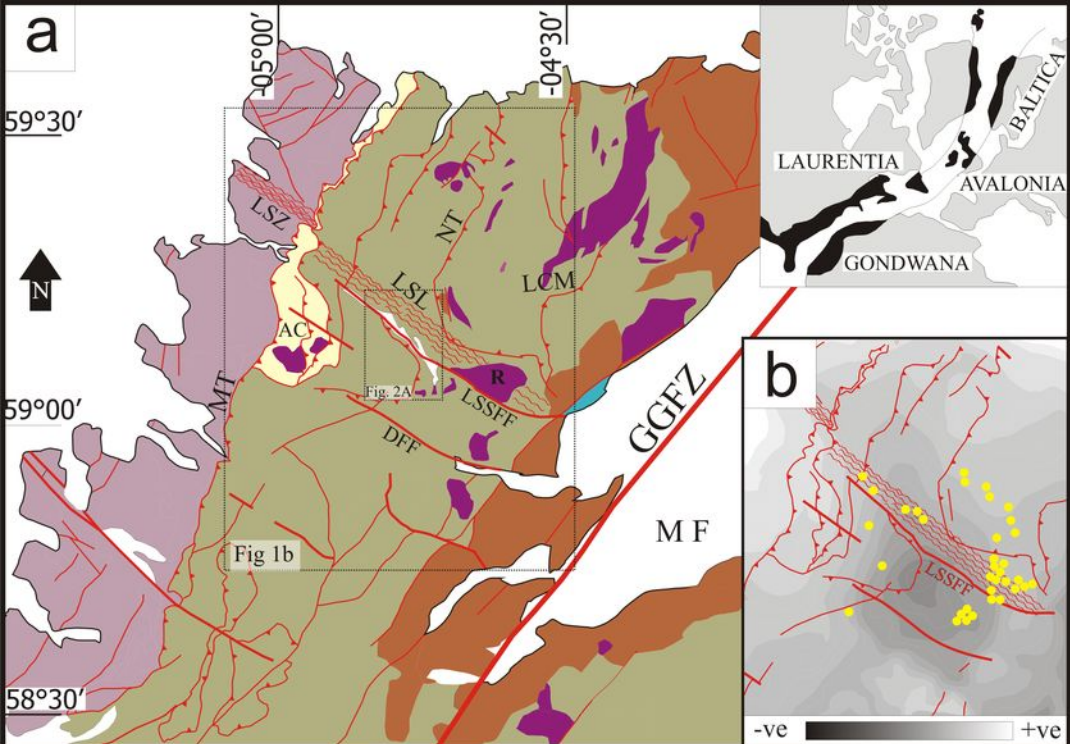
998 **Table II)** Re-Os data for molybdenite from the Loch Shin and Gruide granites.

999

1000 **Table III)** Classification of fluid inclusion types and fluid inclusion micro-
1001 thermometric data from the Loch Shin and Gruide granites.

1002

1003



Moine Supergroup

Caledonian Foreland

Moine Thrust Zone

Silurian -Devonian Granitoids

Devonian ORS

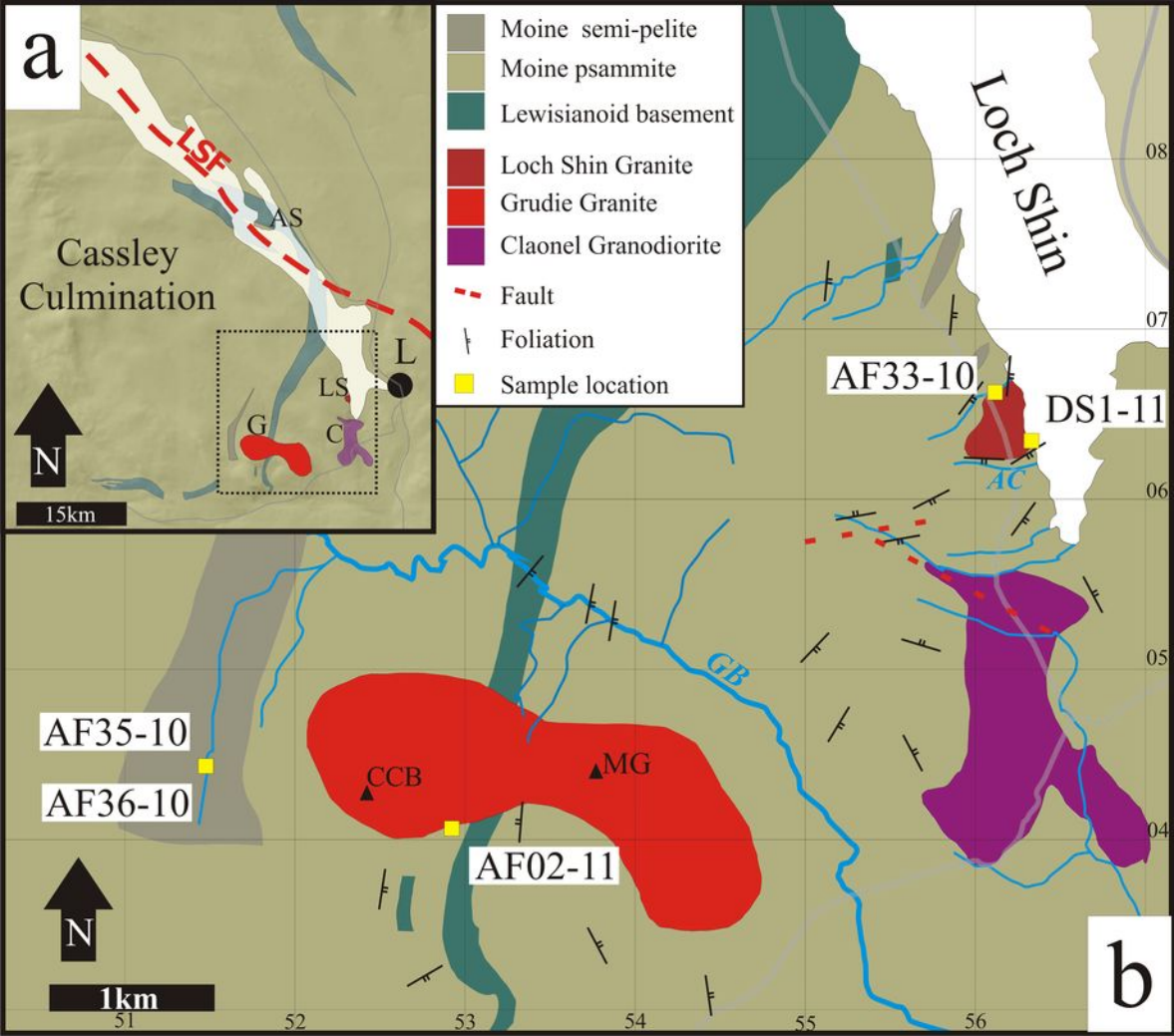
Jurassic Sediments

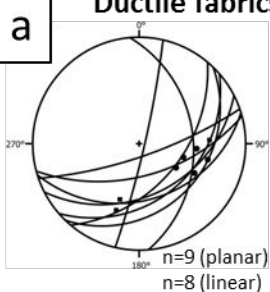
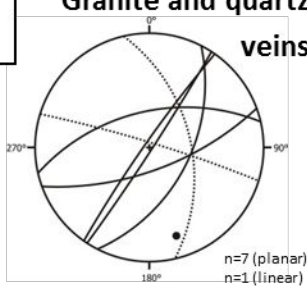
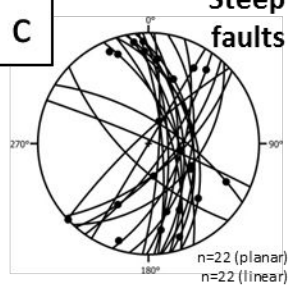
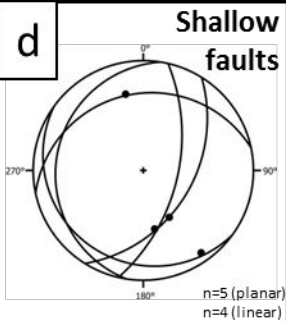
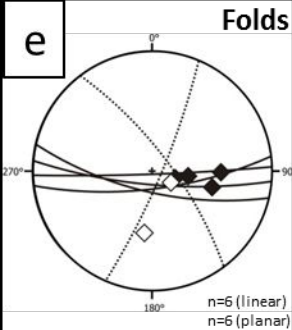
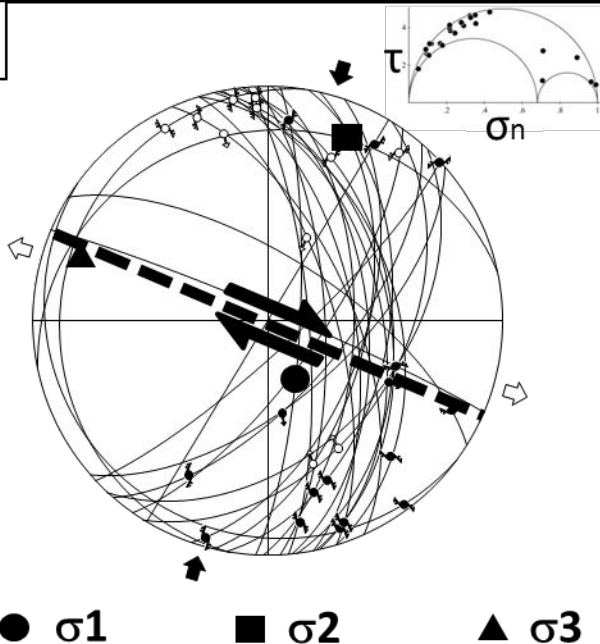
Fault

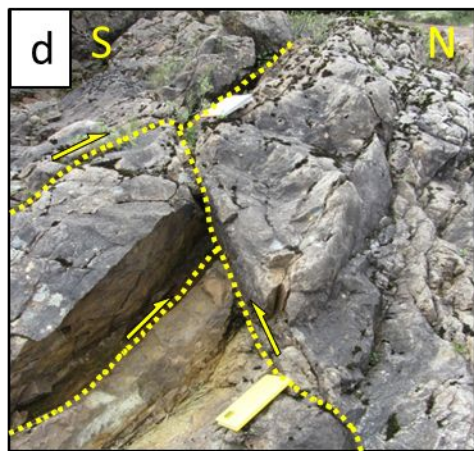
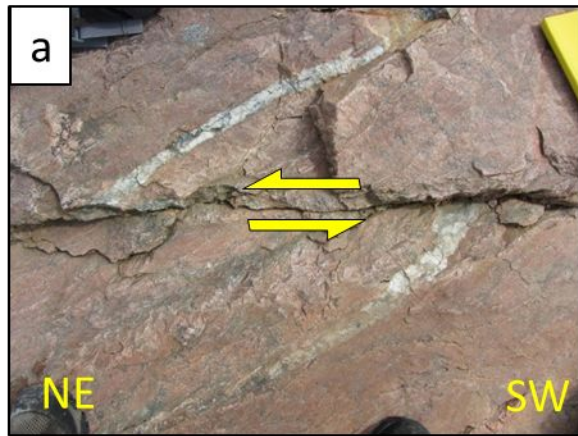
Thrust Fault

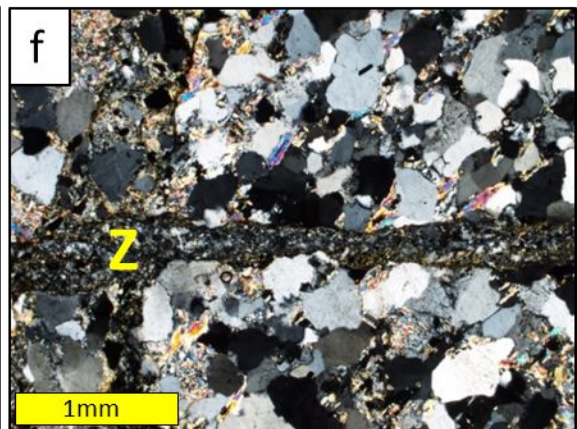
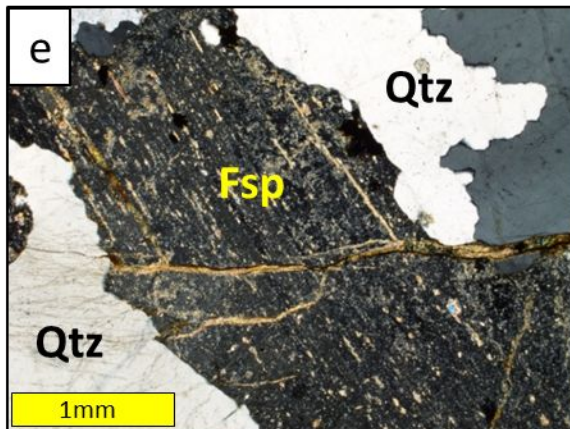
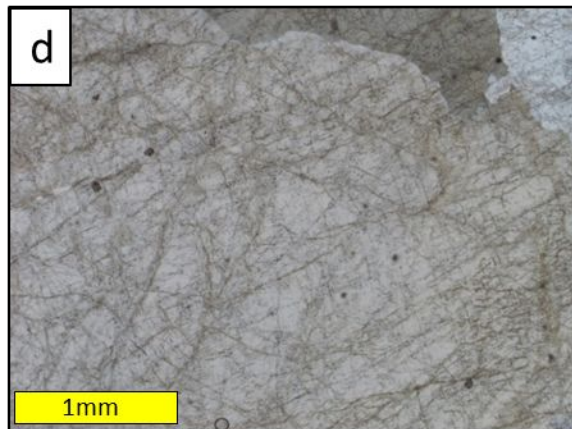
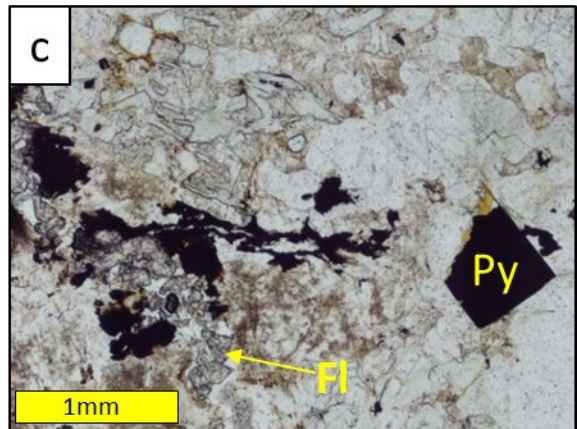
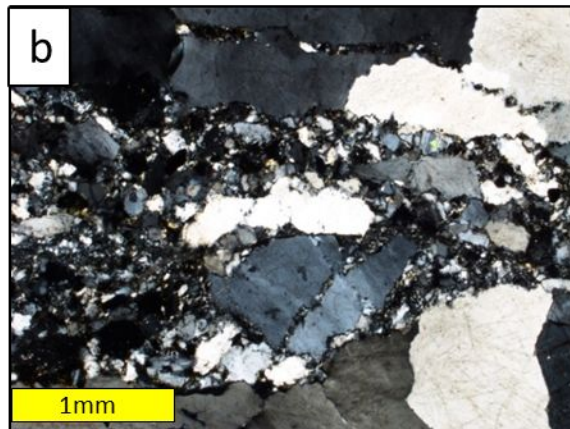
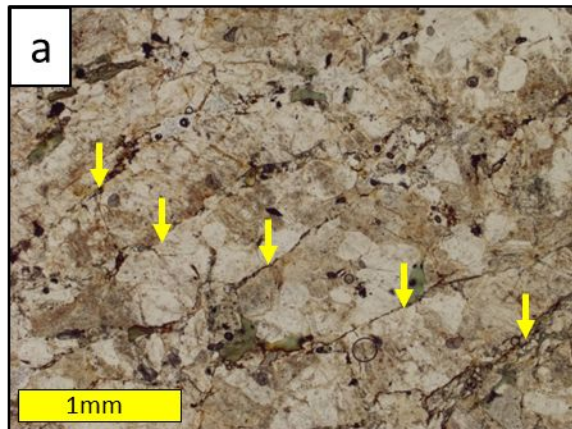
Basement Shear Zone / LSL

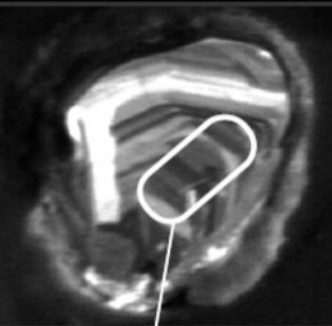
Appinites



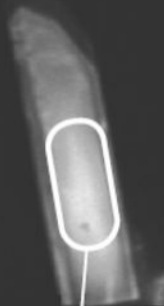
Ductile fabrics**Granite and quartz veins****Steep faults****Shallow faults****Folds****f**



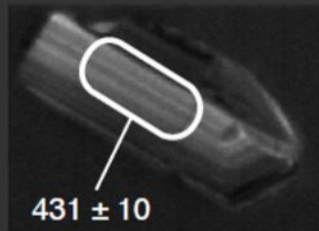




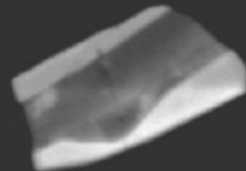
1306 ± 42 (1.7 %)



421 ± 18 (2.6 %)



431 ± 10
(0.5 %)



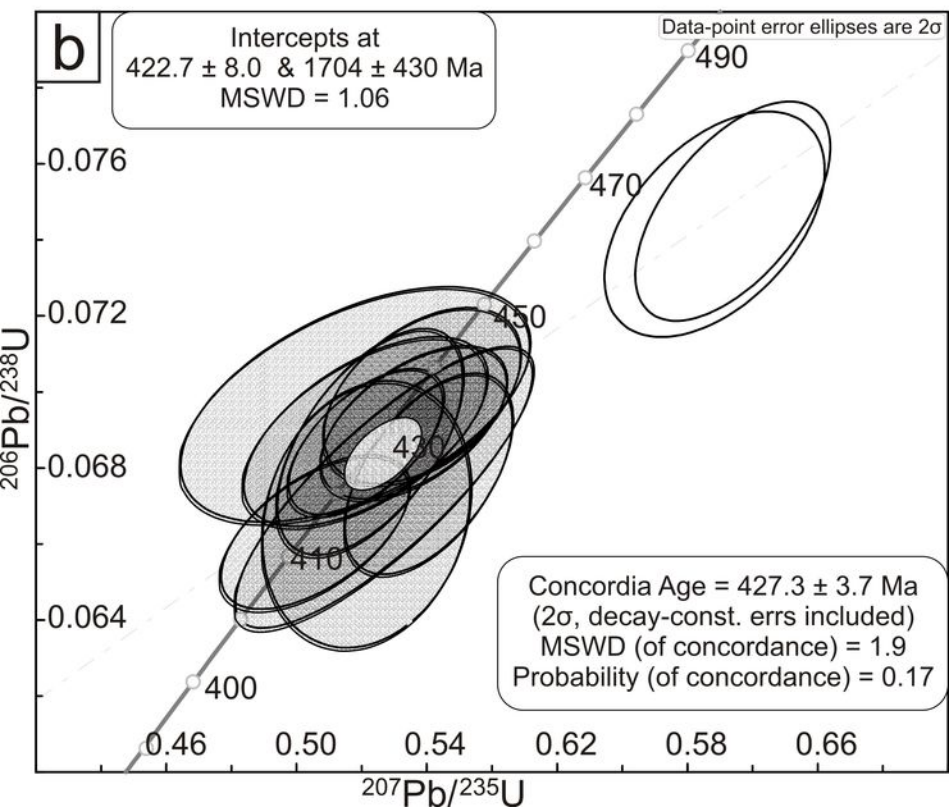
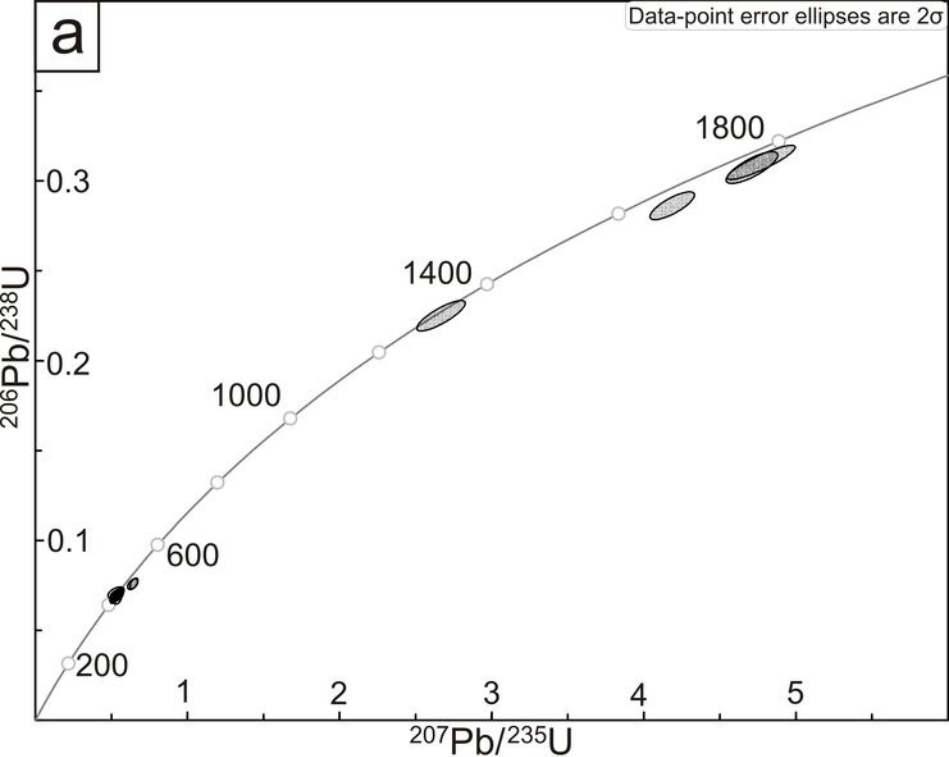
40 μm

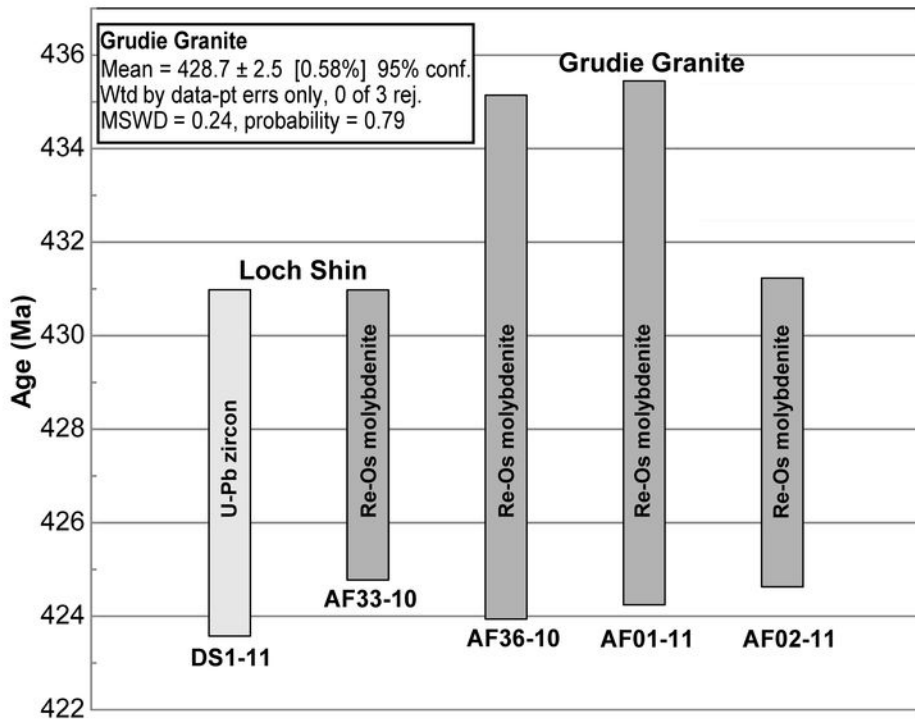


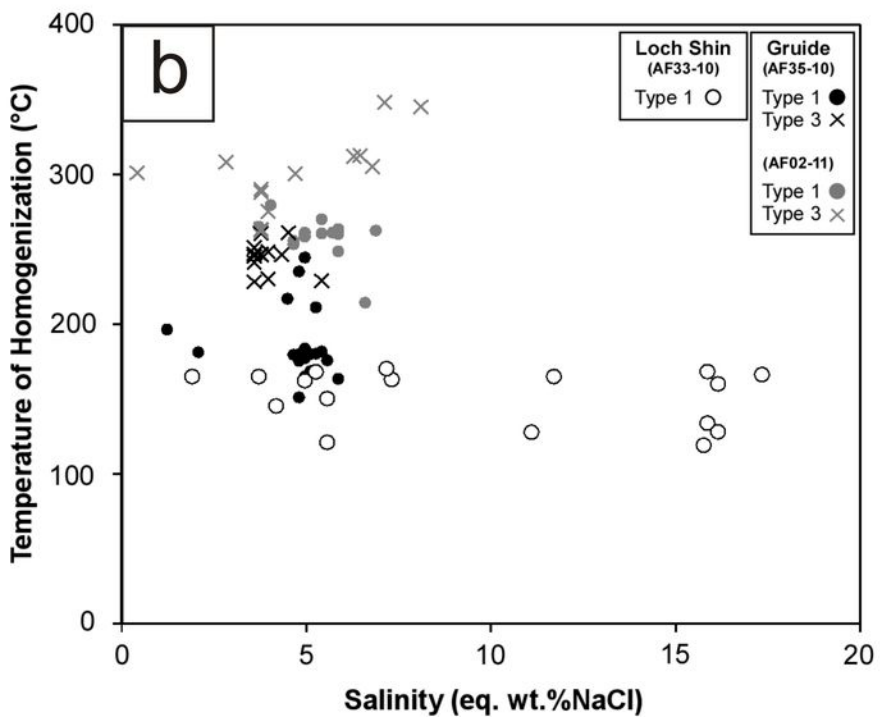
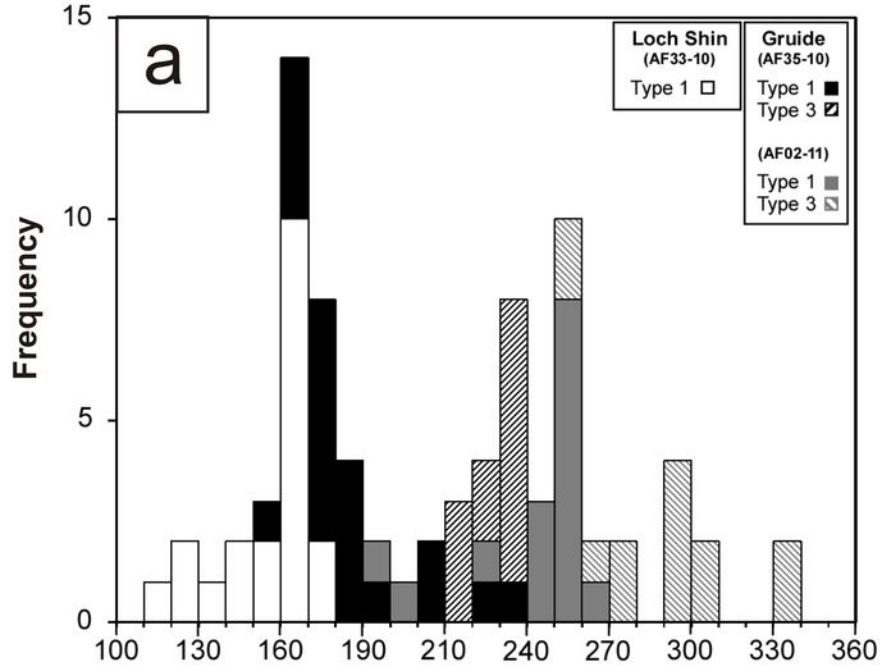
462.7 ± 15
(7 %)

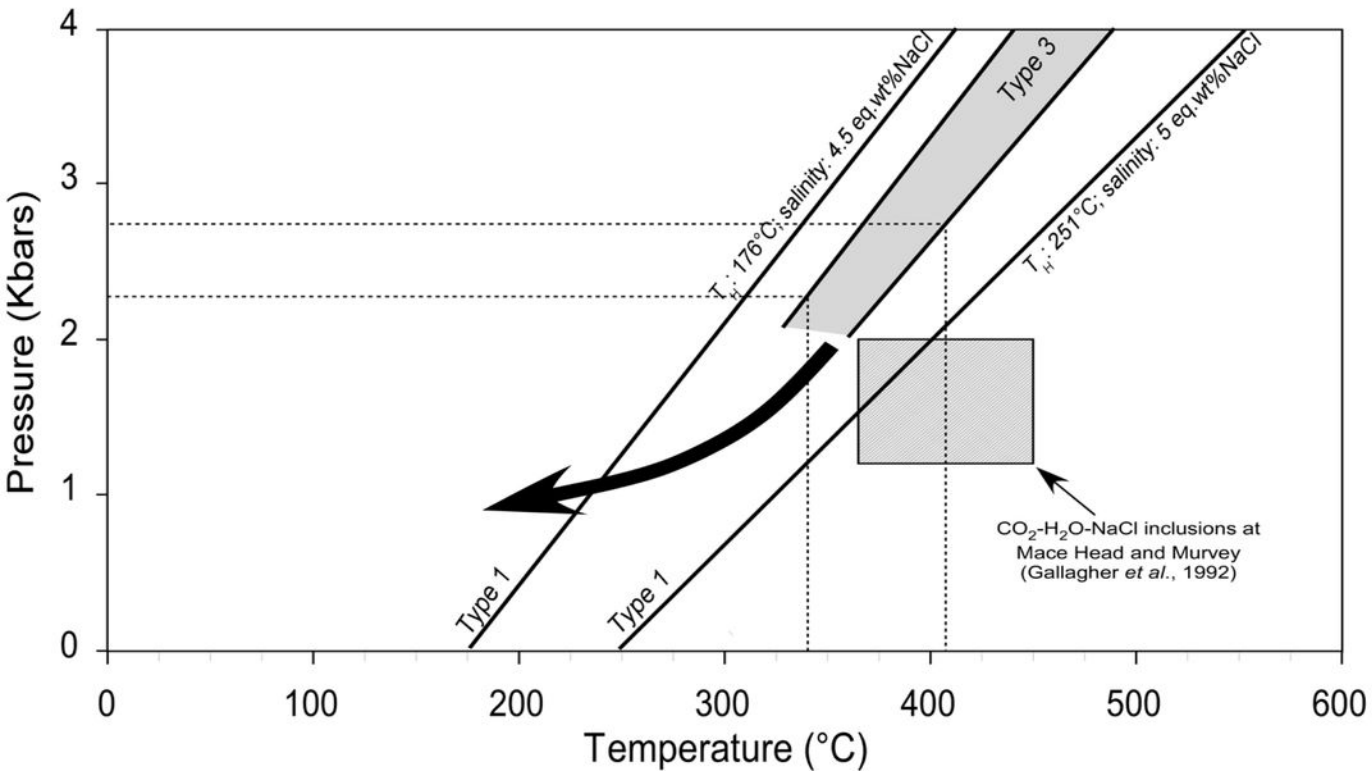


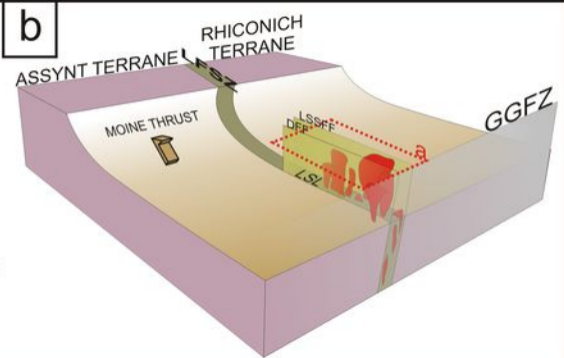
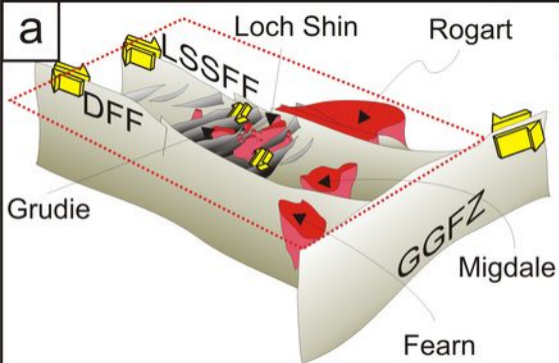
434 ± 16 (1 %)











School of Earth and Environmental Sciences, University of Portsmouth				Data for Tera-Wasserburg plot2					Data for Wetherill plot2					Age2								
Identifier	Comments	Beans (µm)	U (ppm)	Th (ppm)	Th/U	²⁰⁶ Pb/ ²³⁸ U	²⁰⁷ Pb/ ²³⁵ U	²⁰⁶ Pb/ ²³⁸ U	²⁰⁷ Pb/ ²³⁵ U	Rho	²⁰⁶ Pb/ ²³⁸ U	²⁰⁷ Pb/ ²³⁵ U	²⁰⁶ Pb/ ²³⁸ U	²⁰⁷ Pb/ ²³⁵ U	²⁰⁶ Pb/ ²³⁸ U	²⁰⁷ Pb/ ²³⁵ U	% concord					
Sample DS1-11																						
DE10A05	25x7	254	112	0.44	3525	37	3.494	0.039	0.106	0.001	4.188	0.061	0.286	0.003	0.8	1725	16	1622	18	1672	25	94
DE10A06	25x7	208	92	0.44	2414	32	3.251	0.038	0.107	0.001	4.716	0.070	0.308	0.004	0.8	1742	16	1729	20	1770	26	99
DE10A07	25x7	241	267	1.11	971	22	14.451	0.172	0.055	0.001	0.536	0.012	0.065	0.001	0.5	431	8	431	5	436	10	100
DE10A08	25x7	335	384	1.15	1722	37	14.558	0.258	0.056	0.001	0.523	0.012	0.069	0.001	0.6	435	8	428	8	427	10	99
DE10A09	25x7	105	61	0.58	2759	37	3.222	0.041	0.108	0.001	4.775	0.093	0.310	0.004	0.9	1771	18	1743	22	1780	35	98
DE10A11	25x7	465	481	1.03	505	46	14.738	0.235	0.056	0.001	0.541	0.011	0.068	0.001	0.5	444	8	423	7	439	9	95
DE10A12	25x7	266	228	0.85	891	30	14.558	0.166	0.055	0.001	0.521	0.010	0.069	0.001	0.6	427	6	428	5	426	8	100
DE10A13 High 204Pb - rejected	25x7	105	161	1.54	316	32	14.490	0.197	0.071	0.002	0.695	0.023	0.069	0.001	0.8	945	22	430	6	536	18	46
DE10A14	25x7	51	30	0.59	790	36	4.462	0.070	0.084	0.001	2.662	0.065	0.225	0.004	0.8	1295	19	1306	21	1318	32	102
DE10A15	25x7	141	154	1.1	348	27	15.079	0.188	0.056	0.001	0.506	0.012	0.066	0.001	0.6	454	9	414	5	416	10	91
DE10A16	25x7	282	259	0.92	1135	27	14.817	0.332	0.055	0.001	0.527	0.019	0.067	0.002	0.8	432	9	421	9	430	15	97
DE10B05	15x27	230	412	1.87	1814	36	14.974	0.332	0.056	0.002	0.522	0.013	0.067	0.001	0.2	446	13	417	9	426	11	93
DE10B06 High 204Pb - rejected	15x27	538	690	1.28	566	34	14.391	0.238	0.080	0.002	0.769	0.020	0.069	0.001	0.3	1209	32	433	7	579	15	36
DE10B07	15x27	253	209	0.83	1225	28	13.377	0.211	0.059	0.001	0.634	0.012	0.075	0.001	0.6	572	9	465	7	499	10	81
DE10B08	15x27	64	77	1.19	389	36	14.353	0.260	0.055	0.002	0.518	0.022	0.070	0.001	0.5	429	16	434	8	424	18	101
DE10B09	15x27	123	187	1.52	615	49	14.502	0.214	0.055	0.001	0.521	0.015	0.069	0.001	0.6	409	10	430	6	426	13	105
DE10B10	15x27	144	191	1.33	583	33	14.305	0.193	0.056	0.001	0.539	0.012	0.070	0.001	0.5	436	9	436	6	438	10	100
DE10B11	15x27	290	403	1.39	1043	27	13.439	0.219	0.051	0.001	0.628	0.014	0.074	0.001	0.5	638	13	463	8	495	11	75
DE10B12 High 204Pb - rejected	15x27	674	1044	1.55	242	31	16.781	0.463	0.124	0.008	0.930	0.041	0.060	0.001	-0.9	2021	134	374	9	667	30	18
Standard QJ-1																						
DE10A04	30x45	389	26	0.04	1301	26	10.158	0.095	0.060	0.001	0.807	0.011	0.098	0.001	0.3	594	8	605	6	601	9	102
DE10A12	30x45	266	11	0.04	1245	28	10.125	0.091	0.060	0.001	0.821	0.014	0.099	0.001	0.4	605	9	607	5	609	11	100
DE10A17	30x45	310	5	0	1179	26	10.127	0.143	0.060	0.001	0.818	0.012	0.095	0.001	0.7	609	7	607	9	607	9	100
DE10A04	30x45	300	13	0.04	1306	27	10.133	0.086	0.060	0.001	0.828	0.011	0.099	0.001	0.6	602	6	607	5	613	8	101
DE10A10	30x45	292	13	0.04	1205	27	10.151	0.087	0.060	0.001	0.826	0.011	0.099	0.001	0.7	613	6	606	5	611	8	99
DE10A17	30x45	289	12	0.04	1441	36	10.161	0.097	0.060	0.001	0.822	0.012	0.098	0.001	0.7	600	7	605	6	609	9	101
DE10B04	30x45	270	12	0.04	1176	42	10.155	0.118	0.060	0.001	0.831	0.013	0.098	0.001	0.7	590	7	605	7	614	10	103
DE10B13	30x45	267	11	0.04	965	34	10.179	0.120	0.059	0.001	0.814	0.013	0.098	0.001	0.7	580	7	604	7	605	10	104
Standard Temora 2																						
DE10A05	30x45	141	83	0.6	1043	30	15.103	0.234	0.055	0.001	0.509	0.010	0.066	0.001	0.4	429	9	413	6	418	8	96
DE10A06	30x45	143	85	0.6	1313	23	14.686	0.180	0.055	0.001	0.518	0.010	0.068	0.001	0.4	415	8	425	5	424	8	102
DE10A07	30x45	145	86	0.6	1067	36	14.767	0.181	0.055	0.001	0.514	0.010	0.068	0.001	0.4	426	8	422	5	421	8	99
DE10A08	30x45	144	100	0.7	1122	31	14.805	0.189	0.055	0.001	0.511	0.010	0.068	0.001	0.4	427	8	421	5	419	8	99
DE10C05	30x45	373	205	0.5	845	28	14.813	0.194	0.055	0.001	0.504	0.009	0.068	0.001	0.7	417	6	421	6	415	8	101
DE10C06	30x45	327	193	0.6	1187	34	14.974	0.206	0.055	0.001	0.498	0.009	0.067	0.001	0.7	411	6	417	6	410	7	101
DE10C14	20x20	346	188	0.5	1042	34	15.231	0.177	0.055	0.001	0.504	0.009	0.066	0.001	0.7	416	6	410	5	414	8	98
DE10C15	20x20	292	163	0.6	1063	29	15.126	0.180	0.055	0.001	0.501	0.009	0.066	0.001	0.6	414	6	413	5	413	7	100

1 concentration uncertainty < 20%

2 data not corrected for common Pb

3 2-sigma outlier based on (2006-2010) ages 207Pb/235U age/100

Decay constants of Jaffey et al 1971 used

Table II.

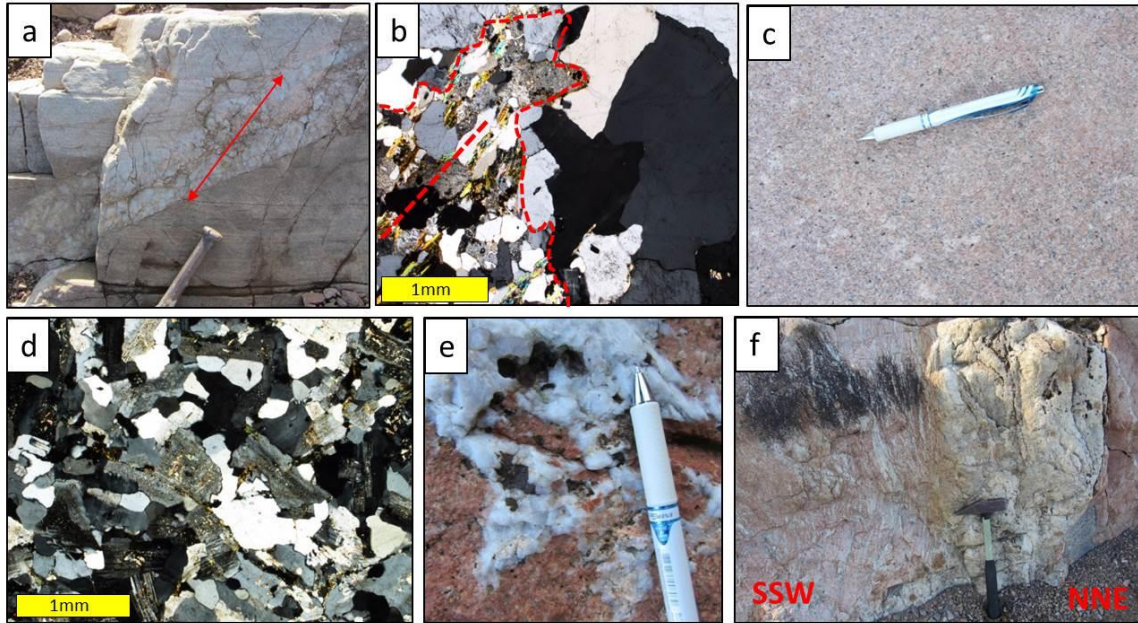
Sample	Location	wt	Re (ppm)	±	¹⁸⁷Re (ppm)	±	¹⁸⁷Os (ppb)	±	Age (Ma)	±	± (λ¹⁸⁷Re uncert)
<i>Loch Shin</i>											
AF33-10	Loch Shin, NC 56139, 06495	0.021	1.67	0.01	1.05	0.01	7.5	0.0	427.9	2.8	3.1
<i>Gruidie Granite</i>											
AF36-10	Moly Burn (Gallagher & Smith, 1975), NC 51530, 04646	0.012	3.53	0.03	2.22	0.02	15.9	0.1	429.6	5.2	5.6
AF01-11	Edge of Gruidie Granite NC 52726, 03797	0.014	3.40	0.03	2.14	0.02	15.4	0.1	429.9	5.2	5.6
AF02-11	Edge of Gruidie Granite NC 52726, 03797	0.010	8.04	0.05	5.05	0.03	36.1	0.2	428.0	3.0	3.3

Fluid Inclusion Types	Loch Shin granite	Gruide granite	
	sample AF33-10	sample AF35-10	sample AF02-11
Type 1 two-phase (L+V) liquid-rich aqueous inclusions 9-25 μm ; sub-rounded and irregular shapes; occur in trails aligned within annealed fractures; some clusters ¹ fluid composition: $\text{H}_2\text{O}-\text{NaCl}\pm\text{KCl}\pm\text{CO}_2$	F: 0.85-0.9 T_{FM} : -45.5° to -50.5° (mean: -47.9°C; N=7) T_{LM} : -13.5° to -1.1° (mean: -6.9°C; N=17) Salinity: 1.9 to 17.3 eq. wt%NaCl (mean: 9.7; N=17) $T_{\text{H}\rightarrow\text{L}}$: 119° to 170.1° (mean: 152.9°C; N=20) Abundant	F: 0.8-0.95 T_{LM} : -3.6° to -0.7° (mean: -2.8°C; N=20) Salinity: 1.2 to 5.9 eq. wt%NaCl (mean: 4.4; N=20) $T_{\text{H}\rightarrow\text{L}}$: 151° to 244.4° (mean: 185.6°C; N=20) Abundant	F: 0.7-0.9 T_{FM} : -22.5° to -23° (mean: -22.8°C; N=2) T_{LM} : -4.3° to -2.2° (mean: -3.3°C; N=20) Salinity: 3.7 to 6.9 eq. wt%NaCl (mean: 5.4; N=20) $T_{\text{H}\rightarrow\text{L}}$: 214.2° to 279.5° (mean: 258.3°C; N=20) Abundant
Type 2 monophasic (L) liquid aqueous inclusions 1-5 μm ; rounded to sub-rounded shapes; occur in trails within annealed fractures and randomly distributed fluid composition: $\text{H}_2\text{O}-\text{NaCl}$	² Trapping T < 50°C Abundant	Trapping T < 50°C Abundant	Trapping T < 50°C Abundant
Type 3 three- phase (L+L+V) aqueous-carbonic inclusions 4-17 μm ; elongated and irregular shapes; occur in trails aligned within annealed fractures; isolated or in clusters fluid composition: $\text{H}_2\text{O}-\text{CO}_2.\text{NaCl}\pm\text{H}_2\text{S}\pm\text{H}_2$	F: 0.8-0.9 Common	F: 0.8-0.9 T_{MCO_2} : -57.1° to -56.5° (mean: -56.7°C; N=20) T_{Mclath} : 7.2° to 8.2° (mean: 8°C; N=18) $T_{\text{HCO}_2\rightarrow\text{fading}}$: 30.5° to 31.1° (mean: 30.8°C; N=20) Salinity: 3.6 to 5.4 eq. wt%NaCl (mean: 4; N=18) Density: 0.468 g/cm ³ $T_{\text{HTOT}\rightarrow\text{L}}$: 228.2° to 261° (mean: 243.5°C; N=20)	F: 0.4-0.85 T_{MCO_2} : -57.2° to -56.2° (mean: -56.7°C; N=17) T_{Mclath} : 5.6° to 9.9° (mean: 7.2°C; N=19) $T_{\text{HCO}_2\rightarrow\text{L}}$: 28° to 30.9°; $T_{\text{HCO}_2\rightarrow\text{fading}}$ 31.1° Salinity: 0.2 to 8.1 eq. wt%NaCl (mean: 4.4; N=19) Density: 0.468 to 0.655 g/cm ³ $T_{\text{HTOT}\rightarrow\text{L}}$: 262° to 312.5° (mean: 243.5°C; N=10) $T_{\text{HTOT}\rightarrow\text{V}}$: 305° to 348° (mean: 332.7°C; N=3) Abundant
Type 4 monophasic (L) carbonic inclusions 5-10 μm ; rounded to sub-rounded shapes; occur in trails aligned within annealed fractures; some isolated fluid composition: $\text{CO}_2\pm\text{H}_2\text{S}$	Rare	Not Observed	Rare

Classification is based upon FI morphology and the volumetric proportion of phases observed at room temperature. L = liquid, V = vapour. ¹Bulk composition based on combined microthermometry and Raman spectroscopy. ²The presence of monophasic aqueous liquid FIs indicate trapping temperatures of < 50°C. \pm : trace or minor constituent. T_{FM} : temperature of first ice melting; T_{LM} : temperature of last ice melting; $T_{\text{HTOT}\rightarrow\text{L}}$: homogenisation temperature (to L); $T_{\text{HTOT}\rightarrow\text{V}}$: homogenisation temperature (to V); T_{Mclath} : temperature of clathrate melting; F: degree of fill; F=vol. liquid / (vol. liquid+vol. vapour).

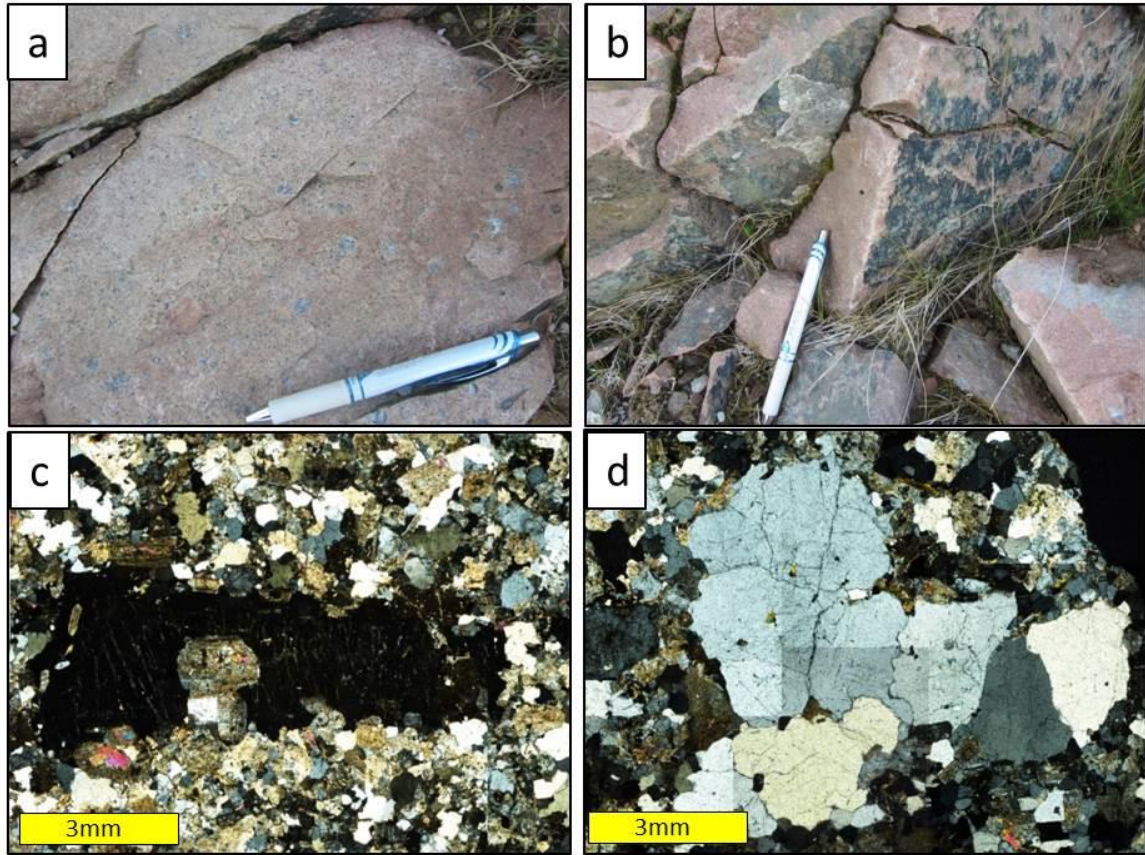
Appendices for Holdsworth et al. *Silurian-Devonian magmatism, mineralization, regional exhumation and brittle strike-slip deformation along the Loch Shin Line, NW Scotland*

Appendix A



The country rocks, Loch Shin granite and associated veins viewed in the field and thin section. **a)** Oblique view looking down onto undeformed granite pegmatite vein (077/55 NNW) cutting ACW of compositional banding in Moine psammites (100 metres to the SE of the Loch Shin granite (NC 5639 0613). Arrow shows inferred direction of vein opening based on offsets of thin semipelite layer. **b)** Thin section of undeformed granite pegmatite vein shown in (a) cross-cutting S0-S1-S2 fabric in Moine psammites (dashed yellow line). View in crossed polars, with igneous contact shown in red. **c)** Plan view in the field (NC 5631 0650) and **d)** in thin section (crossed polars) of typical undeformed Loch Shin granite (NC 5635 0631). **e)** Close-up plan view of irregular quartz-pyrite veins cutting Loch Shin granite (NC 5631 0650). **f)** Cross-section view of large NW-SE-trending quartz-galena veins (107/85N) cutting Loch Shin granite (NC 5630 0659).

Appendix B



Field and thin section views of the Grudie granite. **a)** Plan view of typical unfoliated Grudie granite with large pink K-feldspar and grey quartz phenocrysts/xenocrysts (NC 5268 0450). **b)** Oblique section view of slickenlined joints with chlorite and epidote mineralization (NC 5267 0444). **c)** Thin section of typical K-feldspar (in extinction) and **d)** polycrystalline quartz xenocryst/phenocrysts within Grudie granite (NC 5310 0427).

Appendix C

ZIRCON U-Pb ISOTOPE ANALYSIS

Sample, mineral separation and analytical protocols

A representative sample of Loch Shin granite from the SW west shore of Loch Shin (DS1-11; Fig. 2b, NC 5635 0625) was selected for Zircon U-Pb LA-ICP-MS geochronology. Zircons were separated from sample DS1-11 using heavy liquids and an isodynamic magnetic separator. The zircon fraction for analysis was handpicked under a binocular microscope and mounted in epoxy resin along with grains of the zircon reference material Temora 2 (Black *et al.* 2004). After polishing and carbon coating, cathodoluminescence (CL) images of the zircons were taken with a KeDev Centaurus CL detector housed on a JEOL 6060LV SEM at the University of Portsmouth (accelerating voltage = 15 kV).

Laser ablation (LA)-ICP-MS U-Pb isotope analyses were undertaken at the University of Portsmouth, using a New Wave 213 nm Nd:YAG laser coupled with an Agilent 7500cs quadrupole ICP-MS. Analytical protocols and instrument conditions are described in detail by Darling *et al.* (2012). Key points of the methodology are: (i) line-raster ablation (aspect ratio 1:1.5), in order to minimise time-dependent elemental fractionation; and (ii) external normalisation to the zircon standard Plesovice (Slama *et al.* 2008) using a 30 μm beam diameter. Laser beam diameters used on unknown zircons ranged from 30 to 15 μm , reflecting the scale of target domains within the crystals. Accuracy was monitored via analyses of the zircon reference materials Temora 2 and GJ-1. Eight analyses of Temora 2 (20 to 30 μm beam diameter) yield a U-Pb concordia age of 417.4 ± 3.5 Ma, and eight analyses of GJ-1 (30 μm beam diameter) yield a U-Pb concordia age of 606.6 ± 3.8 Ma: both of which are within uncertainty of the ID-TIMS reference ages for these materials (Black *et al.* 2004, Jackson *et al.* 2004).

Appendix D

RHENIUM-OSMIUM MOLYBDENITE GEOCHRONOLOGY

Mineral separation and analytical protocols

Molybdenite samples present in the area of the Grudie Granite were isolated using traditional methods of crushing, heavy liquids, and water flotation (Selby & Creaser, 2004). In contrast, given the minor abundance of molybdenite in the Loch Shin Granite sample (AF33-10), and to avoid losing molybdenite during crushing, the mineral separate was achieved using a room temperature HF dissolution of quartz protocol (Lawley & Selby, 2012).

The Re-Os analysis follows that outlined by Selby & Creaser (2004), which determines the Re and Os abundance of the molybdenite using isotope dilution negative thermal ionization mass spectrometry (ID-NTIMS). An aliquant of molybdenite, together with a known amount tracer solution (isotopically normal Os + ^{185}Re) are digested and equilibrated in a carius tube with 1ml 11N HCl and 3ml 15N HNO₃ for 24hrs at 220°C. Osmium is isolated and purified from the acidic solution using solvent extraction (CHCl₃) and micro-distillation methods. The Re is separated and purified using anion chromatography. The separated Re and Os were loaded on Ni and Pt wire filaments with BaNO₃ and BaOH activators, respectively, and analyzed for their isotope compositions using NTIMS via static Faraday collection. Analytical uncertainties are propagated and incorporate uncertainties related to Re and Os mass spectrometer measurements, blank abundances and isotopic compositions, spike calibrations, and reproducibility of standard Re and Os isotope values. The molybdenite analyses of this study were conducted during the same period as those of Lawley & Selby (2012). This study reported Re and Os blanks of <4 and 1 pg, respectively, with the $^{187}\text{Os}/^{188}\text{Os}$ of the blank being 0.25 ± 0.02 (n = 2). Further, Re-Os model ages determined using the ^{187}Re decay constant of $1.666 \times 10^{-11} \text{ a}^{-1}$ (Smoliar *et al.*, 1996) of molybdenite reference materials (NISTRM8599 = 27.6 ± 0.1 and 27.6 ± 0.1 Ma; HLP-5 = 220.0 ± 0.9 Ma), which are in good agreement with their accepted values determined at other laboratories and those previously reported at Durham University (Markey *et al.*, 1998, 2007; Porter & Selby, 2010).

Appendix E

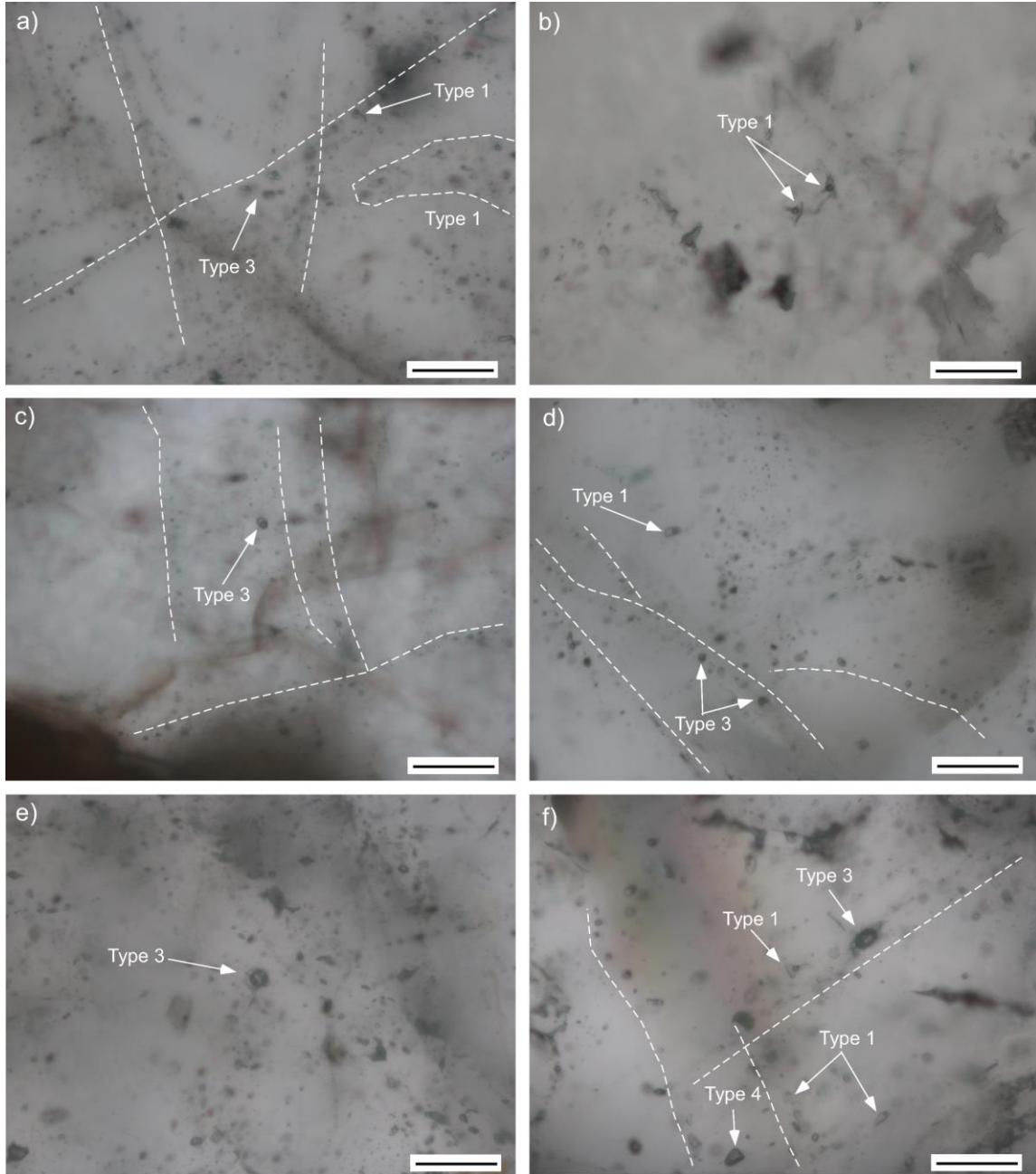
FLUID INCLUSION ANALYSIS

Analytical protocols

Microthermometric analysis was performed on doubly polished wafers (~100 μm thick) using a Linkam THMGS 600 heating freezing stage, mounted on an Olympus transmitted polarised light microscope. The instrument is equipped with a range of special long working distance objective lenses ranging up to 100x magnification. Calibration of the stage was performed using synthetic fluid inclusion standards (pure CO_2 and H_2O). Precision is $\pm 0.5^\circ\text{C}$ at 300°C and $\pm 0.2^\circ\text{C}$ at -56.6°C . Following procedures outlined by Shepherd *et al.* (1985), the temperature of first ice melting T_{FM} , the temperature of last ice melting T_{LM} and the temperature of homogenisation T_{H} were measured in quartz hosted two-phase liquid+vapour inclusions in all wafers (Fig. 9a). Fluid salinities were calculated using T_{LM} and the equations of Bodnar (1993). In addition, clathrate melting temperatures recorded in three-phase ($\text{L}_{\text{H}_2\text{O}}+\text{L}_{\text{CO}_2}+\text{V}_{\text{CO}_2}$) aqueous-carbonic inclusions were used with the equations of Duan *et al.*, (1996) to calculate their fluid salinities (Fig.9b).

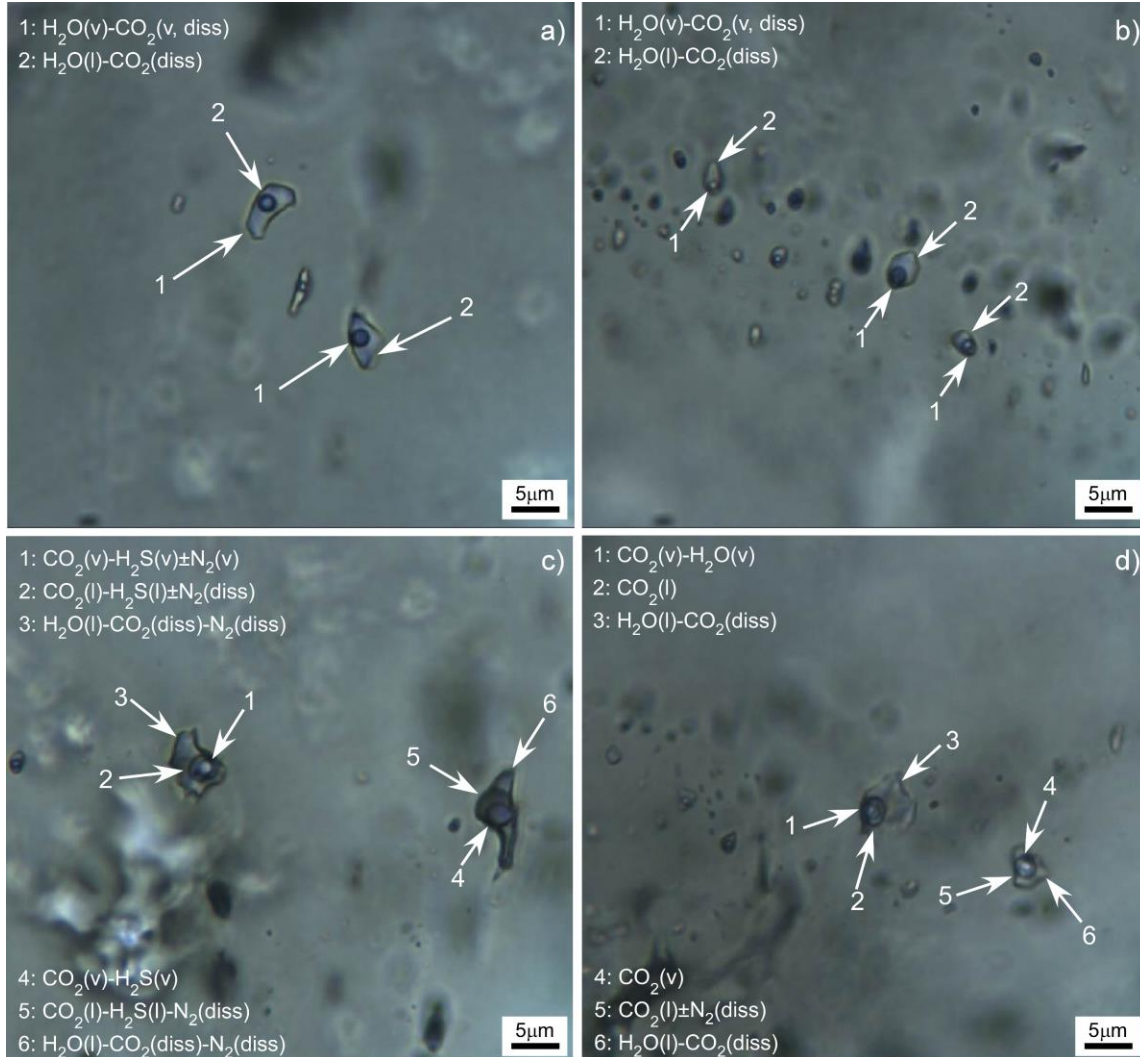
Laser Raman Microspectroscopy (LRM) of fluid inclusions was performed using a Horiba LabRam II laser Raman spectrometer. The instrument is equipped with a 600 groove mm^{-1} diffraction grating, a confocal and optical filter system, a Peltier-cooled CCD detector (255 x 1024 pixel array), and is coupled to an Olympus BX51 microscope. Fluid inclusion gas and liquid phases were analysed at room temperature using a 532 nm laser focused through either a 50x or 100x microscope objectives. The spatial resolution of the 532 nm laser at the sample was approximately 2 μm . Individual analyses were performed for between 10 to 60 seconds over the spectral range 1100 cm^{-1} to 4200 cm^{-1} . The number of spectral accumulations per analysis typically ranged between 2 to 5 in order to maximize the signal-to-noise efficiency of the spectrometer. Calibration of the instrument was routinely performed between analyses using the Raman peak of a pure silicon standard (520.7 cm^{-1}). Spectral uncertainty associated with the generation of Raman peak positions is estimated to be $\pm 1.5 \text{ cm}^{-1}$ (2σ ; 0.3%) based on replicate analyses of the standard.

Appendix F



Photographs of fluid inclusions (FI) trails from samples AF33-10 – Loch Shin Granite (**a,b**); AF35-10 (**c,d**) and AF02-10 (**e,f**) both from Gruide Granite. Scale bar = 50 μm .

Appendix G



Photomicrographs of Type 1 and Type 3 inclusions within quartz grains in sample AF35-10 (Grude granite) analysed under Laser Raman Spectroscopy. Type 1 two-phase liquid-rich aqueous inclusions distributed in isolated cluster **(a)** and trails **(b)**. Type 3 three-phase aqueous-carbonic inclusions distributed in clusters **(c and d)**.

Appendix H:
Re-Os data for molybdenite from Lagalochan Porphyry Cu-Mo system

Sample	location	wt (g)	Re(ppm)	±	187Re(ppm)	±	187Os(ppb)	±	Age	±
RO562-4_RC843047	Hole LD13-1A: 77.45-77.55m depth: 56deg 15' 19.0" N and 05deg 25' 41.1"	0.012	1215.9	5.5	764.2	3.5	5405.0	22.1	423.0	1.7
RO562-5_RC843048	Hole LD13-1A: 115.80-115.90m depth: 56deg 15' 19.0" N and 05deg 25' 41.1"	0.011	254.4	1.2	159.9	0.8	1131.5	4.9	423.2	1.7

Appendix I: Coire Buidhe Fluid Inclusions

Three types of fluid inclusion (Type 1, 2 and 3) were identified in vein quartz from Coire Buidhe (CB1A, B; see Porter & Selby 2010 for sample details), based on their morphology, composition and phase relations at room temperature. Type 1 inclusions are three-phase (liquid H₂O + liquid CO₂ + CO₂ vapour) at room temperature (~25°C). They are found as isolated inclusions, randomly distributed throughout the quartz, and appear to be primary or pseudosecondary in origin. On cooling, the carbonic phase in Type 1 inclusions freezes at ~ -120°C. Melting of the carbonic phase in all inclusions occurs between -57.1 and -56.5°C. Most melting temperatures are close to the triple point of CO₂ (-56.6°C), indicating that the inclusions contain almost pure CO₂. This is confirmed by Laser raman analysis that shows the volatile phase contains < 97.7% CO₂, with minor CH₄, N₂ and H₂S. Clathrate melting, in the presence of liquid CO₂ occurred between 3.7 and 6°C and yields salinities between 7.1 and 11.1 eq. wt% NaCl using the equation of Duan *et al.* (1995) and the software program CLATHRATES (Bakker, 1997). Homogenisation of CO₂ (to the liquid phase) occurs between 27 and 30.4°C, indicating a CO₂ phase density of 0.57 to 0.68 g/cc. Total homogenization to the liquid phase occurs between 291 and 353°C. Type 2 aqueous inclusions are two-phase (liquid + vapour) inclusions and occur along healed microfractures that crosscut quartz grain boundaries. The temperature of first ice melting (T_{FM}) takes place between -20.7 and -23.8°C, close to the eutectic temperature of the H₂O-NaCl-KCl system (-22.9°C). T_{LM} values are between -3.1 and -5.1°C. Clathrate melting between 1.2 and 2.9°C was observed in some Type 2 inclusions. This indicates the presence of non-aqueous phases in Type 2 inclusions. Laser raman analysis of Type 2 inclusions confirmed that the vapour phase contains > 98% CO₂ with minor amounts of CH₄, N₂ and H₂S. Clathrate melting temperatures have been used to calculate salinities of 5 to 5.2 eq. wt% NaCl using software program CLATHRATES (Bakker, 1997). Type 2 inclusions homogenised to the liquid phase (L + V → L) between 261.9°C and 282.9°C.

Type 3 inclusions are found in trails along annealed microfractures, that are occasionally crosscut trails of Type 2 inclusions. First ice melting temperatures were recorded between -21.3 and -24.6°C. T_{LM} values lie between -2.7 and -4.9°C and

correspond to salinities of 4.5 to 7.7 eq. wt% NaCl (Bodnar, 1993). Type 3 inclusions homogenized to the liquid phase ($L + V \rightarrow L$) between 165.5 and 218°C.

Additional References Cited in Appendices

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