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1 **Multi-chronometer dating of the Souter Head Complex: rapid exhumation**
2 **terminates the Grampian Event of the Caledonian Orogeny**

3
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18
19 **ABSTRACT**

20
21 The Souter Head sub-volcanic complex (Aberdeenshire, Scotland) intruded the high-
22 grade metamorphic core of the Grampian orogen at 469.1 ± 0.6 Ma (²³⁸U-²⁰⁶Pb
23 zircon). It follows closely peak metamorphism and deformation in the Grampian
24 Terrane and tightly constrains the end of the Grampian Event of the Caledonian
25 Orogeny. Temporally coincident U-Pb and ⁴⁰Ar/³⁹Ar data show the complex cooled
26 quickly with temperatures decreasing from ca. 800 °C to less than 200 °C within 1
27 Ma. Younger Re-Os ages are due to post-emplacement alteration of molybdenite to
28 powellite. The U-Pb and Ar/Ar data combined with existing geochronological data

29 show that D2/D3 deformation, peak metamorphism (Barrovian and Buchan style) and
30 basic magmatism in NE Scotland was synchronous at ca. 470 Ma and is associated
31 with rapid uplift (5-10 km/Ma) of the orogen, which by ca. 469 Ma had removed the
32 cover to the metamorphic pile. Rapid uplift resulted in decompressional melting and
33 generation of mafic and felsic magmatism. Shallow slab breakoff (50-100 km) is
34 invoked to explain the synchronicity of these events. This interpretation implies that
35 peak metamorphism and D2/D3 ductile deformation were associated with extension.
36 Similarities in the nature and timing of orogenic events in Connemara, western
37 Ireland with NE Scotland suggest that shallow slab breakoff occurred in both
38 localities.

39

40 **INTRODUCTION**

41

42 The Caledonides of Britain and Ireland have inspired numerous studies, many of
43 fundamental importance, seeking to understand orogenic processes. Central to this
44 aim is providing a robust geochronological framework to test prospective
45 tectonothermal models. There is a large geochronological database for the Grampian
46 Event of the Caledonian Orogeny (referred to as the Grampian Event from now)
47 based on the ages of metamorphic minerals (*in situ* and detrital) and syn- and post-
48 orogenic intrusions (Baxter et al., 2002; Dewey 2005; Friedrich et al., 1999; Oliver et
49 al., 2001, 2008; Viete et al., 2013). Despite this considerable geochronological
50 framework, which is accompanied by detailed field, geochemical and isotopic studies
51 that have spawned a plethora of plate tectonic models, the causes of the rapid,
52 synchronous, Grampian orogenic peak remain enigmatic (Ague & Baxter, 2007;
53 Chew & Strachan, 2013). Here we present a multi-chronometer study of an
54 Ordovician sub-volcanic intrusion at Souter Head near Aberdeen, which is emplaced
55 within the high grade Barrovian core of the Grampian orogen. The Souter Head
56 Complex (SHSC) was emplaced immediately following main stage deformation and

57 provides a unique opportunity to test cause-and-effect processes/relationships at the
58 termination of the Grampian Event.

59

60 The multi-chronometer ($^{40}\text{Ar}/^{39}\text{Ar}$, ^{238}U - ^{206}Pb , Re-Os) approach facilitates detailed
61 temporal framework for the SHSC. The data yield insights into structural changes
62 associated with the termination of the Grampian Event and through comparison with
63 numerical simulations and modern-day subduction zones, highlights that shallow slab
64 breakoff (50-100 km) explains the synchronicity of events occurring across the
65 subduction zone in Ireland and Scotland.

66

67 **GEOLOGICAL BACKGROUND**

68

69 Neoproterozoic-Cambrian Dalradian sediments were deposited on the passive
70 margin of the Laurentian continent and deformed and metamorphosed during the
71 Grampian Event following a continent-arc collision (Chew & Strachan, 2013 and
72 references therein). Below we summarise the sequences of events in terms of the
73 onset of orogenesis, the timing of deformation and the termination of the event.

74

75 **Onset of the Grampian event**

76

77 The rocks of the Grampian Terrane are continuous between NE Scotland and
78 Connemara, western Ireland (Figure 1) and are likely the telescoped end-result of
79 contraction of a passive margin. A maximum age for the start of the Grampian event
80 was suggested using the age (ca. 478 Ma) of the youngest deformed Dalradian
81 sedimentary rocks (assuming the Dalradian extends into the Ordovician, Tanner,
82 2014 and references therein). A minimum pre-early Silurian age is demonstrated in
83 Connemara, where Upper Llandovery strata rest unconformably on Dalradian
84 sediments (Soper et al., 1999; McKerrow & Campbell, 1960). The ca. 478 Ma age of

85 Tanner (2014) may well be correct but this constraint does not preclude deformation
86 having been initiated further outboard in the subduction zone significantly earlier.

87

88 Ophiolites (dismembered and ophiolites *sensu stricto*), located at Unst, Bute, Tyrone
89 and Clew Bay (Spray & Dunning, 1991; Chew et al., 2010; Crowley & Strachan,
90 2015) (Figure 1A-B) are slices of oceanic-type lithosphere that formed in supra-
91 subduction zone arc-forearc environments prior to orogenesis, and were
92 subsequently obducted onto a colliding passive margin. As such, they record the
93 initial stages in the closure of the Iapetus Ocean (e.g., Chew et al., 2010) and
94 determination of accurate (cooling)-ages for these ophiolites would provide additional
95 constraints on the timing of onset of deformation. In comparison to Tanner (2014)
96 these ages should pre-date ca. 478 Ma. Ages obtained from the metamorphic soles
97 of these ophiolites suggest that obduction was initiated at 490 ± 4 (Clew Bay, Chew
98 et al., 2010), 492 ± 1 (Bute, Chew et al., 2010), 484 ± 4 (Unst, Crowley & Strachan,
99 2015), and 492 ± 3 Ma (Unst, Spray & Dunning, 1991). There is also an age
100 constraint (477.6 ± 1.9 Ma, Sm-Nd garnet) for the ophiolite at Ballantrae (Stewart et
101 al., 2017). However, this site (Figure 1B), which is located on the opposite side of the
102 Midland Valley Arc to the other ophiolite complexes, potentially relates to a different
103 and younger phase of the collision event and thus the data are not considered
104 further. Recently, Johnson et al. (2017) proposed the existence of an island arc that
105 may be temporally associated with ophiolite obduction. However, the large age
106 uncertainties reported by Johnson et al. (2017, $\pm 8-9$ Ma) and scatter in the data
107 mean the relationship, if any, of this island arc to the onset of the Grampian event is
108 unclear.

109

110 Although locally the onset of the Grampian Event (and any orogenic collision) was
111 likely to have been diachronous, large age uncertainties (e.g., in excess of 2 Ma)
112 mean that we currently lack the temporal resolution to dissect the evolution of the

113 orogenesis. Therefore the best age for the onset of the Grampian Event is calculated
114 by taking the weighted average of the ophiolite age constraints (Clew Bay, Bute,
115 Unst) and accounting for the scatter in the data by reporting the age with an
116 uncertainty that is multiplied by the square-root of the mean square weighted
117 deviates (MSWD, or reduced chi-squared). This approach suggests that Grampian
118 Event deformation commenced at 491 ± 2 Ma.

119

120 **Grampian deformation and magmatism**

121

122 Four phases of deformation are commonly recognised in NE Scotland (D1 to D4) of
123 which the first three include the main compressional and nappe-building phases
124 (Chew & Strachan 2013). Multiple phases of deformation are found in the various
125 Dalradian inliers in Ireland but these cannot be correlated accurately with NE
126 Scotland. Barrovian style metamorphism is found throughout this sector. Buchan
127 style metamorphism is restricted to the Buchan block in NE Scotland and southern
128 Connemara, in conjunction with syn- and post-orogenic intrusions (granites and
129 voluminous basic intrusions) (Chew & Strachan 2013). Amongst these intrusions are
130 foliated gabbros and granites (e.g., Inch and Strichen), which attest to emplacement
131 pre- or syn-deformation. The intrusions yield similar U-Pb and Ar/Ar radioisotopic
132 ages for peak regional metamorphism and deformation at ca. 475-470 Ma (Kneller
133 and Aftalion 1987; Friedrich et al., 1999; Dempster et al., 2002; Oliver et al 2008).
134 High-grade pelites also yield Ar/Ar and U-Pb ages for peak metamorphism in this
135 range (Viète et al., 2013; Vorhies et al., 2013).

136

137 In the Glen Clova area of Scotland (Figure 1C) younger radioisotopic (e.g., Sm-Nd
138 garnet) ages suggest continued metamorphism and deformation to 464.8 ± 2.7 Ma
139 (2-sigma, analytical precision) (Baxter et al., 2002; Vorhies et al., 2013; Viète et al.,
140 2013). However, this datum (Baxter et al., 2002) is the rim age of one garnet derived

141 from the weighted subtraction of an 'inferred' garnet core age from a measured bulk
142 garnet age. This model date likely reflects mixing and requires an unlikely
143 assumption of constant Nd concentration across the duration of garnet growth (ca. 8
144 Ma) and as such we do not consider this age constraint further. Baxter et al. (2002)
145 reported a bulk garnet age of 466.8 ± 1.9 Ma (2-sigma, analytical precision). This age
146 with associated decay constant uncertainty incorporated to allow for inter-
147 chronometer comparison is 466.8 ± 3.2 Ma and we contend this is the most accurate
148 minimum age constraint for syn- to slightly post-D3 deformation at Glen Clova (based
149 on textural analyses of McLellan 1985, 1989).

150

151 **Termination of the Grampian Orogeny**

152

153 The ages of unfoliated intrusions (post-deformation) currently provide the best
154 constraints for the termination of orogenesis, including the Oughterard granite in
155 western Ireland (463 ± 3 Ma, Friedrich et al., 1999), the Kennethmont granite in NE
156 Scotland (457 ± 1 Ma, Oliver et al., 2000) and an undeformed quartzo-feldspathic
157 pegmatite (474 ± 5 Ma) at Portsoy also in NE Scotland (Carty et al., 2012) (Figure
158 1B). The non-foliated Cove granite (458 ± 5 Ma) and the Nigg Bay granite (465 ± 5
159 Ma) are also located in NE Scotland (Appleby et al., 2010).

160

161 **THE SOUTER HEAD SUB-VOLCANIC COMPLEX (SHSC)**

162

163 The SHSC is emplaced in metasedimentary rocks of the Dalradian Aberdeen
164 Formation (Southern Highland Group) on the coast between Aberdeen and Findon
165 (Figure 1B, D), 10 km south of the foliated Aberdeen Granite (470 ± 2 Ma, Kneller &
166 Aftalion, 1987). The Formation is well exposed along this stretch of coastline
167 whereas inland, exposure is generally poor (Munro 1986). The SHSC is also on the
168 southern edge of a granite vein complex occupying a large area south of the Dee

169 fault (Figure 1D). This fault separates the complex from the foliated Aberdeen
170 Granite (Kneller & Aftalion, 1987).

171

172 Much of the granite in the complex is migmatitic and typically occurs as lenses and
173 sheets at the cm- to 10 m-scale. Larger bodies do occur and the weakly foliated
174 (magmatic foliation) granite at Nigg Bay (Figure 1D) has been dated at 465 ± 5 Ma
175 (Appleby et al., 2010). The unfoliated Cove granite to the south of the complex is
176 dated at 458 ± 5 Ma (Figure 1D) (Appleby et al., 2010). Kneller and Aftalion (1987)
177 distinguished granite veins in the complex ranging in structural age from pre D3 or
178 syn D3 to post D3 and concluded that (1) the foliated Aberdeen granite is 'broadly'
179 syn D3, and (2) that the veins and larger bodies of granite represent a period of
180 intrusion that overlapped D3. Unfoliated granites in the complex are therefore post
181 D3. The geochronological age of the SHSC at ca. 469 Ma is discussed in this
182 structural and magmatic context.

183

184 The SHSC lies in the sillimanite zone of Barrovian metamorphism (Figure 1C)
185 (Kneller and Gillen, 1987). The metamorphic grade increases systematically from the
186 Highland Boundary Fault (HBF) northwards. Peak metamorphism (sillimanite grade)
187 is reached at Findon 4 km south of the SHSC (Harte et al., 1987) and Munro (1986)
188 records sillimanite as being widespread in the Aberdeen Formation. Sillimanite is
189 reported also in the syn D3 Aberdeen granite (Mackie 1926). Thus, we conclude that
190 the SHSC host rocks are within the sillimanite zone.

191

192 Exposure of the SHSC reveals a multistage history of repeated intrusion, breccia
193 formation, hydrothermal activity, mineralisation and faulting (Rice & Mark, this issue).
194 Two-mica granites and intrusive breccia are the dominant rock types, with minor
195 pegmatite, quartz porphyry, felsite and dolerite rocks. There is an inner sequence
196 (described by Porteous, 1973) separated by faults from two previously un-described

197 outer granites (Burnbanks and Bunstane, Figure 2). In addition, there is widespread
198 quartz veining, associated hydrothermal alteration and localised molybdenite
199 mineralisation. The SHSC has been interpreted as sub-volcanic and, in the absence
200 of a significant foliation, temporally linked to the Silurian-Devonian Newer Granites
201 (Porteous, 1973; Kneller and Gillen, 1987) that span the period of late Caledonian
202 orogenic convergence and uplift (Strachan et al., 2002; Oliver et al., 2008).

203

204 The relative timing of crystallisation for the members of the inner sequence of the
205 SHSC can be established from intrusive relationships to be from oldest to youngest:
206 (1) intrusive breccia, (2) two mica granites, (3) pegmatite, (4) quartz porphyry and (5)
207 most quartz veins. Felsite dykes are coeval with the SHSC and dolerite is younger,
208 but these intrusive rocks occur regionally and are not genetically linked to the SHSC.

209

210 Intrusive breccia occurs as three main masses separated by granite (Figure 2). Rare
211 original contacts show that granite intrudes the breccia. Breccia clasts are mainly
212 angular semi-pelite with rare rounded granite. Maximum clast dimensions are
213 typically 10-20 cm but can range up to 30 m. The northern granite mass contains
214 xenoliths of semi-pelite and rare amphibolite and exhibits a weak and patchy foliation
215 defined by alignment of biotite grains. The foliation is interpreted as magmatic since
216 the biotite is enclosed by non-aligned minerals with an igneous texture (Paterson et
217 al., 1989). In contrast, the southern mass is non-xenolithic and lacks any foliation (as
218 do all other units in the inner sequence) indicating emplacement post-D3
219 deformation. The xenolithic granite is interpreted as the marginal facies of the non-
220 xenolithic granite, which likely explains the foliation.

221

222 Pegmatites cut the two granites and breccias and are composed of quartz, K-
223 feldspar, muscovite and biotite grains. They occur mainly as linear veins that extend
224 up to 70 m along strike. With the exception of the dolerite, quartz veins cut all of the

225 intrusive rocks. The quartz veins are generally straight-sided, massive and can be
226 traced for up to 130 m. They mostly strike N-S and are either vertical or dip easterly
227 at a shallow angle. One of these veins that extends for over 50 m and cuts the
228 breccia, non-xenolithic granite and quartz porphyry contains thin margin parallel
229 bands and clusters of intergrown muscovite and molybdenite (Figure 3).

230

231 **Structural age of the SHSC**

232

233 The structural age of the SHSC cannot be obtained by examination of the contacts
234 between the metasediments and the outer granites or the inner sequence due to lack
235 of exposure or accessible exposure (Rice and Mark, 2019, this volume). However,
236 constraints can be placed upon it by (1) assuming that N-S striking quartz veins
237 cutting metasediments north of Souter Head are the same age as similarly orientated
238 veins in the SHSC, (2) examining truncated structures in clasts and xenoliths in the
239 SHSC, and (3) comparing the structures in the SHSC with those in better exposed
240 areas (Appendix DM1).

241

242 Quartz veins: N-S striking quartz veins cut the metasediments in the Altens Haven
243 area and are demonstrably post D1. Since they lack significant deformation they
244 were emplaced late in the structural sequence and are probably post D3. If these
245 veins are related to the late N-S striking veins in the SHSC, the latter are also likely
246 post D3.

247

248 Clasts and xenoliths in the SHSC: The structural age of the SHSC must be younger
249 than any structures seen in clasts and xenoliths that are truncated at the margins.
250 Most clasts and xenoliths are semi pelites like the country rocks and possess a fabric
251 identical in character to the country rocks (Appendix DM1). In keeping with the

252 general lack of folding in the host rocks the fabric is planar, even in the 30 m rafts.

253 There is rare cm scale folding of this fabric placing the SHSC as post D2 or D3.

254

255 Comparison with other areas: The absence of a tectonic foliation suggests the SHSC

256 is of a comparable structural age (post D3) to the late unfoliated granite veins

257 elsewhere in the granite vein complex which cut D3 structures (Kneller and Aftalion

258 1987). The other evidence presented above is consistent with this conclusion.

259

260 **SAMPLES**

261

262 $^{40}\text{Ar}/^{39}\text{Ar}$, ^{238}U - ^{206}Pb and Re-Os dating were used to construct a chronological

263 framework for the inner sequence and an outer granite of the SHSC (Figure 2)

264 together with a late molybdenite-bearing quartz vein (Figure 3). The chronology for

265 the inner sequence, which lacks a tectonic foliation, provides a chronological marker

266 for the termination of peak metamorphism and deformation in the Grampian Event in

267 northeast Scotland (Rice & Mark, 2019). Further, the chronology, integrated with the

268 structural setting and metamorphic grade, permits interrogation of subduction zone

269 response (i.e., topographical change, exhumation) during the termination of the

270 Grampian Event. Such investigations are possible due to the excellent field relations

271 exposed in the Souter Head coastal transect, which provide a robust petrogenetic

272 framework and a wide variety of dating targets. Specifically, we targeted muscovite

273 from the outer Burnbanks granite (sample location 4, Figure 2), zircon and muscovite

274 from the inner non-xenolithic granite (sample location 1, Figure 2), K-feldspar and

275 muscovite from pegmatite cutting the non-xenolithic granite (sample location 2,

276 Figure 2), and finally intergrown and demonstrably coeval muscovite and

277 molybdenite from the late quartz-molybdenite vein (sample location 3, Figure 2,

278 Figure 3C).

279

280 **Petrography of the late muscovite-molybdenite-bearing quartz vein**

281

282 The dominant mineral is quartz of which three types can be distinguished in
283 cathodoluminescence (CL) (Figure 4). The most common and earliest is medium
284 grey in CL. This has recrystallised to grains in the size range (0.1 - 5.0 mm). In
285 places primary oscillatory zoning is preserved (Figure 4). A network of dark quartz,
286 which follows the grain boundaries and fills fractures, postdates this quartz (Figure
287 4). The third and latest is margin parallel bright quartz veinlets. These, and fracture-
288 controlled inclusion trails, both crosscut the network (Figure 4).

289

290 Molybdenite shows a close spatial and temporal relationship with muscovite. The two
291 minerals are mainly found in clusters with muscovite and molybdenite crystals up to 2
292 mm and 0.4 mm, respectively. In the clusters molybdenite occurs between muscovite
293 crystals but also crosscutting crystals and along muscovite cleavages (Figure 4). The
294 siting of the clusters is closely linked to the dark quartz network (Figure 4). Lesser
295 amounts of molybdenite and muscovite are found in margin parallel veinlets (Figures
296 4) and dispersed in the dark quartz network. In these last two locations, the
297 molybdenite and muscovite are finer grained (typically up to 150 μm). The close
298 spatial association of molybdenite with muscovite within the dark quartz network
299 suggests all three minerals are broadly coeval.

300

301 **METHODS**

302

303 $^{40}\text{Ar}/^{39}\text{Ar}$ dating: Samples for $^{40}\text{Ar}/^{39}\text{Ar}$ dating were prepared using the methodologies
304 outlined in Mark et al. (2011). Briefly, samples were crushed and subjected to
305 magnetic separation. The sericite-bearing fraction was run down a shaking table and
306 relatively pure muscovite splits collected. K-feldspar samples were purified using
307 heavy liquids and cleaned by leaching in 5% HF for 2 minutes. Subsequently clean

308 grains (no visible inclusions) were hand-picked under a binocular microscope with all
309 samples (wafers and separates) further cleaned in ethanol and de-ionised water.

310

311 Samples were parcelled in high purity Al discs for irradiation. Standards Fish Canyon
312 sanidine (FCs-EK, Morgan et al., 2014) (28.294 ± 0.036 Ma, Renne et al., 2011),
313 GA1550 biotite (99.738 ± 0.104 Ma, Renne et al., 2011) and Hb3gr (1081.0 ± 1.2 Ma,
314 Renne et al., 2011) hornblende were loaded adjacent to the samples to permit
315 accurate characterisation of the neutron flux (J-parameter). Samples were irradiated
316 for 2,700 minutes in the Cd-lined facility of the CLICIT Facility at the Oregon State
317 University TRIGA reactor. Standards were analysed on a MAP 215-50 system
318 (described below briefly and in more detail by Mark et al., 2011) – FCs was analyzed
319 by CO₂ laser total fusion as single crystals (n = 20), GA1550 (n = 5) and Hb3gr (n =
320 5) were step-heated using a CO₂ scanning laser (e.g., Barfod et al., 2014). Using
321 GA1550 the J-parameter was determined to a precision c. 0.1% uncertainty. Using
322 the J-parameter measurements from GA1550 ages were determined for FCs and
323 Hb3gr. The ages overlapped at the 68% confidence (1-sigma) with the ages reported
324 by Renne et al. (2011), showing the J-parameters determined from GA1550 to be
325 accurate.

326

327 The samples were step-heated using a CO₂ laser (approximately 500-1,500 °C,
328 optical pyrometer measurements). Extracted gases were subjected to 300 seconds
329 of purification by exposure to two SAES GP50 getters (one maintained at room
330 temperature, the other held at c. 450 °C). A cold finger was maintained at -95.5 °C
331 using a mixture of dry ice (CO_{2(S)}) and acetone. Ion beam intensities (i.e., Ar isotope
332 intensities and hence ratios) were measured using a GVI ARGUS V noble gas mass
333 spectrometer in 'true' multicollection mode (Mark et al., 2009). Faraday cups (10^{11}
334 ohm ⁴⁰Ar, 10^{12} ohm ³⁹⁻³⁶Ar) were used to make measurements. The system had a
335 measured sensitivity of 7.40×10^{-14} moles/Volt. The extraction and clean-up, as well

336 as mass spectrometer inlet and measurement protocols and data acquisition were
337 automated. Backgrounds (full extraction line and mass spectrometer) were made
338 following every two analyses of unknowns. The average background \pm standard
339 deviation ($n = 162$) from the entire run sequence was used to correct raw isotope
340 measurements from unknowns and air pipettes. Mass discrimination was monitored
341 by analysis of air pipette aliquots after every five analyses of unknowns ($n = 63$, 7.32
342 $\times 10^{-14}$ moles ^{40}Ar , $^{40}\text{Ar}/^{36}\text{Ar} = 299.81 \pm 0.19$).

343

344 All Ar isotope data were corrected for backgrounds, mass discrimination, and reactor-
345 produced nuclides and processed using standard data reduction protocols (e.g.,
346 Mark et al., 2005) and reported according to the criteria of Renne et al. (2009). The
347 atmospheric argon isotope ratios of Lee et al. (2006), which have been independently
348 verified by Mark et al. (2011), were employed. The ages were calculated using the
349 optimisation model approach of Renne et al. (2010) using the parameters of Renne
350 et al. (2011). The $^{40}\text{Ar}/^{39}\text{Ar}$ ages are reported as $X \pm Y/Z$ where Y is the analytical
351 uncertainty and Z is the full external precision, including uncertainties from the decay
352 constant. All ages are reported at the 2 sigma confidence interval.

353

354 Isotope dilution thermal ionisation mass spectrometry (ID-TIMS) U-Pb
355 geochronology: zircons were hand-picked after separation using conventional
356 techniques. Analyses were performed at the NERC Isotope Geosciences Laboratory
357 (NIGL) at the British Geological Survey, Keyworth, United Kingdom following
358 established protocols (e.g., Noble & Condon et al., 2014, Noble et al., 2014). This
359 includes a chemical abrasion procedure (Mattinson, 2005) and U/Pb determinations
360 calibrated using the EARTHTIME (ET535) tracer solution (Condon et al., 2015, CA-
361 ID-TIMS). For data reduction and uncertainty propagation, we followed the strategy
362 of Bowring et al. (2011) and McLean et al. (2011).

363

364 As we are not dealing with geologically 'young' rocks and thus our data will not be
365 precise enough to concern ourselves with 'over-interpretation' of the zircon U-Pb age
366 data (i.e., youngest zircon versus weighted mean age; Ickert et al., 2015, Mark et al.,
367 2017), we used a weighted mean of the youngest population of each sample. Each
368 youngest population contained three or more ages that give an MSWD that is
369 acceptable for a single population (Wendt and Carl, 1991). The $^{206}\text{Pb}/^{238}\text{U}$ ages
370 presented in this paper are corrected for initial Th disequilibrium and uncertainties
371 are quoted at the 2 sigma confidence level, unless stated otherwise. Uncertainties
372 are listed as $\pm X/Y/Z$, where X is the analytical uncertainty, with Y and Z including the
373 propagated uncertainties for tracer calibration, and respectively tracer calibration and
374 the ^{238}U decay constant.

375

376 Re-Os dating: Three molybdenite separates were obtained. Two independent mineral
377 separates were isolated using traditional mineral separation protocols, e.g., crushing,
378 magnetic Frantz separation, heavy liquids, water floatation and hand picking (Selby
379 and Creaser, 2004). The mineral separates of samples SH23A and SH23B were
380 achieved utilising the HF isolation approach (Lawley and Selby, 2012). The latter
381 uses concentrated HF at room temperature to aid in liberating the molybdenite from
382 the silicate matrix.

383

384 The Re-Os analytical protocol follows that described by Selby and Creaser (2001),
385 with a slight modification to the isolation protocol of Re. An aliquot of molybdenite
386 doped with a known amount of tracer solution comprising ^{185}Re and normal Os
387 isotope composition was loaded into a carius tube with a 1:3 mL mix of concentrated
388 HCl and HNO_3 . The tube was sealed and then heated to 220 °C for 24 hours. The Os
389 was isolated from the acid solution using solvent extraction with CHCl_3 and further
390 purified using micro-distillation. The Re was isolated using solvent extraction by
391 NaOH and Acetone, and then further purified using anion $\text{HNO}_3:\text{HCl}$

392 chromatography.

393

394 The isotope compositions of the Re and Os fractions were determined using
395 Negative Thermal Ionisation Mass Spectrometry (N-TIMS – Creaser et al., 1991;
396 Volkening et al., 1991) using a Thermo Electron TRITON mass spectrometer at the
397 University of Durham. Measurements were made statically using the Faraday Cups
398 for both Re and Os. The measured Re and Os isotopic ratios were oxide corrected
399 offline. The data were corrected for fractionation. Analytical uncertainties are
400 propagated and incorporate uncertainties related to Re and Os mass spectrometer
401 measurements, blank abundances and isotopic compositions, spike calibrations and
402 reproducibility of standard Re and Os isotope values. Procedural blanks conducted
403 during the period of the molybdenite analysis are negligible relative to the Re and Os
404 abundances measured in the samples (Re 2.1 ± 0.2 ppt, Os 0.1 ± 0.2 ppt,
405 $^{187}\text{Os}/^{188}\text{Os} = 0.22 \pm 0.05$; $n = 2$). In-house reference solutions run during the analysis
406 (Re std = 0.5983 ± 0.0011 ; DROsS = 0.16089 ± 0.0001 ; $n = 2$) are similar to long-
407 term reproducibility data reported by Lawley and Selby (2012) (and references
408 therein). The Re-Os ages are presented as model ages from the simplified isotope
409 equation [$t = \ln(^{187}\text{Os}/^{187}\text{Re} + 1)/\lambda$, where t = model age, and λ = ^{187}Re decay
410 constant] and assumes no initial radiogenic Os. Inclusion of decay constant
411 uncertainty and reporting of data with 2 sigma uncertainty allows for direct
412 comparison of the Re-Os ages with the $^{206}\text{Pb}/^{238}\text{U}$ and $^{40}\text{Ar}/^{39}\text{Ar}$ ages. The Re-Os
413 ages are provided as $X \pm Y/Z$ with Y and Z with and without the decay constant
414 uncertainty, respectively.

415

416 Appendix DM2 contains raw age data. Note, all age data throughout are reported at
417 the 2-sigma confidence level. Also, all published data have been recalculated (where
418 relevant) to the latest decay constants and monitor ages/spike calibrations.

419

420 **RESULTS**

421

422 **Inter-chronometer comparison**

423

424 It is important to note that when considering the relative timing of different units using
425 a single chronometer, only the analytical uncertainty is required as mineral standard
426 age uncertainties, tracer calibration and decay constant uncertainties are all
427 systematic, and have a predictable and similar effect on each sample. The age
428 standard, tracer and decay constant uncertainties, combined, yield the 'total'
429 uncertainty and this is used when comparing data from different chronometers. All
430 inter-chronometer comparisons throughout this contribution are made at the 2-sigma
431 (95.4 %) confidence interval and incorporate systematic uncertainties.

432

433 The weighted means (single zircon ID-TIMS) for the $^{207}\text{Pb}/^{206}\text{Pb}$ and $^{206}\text{Pb}/^{238}\text{U}$ ages
434 are $469.4 \pm 0.8/0.8/4.6$ Ma (analytical precision/tracer calibration/decay constant
435 uncertainties) and $469.1 \pm 0.1/0.2/0.6$ Ma, respectively (Figure 5). We interpret the
436 $^{206}\text{Pb}/^{238}\text{U}$ age to constrain crystallisation of the unfoliated non-xenolithic granite.

437

438 All $^{40}\text{Ar}/^{39}\text{Ar}$ age spectra (Figure 5) show, to varying degrees, a ca. 405 Ma
439 disturbance of the low temperature steps. With the exception of the K-feldspar from
440 the pegmatite all age spectra step-up to define robust plateau ages. The outer
441 Burnbanks Granite (muscovite) is $468.7 \pm 0.3/0.4$ Ma (analytical precision/decay
442 constant uncertainties), the inner unfoliated non-xenolithic granite (muscovite) is
443 $468.4 \pm 0.3/0.4$ Ma, the pegmatite (muscovite) is $468.6 \pm 0.7/0.7$ Ma, the late vein
444 (muscovite) that the cuts granite, breccia and porphyry rocks is $468.3 \pm 0.6/0.7$ Ma.
445 The K-feldspar data step-up also from ca. 405 Ma to define a mini-plateau age (468.2
446 $\pm 0.3/0.4$ Ma) that is indistinguishable from the age of the muscovite from the same
447 rock. All $^{40}\text{Ar}/^{39}\text{Ar}$ ages are indistinguishable from each other and the $^{206}\text{Pb}/^{238}\text{U}$

448 zircon age. The close temporal association between the U-Pb zircon and the $^{40}\text{Ar}/^{39}\text{Ar}$
449 muscovite ages shows that the SHSC, including late hydrothermal activity, was
450 emplaced and cooled very quickly, within 0.5 ± 0.9 Ma (possibly due to rapid uplift,
451 see discussion below). The reproducible ages (ca. 469-468 Ma) for the outer
452 Burnbanks granite and the SHSC together with petrographic similarities and the
453 symmetrical position of the outer granites on the northern and southern sides of the
454 complex (Figure 2), strongly suggest that they are part of the SHSC.

455

456 A molybdenite sample from the late quartz-molybdenite vein that presents as coeval
457 with the muscovite (Figure 4) (Table 1) defines a model age of $440.2 \pm 3.1/3.4$ Ma
458 (analytical precision and tracer calibration/decay constant uncertainties). Although
459 there is excellent agreement between the $^{40}\text{Ar}/^{39}\text{Ar}$ and U-Pb systems (Figure 5-6),
460 the Re-Os age is surprisingly not concordant with the other chronometers (Table 1).
461 We do not consider the Re-Os age to be geologically robust with respect to an
462 emplacement age (see discussion below) owing to the later alteration of the
463 molybdenite to powellite, a mineral found in the oxidation zones of molybdenite-
464 bearing hydrothermal deposits (Anthony et al., 2003).

465

466 **DISCUSSION**

467

468 **Re-Os chronology**

469

470 The young Re-Os age (Table 1, Figure 6) was unexpected given the relatively high
471 closure temperature (> 500 °C, certainly higher than $^{40}\text{Ar}/^{39}\text{Ar}$ muscovite) associated
472 with the Re-Os molybdenite system – suggesting either the molybdenite age is ‘reset’
473 or the molybdenite was emplaced significantly later than the SHSC. Temperatures
474 required to thermal reset the molybdenite would have completely reset the $^{40}\text{Ar}/^{39}\text{Ar}$
475 geochronometer and as such, we would not have recovered ages for the vein

476 muscovite equivalent to the U/Pb age of the granites. The textural evidence indicates
477 that the molybdenite and muscovite have a close spatial association/coeval (see
478 discussion above) (Figures 4). Therefore, a younger molybdenite emplacement event
479 would require the muscovite formed coevally with the molybdenite to be younger than
480 the granites, but the muscovite fraction dated (grainsize 50 to 100s μm) is not
481 temporally resolvable from the age of the granites. In an attempt to resolve this
482 conundrum we re-examined the samples by Scanning Electron Microscopy, which
483 revealed the presence of powellite.

484

485 Powellite (calcium molybdate) in our samples is closely associated with molybdenite
486 and best developed in the clusters and to a lesser extent in the margin parallel
487 veinlets (Figure 7). There is evidence that powellite was formed during a separate
488 and later event to molybdenite. Thus, ragged molybdenite crystals occur floating in
489 powellite, which also penetrates along molybdenite cleavages supporting a replacive
490 relationship (Figure 7). Commonly, molybdenite crystals or terminations of crystals
491 enclosed by quartz are not coated by powellite (Figure 7), which is consistent with
492 quartz protecting molybdenite from alteration by later fluids. Powellite is present by
493 itself in fracture fills in muscovite (Figure 7). Further petrographic evidence for a later
494 tectono-hydrothermal event, possibly linked to powellite deposition, are margin
495 parallel bright quartz veinlets and fracture-controlled inclusion trails cutting the dark
496 quartz network (Figure 7).

497

498 We therefore propose an alternate scenario, which suggests caution must be
499 employed if using the Re-Os molybdenite-dating tool (model ages) in isolation of
500 other chronometers in settings that have experienced multiple magmatic and
501 hydrothermal events. The alteration of molybdenite to powellite, has resulted in
502 (owing to bulk dissolution/sampling approaches) a two-component hybrid age that
503 has no geological significance. This is supported by the $^{40}\text{Ar}/^{39}\text{Ar}$ step-heating data

504 recording an alteration/fluid flushing event at ca. 405 Ma (Figure 5-6), which is likely
505 linked to the emplacement of the nearby Mt. Battock granite of equivalent age (Oliver
506 et al., 2008). Hydrothermally driven fluid flushing events (e.g., Mark et al., 2007) likely
507 formed the powellite whilst also disturbing the Ar-systematics of the low closure
508 temperature minerals (e.g., feldspar), producing the 405 Ma overprint. The primary
509 molybdenite age (ca. 468 Ma) mixed in the correct portions with a younger powellite
510 age (ca. 405 Ma), would yield a 438-441 Ma hybrid age. Note, there is no known
511 thermal event that is coincident with the ca. 440 Ma model Re-Os age in the area of
512 study.

513

514 Although powellite has been found in other systems a disturbance of the Re-Os ages
515 has not been reported, which is likely because the powellite formed soon after
516 molybdenite deposition; the molybdenite age uncertainties would thus mask any
517 disturbances to the Re-Os system (the delta-time between the molybdenite and
518 powellite is small, whereas at Souter Head, the delta-time between the molybdenite
519 formation and subsequent powellite formation is large). The powellite at Souter Head
520 formed much later than the molybdenite, beyond the uncertainties of the molybdenite
521 Re-Os age as suggested by the young (ca. 405 Ma) $^{40}\text{Ar}/^{39}\text{Ar}$ age. Several studies
522 have conducted experiments to establish the effect of such alteration on the Re-Os
523 chronometer (e.g., Suzuki et al., 2000) with much discussion and debate (e.g., Selby
524 et al., 2004; Suzuki, 2004). Suzuki et al. (2000) showed experimentally that the Re-
525 Os system in molybdenite can behave as an open-system in the presence of
526 aggressive advecting fluids. □□□

527

528 **The age of the SHSC**

529

530 The ages obtained for the SHSC show that it belongs to the Ordovician late tectonic
531 granites (ca. 475-457 Ma) of the NE Grampian Terrane (Strachan et al., 2002; Oliver
532 et al., 2008) rather than the Silurian-Devonian Newer Granites (Figure 8). The former
533 are mostly S-type two-mica granites, garnet-bearing and commonly foliated with
534 $^{87}\text{Sr}/^{86}\text{Sr}$ ratios consistent with the melting of sedimentary protoliths (Chappell &
535 White, 1974; Harmon 1983). Infracrustal sources may also be involved (Appleby et
536 al., 2010).

537

538 Stages of deformation associated with the Grampian Event in NE Scotland are D1-3
539 and, locally, D4 (Harte et al., 1984; Kneller, 1987; Strachan et al., 2002). The SHSC
540 intrudes the Barrovian metamorphic core (sillimanite zone) in this area (Figure 1C)
541 where peak metamorphism is closely associated with D3 (Harte et al., 1984;
542 McClellan, 1989). The nearby-foliated Aberdeen granite is broadly D3 and has been
543 dated at 470 ± 2 Ma (Kneller and Aftalion, 1987) (Figure 1D). The SHSC is similar in
544 terms of mineralogy to these granites but critically, lacks a tectonic foliation. The
545 geochronology, absence of a tectonic foliation and widespread evidence of brittle
546 rather than ductile deformation in the SHSC as well as the absence of high grade
547 indicator minerals and significant recrystallisation, shows the SHSC immediately
548 post-dates and thus constrains the end of main (D1-3) Grampian deformation and
549 metamorphism in the NE Grampian terrane to 469.1 ± 0.6 Ma (Figure 3).

550

551 **Termination and duration of the Grampian Event.**

552

553 The post-deformation SHSC age is indistinguishable from garnet Sm-Nd ages that
554 place the end of D3 at Glen Clova at 466.8 ± 3.2 Ma (2-sigma, full external
555 uncertainties, Baxter et al., 2002) (Figure 1B). Similarly, the SHSC age is coincident
556 with the termination of deformation to the north at Portsoy, as constrained by an
557 undeformed pegmatite at 474 ± 5 Ma (Carty et al., 2012).

558

559 Given that the ages for the SHSC, Glen Clova and Portsoy are indistinguishable at
560 the 2-sigma confidence interval we have calculated a weighted average of the
561 termination of D3 deformation and peak metamorphism in NE Scotland of 469.2 ± 1.3
562 Ma. Thus, the duration of the Grampian Event at best can be confined to 22.8 ± 2.4
563 Ma, from the onset of collision (i.e., ophiolite obduction) to the termination of
564 Grampian Event D3 deformation. A later phase of deformation D4 is developed
565 locally north of the Highland Boundary Fault but the age of this event is currently
566 unconstrained. It has been linked to late stage uplift of the orogen (Harte et al, 1984).

567

568 **Termination and rapid uplift**

569

570 There is direct evidence that rapid uplift was temporally associated with the
571 termination of the orogenic peak in NE Scotland. The SHSC was emplaced in
572 sillimanite zone rocks in the upper crust (above ca. 10 km) as demonstrated by the
573 presence of porphyritic rocks and widespread evidence of brittle fracture – i.e.,
574 intrusive breccias with angular clasts and parallel-sided quartz and pegmatite veins
575 (Seedorf et al., 2005).

576

577 A further estimate of depth was obtained using the normative quartz and albite plus
578 orthoclase barometers (Yang, 2017). We applied this method to four granitic
579 members of the Souter Head suite to constrain the depth (Appendix DM3). Three of
580 them were at the extreme limits of the calibration of the method and the results
581 should be treated with caution. However, a quartz-feldspar porphyry falls well within
582 the range of the calibration method and yields a depth estimate of 13-15 km. Overall
583 an emplacement depth in the range 10-15 km for the SHSC is indicated.

584

585 However, 0.9 ± 2 Ma prior to emplacement of the SHSC, as evidenced by the nearby
586 foliated (syn-D3) Aberdeen Granite (470 ± 2 Ma), the host rocks of the SHSC were in
587 the lower crust at ca. 20 km depth (Vorhies and Ague, 2011) experiencing high-grade
588 metamorphism under ductile conditions. Within a short time period these rocks thus
589 underwent a 5-10 km change in their structural level – requiring an exhumation rate
590 of 5-10 km/Ma, comparable to rates found in modern arc-continent collision zones
591 (Brown et al., 2011).

592

593 By ca. 469 Ma much of the metamorphic cover was removed, which is consistent
594 with high-grade orogenic debris arriving in the South Mayo Trough, Connemara and
595 Midland Valley Basin (Kirkland Conglomerate) at 465 ± 3 Ma and with the oldest
596 mica (Dalradian and detrital) cooling ages (Oliver, 2000; Oliver, 2001; Clift et al.,
597 2004; Dewey, 2005). The lag time for sediment transport to the Midland Valley basin
598 was exceptionally short, potentially 0.1 Ma, allowing for the large uncertainty
599 associated with the lower (ca. 465 Ma) bound for the Kirkland Conglomerate. These
600 data support our age for exhumation of the metamorphic core.

601

602 **Slab break-off and timing of events during the metamorphic peak**

603

604 The Grampian orogenic peak is defined by temporally overlapping D2/3 deformation
605 and peak metamorphism, basic magmatism and rapid uplift all within ca. 5 Ma, which
606 points towards a critical and abrupt change in the subduction zone (Figure 9).
607 Through comparison with numerical simulations, see below, this is best explained by
608 slab break-off, which likely occurred soon after buoyant material (a spreading ridge or
609 the Midland Valley Arc) entered the trench and stalled subduction (Oliver et al., 2008;
610 Tanner, 2014). Although slab roll-back, slab tearing or slab parallel asthenospheric
611 melting are other potential explanations, numerical simulations suggest none of these
612 mechanisms are congruent with an abrupt event that results in a structural change of

613 5-10 km within the crust within 0.9 ± 2 Ma (Menant et al., 2016; Cassel et al., 2018
614 and references within).

615

616 When slab breakoff occurs, part of a subducted lithospheric plate detaches abruptly
617 and sinks into the asthenosphere inducing upwelling. The dynamics of slab breakoff
618 has been investigated extensively, and it has been shown that the strength of the
619 subducting lithosphere, in part influenced by the oceanic slab age, convergence
620 velocity, continental crustal and lithospheric thicknesses, and the mechanism of
621 detachment, all exert control on the depth of breakoff (Andrews and Billen, 2009;
622 Duretz et al., 2011; Gerya et al., 2004). Numerical models have shown a wide range
623 in this depth, from 40 to over 500 km (Baumann et al., 2010; Duretz et al., 2011), but
624 few numerical modelling studies have quantitatively examined the topographic
625 response (rate and amount of uplift) to slab detachment. For example, Buitter et al.
626 (2002) predicted topography uplift in the range of 2 to 6 km using an elastic model,
627 whereas Gerya et al. (2004) predicted lower uplift values (< 2 km) using a visco-
628 plastic model. Analysis of ancient orogenic belts, e.g., the Grampian Event, provides
629 a time integrated picture of topographic evolution (as opposed to modern day
630 measurements in active subduction zones) that allows for connection between model
631 and real-world data.

632

633 Some 5-10 km of uplift as recorded by the SHSC over ca. 1 Ma would suggest a slab
634 breakoff depth of either less than 100 km (Buitter et al., 2002) or less than 50 km
635 (Duretz et al., 2011). Slab break-off at ca. 50 km depth would be directly under the
636 collision zone. We suggest that the ensuing rapid uplift caused crustal thinning and
637 decompressional melting of the subcrustal mantle (McKenzie & Bickle, 1989; Hole et
638 al., 2015). This depth is consistent with the shallow sourcing of basic melts to power
639 Buchan metamorphism in NE Scotland (and Ireland, see below) (Viète et al., 2013;
640 Johnson et al., 2017). Crustal thinning could be achieved by erosion and gravity

641 driven detachment faulting, as suggested for Connemara where the uplift rate is
642 similar (Clift et al., 2004). Melting of the lower crust may also have occurred by
643 decompression/invasion by basic magma to produce the syn-and post-D3 granites
644 observed throughout the region (e.g., the Aberdeen granite and the SHSC).

645

646 The precise age of slab break-off during the Grampian Event is uncertain but can be
647 constrained by the age of basic magmatism. The oldest known and precisely dated
648 synorogenic basic rock in NE Scotland is 471.3 ± 1.7 Ma (Carty et al., 2012) and in
649 Connemara ca. 474.5 ± 1 Ma (Friedrich et al., 1999). Rapid uplift likely began no later
650 than 470 Ma and may have occurred along regional shear zones such as the Portsoy
651 Shear Zone, which also controlled magma emplacement (Ashcroft et al. 1984; Carty
652 et al., 2012; Viète et al., 2013). Meanwhile, orogenic detritus accumulated in adjacent
653 sedimentary basins (Dewey & Mange, 1999; Oliver, 2001).

654

655 The above observations suggest that a switch from compressional to extensional
656 tectonics in the orogen occurred in the period 474-471 Ma and overlaps with peak
657 Barrovian and Buchan metamorphism and D2/D3 deformation. While we agree with
658 Viète et al., (2013) that regional extension was the likely tectonic setting for peak
659 metamorphism and deformation, we consider that slab break-off (Oliver et al., 2008;
660 Tanner, 2014) provides a better model to explain the extensional processes involved
661 in this very short event.

662

663 **Scotland and Connemara**

664

665 There are marked similarities in the timings and rates of specific events in NE
666 Scotland with Connemara in western Ireland: peak metamorphism and deformation
667 terminated at ca. 470-468 Ma (Friedrich et al., 1999); uplift rates at the termination of
668 the orogeny in Connemara were ca. 7 km/Ma (Friedrich and Hodges, 2016); and

669 these events are essentially coincident with intrusion of basic syn-D3 plutons (e.g.,
670 Cashel-Lough Wheelaun gabbro, 470.1 ± 1.4 Ma, Friedrich et al., 1999). Such data
671 suggest that shallow slab detachment occurred synchronously in NE Scotland and
672 Connemara. Along-strike heterogeneity in subduction zones is well known (e.g.,
673 Nazca Plate; Chen et al. 2001; Brown et al., 2011 and references therein) and it is
674 likely that slab detachment did not occur in the intervening portion of the subduction
675 zone where Barrovian metamorphism only is found. Here slab dips were mainly
676 shallow and consequently, with the exception of the Tyrone Igneous Complex, arc-
677 related igneous activity was essentially absent (Figure 1B) (Cahill & Isacks, 1992;
678 Chen et al., 2001; Cooper et al., 2011).

679

680 **CONCLUSIONS**

681

682 The Souter Head sub-volcanic complex (Aberdeenshire, Scotland) intruded the high-
683 grade metamorphic core of the Grampian orogen at 469.1 ± 0.6 Ma (^{238}U - ^{206}Pb
684 zircon). Temporally coincident U-Pb and $^{40}\text{Ar}/^{39}\text{Ar}$ data show the SHSC cooled
685 quickly. Intrusion followed closely peak metamorphism and D2/D3 deformation at ca.
686 470 Ma and marks the end of the Grampian Event in NE Scotland. Younger Re-Os
687 ages are due to post-emplacement alteration of molybdenite to powellite and
688 highlight the importance of careful petrographic characterisation of materials prior to
689 determination of model Re-Os ages from molybdenite.

690

691 D2/3 Grampian deformation (the age of D1 remains uncertain), peak metamorphism
692 (Barrovian and Buchan style) and basic magmatism in NE Scotland were
693 synchronous at ca. 470 Ma and associated with rapid uplift (5-10 km/Ma) of the
694 orogen, which largely removed the metamorphic cover by ca. 469 Ma. We suggest
695 that shallow slab breakoff (50-100 km) can explain the rapid uplift and the
696 synchronicity of these events and that decompression led to melting and generation

697 of mafic and felsic melts. This interpretation implies that peak metamorphism and
698 D2/D3 ductile deformation were associated with extension, as previously suggested.
699 Close similarities between the geological histories of NE Scotland and Connemara
700 suggest that shallow slab breakoff occurred in both areas. Our proposed model
701 explains the presence of both Buchan and Barrovian activity across various sectors
702 of the Grampian Orogen.

703

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715

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976

977 **FIGURE CAPTIONS**

978

979 **FIGURE 1:** (A) Map of the United Kingdom showing constraints from ophiolite
980 obduction on Shetland for the onset of Grampian orogenesis: 492 ± 3 and $484 \pm$
981 4 Ma (Spray & Dunning, 1991; Crowley & Strachan, 2015, respectively). (B)
982 Figure showing the timing of onset (green) and termination (red) of Grampian
983 orogenesis in Scotland and Ireland. 1: Unst ophiolite obduction, 492 ± 3 Ma
984 (Spray & Dunning, 1991) and 484 ± 4 Ma (Crowley & Strachan, 2015). 2: Clew
985 Bay ophiolite obduction, 3: 490 ± 4 Ma (Chew et al., 2010). Bute ophiolite
986 obduction, 492 ± 2 (Chew et al., 2010). The blue box shows Ballantrae ophiolite
987 obduction, 477.6 ± 1.9 Ma (Stewart et al., 2017) however, this event may not be
988 related to the Grampian event (see discussion). Figure modified from Chew et
989 al., 2010. (C) Location of Souter Head in relation to the Barrovian metamorphic
990 zones (modified from Figure 1 of Baxter et al., 2002) and the Aberdeen Granite.
991 (D) The location and ages of granites located close to Aberdeen.

992

993 **FIGURE 2:** Simplified geological map of the Souter Head complex and the sample
994 localities (1-4). Also see Rice & Mark (this issue for further information). Sample
995 location (1) zircon and muscovite from the inner non-xenolithic granite; Sample

996 location (2) K-feldspar and muscovite from pegmatite cutting the non-xenolithic
997 granite; sample location (3) intergrown and demonstrably coeval muscovite and
998 molybdenite from the late quartz-molybdenite vein; sample location (4)
999 muscovite from the outer Burnbanks granite.

1000

1001 **FIGURE 3:** Field photograph showing the targeted quartz-molybdenite vein hosted
1002 by intrusive breccia. Non-xenolithic granite can be observed in the background.
1003 Black arrows show molybdenite- and muscovite-rich grey streaks through the
1004 vein. Material recovered from sample point 3 in Figure 2.

1005

1006 **FIGURE 4:** (A) SEM-CL – typical fracture network of dark quartz in quartz vein partly
1007 following the boundaries of recrystallized quartz crystals (medium grey). Clusters
1008 of molybdenite and muscovite (white arrows) are located in the network of dark
1009 quartz, the white mineral in the clusters is powellite. Note, in SEM-CL the
1010 molybdenite and muscovite are not visible, but are shown in SEM-BSE (Figs. 3D
1011 and 4A-B). Finer lines corresponding to fluid inclusion trails are arrowed (black)
1012 and an area of primary oscillatory zoning in quartz is marked (OZ). (B) SEM-CL
1013 – medium grey quartz with network of dark quartz and margin parallel (running
1014 E-W) veinlet (MMPV) containing muscovite and molybdenite (opaque) and
1015 powellite (white). Fine lines corresponding to fluid inclusion trails (black arrow)
1016 and quartz veinlet (BQ) cut the network. (C) BSEM – margin parallel veinlet in
1017 quartz (dark grey) composed of muscovite (medium grey), molybdenite (black
1018 arrows) and powellite (white arrows). Small clusters of molybdenite and
1019 muscovite and isolated crystals of these two minerals are also present. (D)
1020 BSEM – molybdenite-muscovite cluster. Molybdenite (black arrows) is located
1021 between muscovite (medium grey) crystals and cleavages and also cross-cutting
1022 muscovite crystals and cleavages. Fine veinlets of powellite (white arrows) are
1023 present cutting muscovite.

1024

1025 **FIGURE 5:** (A) Zircon U-Pb Concordia plot. (B) Zircon U-Pb weighted average ages.
1026 (C) $^{40}\text{Ar}/^{39}\text{Ar}$ step-heating spectra showing ages for all targeted samples.

1027

1028 **FIGURE 6:** Summary of multi-chronometer geochronology (all data shown at 2-sigma
1029 including all sources of uncertainties). 1: $^{206}\text{Pb}/^{238}\text{U}$ age for zircon from the
1030 unfoliated non-xenolithic granite. 2: $^{40}\text{Ar}/^{39}\text{Ar}$ age for the Burnbanks granite. 3:
1031 $^{40}\text{Ar}/^{39}\text{Ar}$ age for inner unfoliated non-xenolithic granite. 4: $^{40}\text{Ar}/^{39}\text{Ar}$ age for the
1032 pegmatite (muscovite). 5: $^{40}\text{Ar}/^{39}\text{Ar}$ age (mini-plateau) for K-feldspar from
1033 pegmatite. 6: $^{40}\text{Ar}/^{39}\text{Ar}$ age for the late vein. 7: Re-Os age for molybdenite from
1034 late vein. 8: $^{40}\text{Ar}/^{39}\text{Ar}$ reset age range recorded by all $^{40}\text{Ar}/^{39}\text{Ar}$ samples.

1035

1036 **FIGURE 7:** (A) BSEM – molybdenite (white), minor muscovite (dark grey) and
1037 powellite (medium grey) in quartz (dark grey). Note that powellite encloses
1038 molybdenite in the cluster but not where molybdenite terminations are protected
1039 in quartz (white arrows). (B) BSEM – Molybdenite (white) and powellite (medium
1040 grey) in quartz (dark grey) with minor muscovite. Powellite enclosing
1041 molybdenite crystals of varying size and ragged appearance. Note powellite
1042 penetrating along molybdenite cleavages (white arrow). (C) BSEM – Powellite
1043 (medium grey) is enclosing ragged molybdenite crystals. (D) BSEM – powellite
1044 filling fine fractures in muscovite.

1045

1046 **FIGURE 8:** Timescale of showing the age of key metamorphic (meta.) minerals,
1047 basic rocks, unfoliated granites, foliated granites and ophiolite obduction in NE
1048 Scotland and western Ireland in relation to Dalradian sedimentation, peak
1049 orogenesis (oro.) and exhumation (exh.). Data from Baxter et al., 2002; Carty et
1050 al., 2012; Chew et al., 2010; Dempster et al., 2002; Friedrich et al., 1999; Kneller

1051 and Aftalion, 1987; Oliver et al., 2000; Stewart et al., 2017 and Viete et al., 2013.
1052 The various times of deformation (D1-D4 are also shown).

1053

1054 **FIGURE 9:** Cross-section of the Grampian orogen in NE Scotland during and
1055 immediately after slab break-off ca. 471-469 Ma. The orogen is undergoing rapid
1056 uplift and exhumation. Crustal thinning through erosion and detachment faulting
1057 (not shown, see text) as the orogen collapses promotes decompressional
1058 melting to produce mafic and felsic melts. The figure is influenced by Oliver et al.
1059 (2008) and Tanner (2014). AF: Alluvial fan, BO: Ballantrae ophiolite, HBO:
1060 Highland Border ophiolite, MVB: Midland Valley basin, MVM: Midland Valley
1061 microcontinent.

1062

1063 **TABLE CAPTIONS**

1064

1065 **TABLE 1:** Re-Os raw data and ages \pm analytical precision/full external uncertainties
1066 (λ). The data define a weighted average age of $438.6 \pm 1.5/1.9$ Ma.

1067

1068 **APPENDICES**

1069

1070 Appendix DM1: Structural information.

1071

1072 Appendix DM2: Raw $^{40}\text{Ar}/^{39}\text{Ar}$ and U-Pb geochronological data.

1073

1074 Appendix DM3: Details of depth estimates for the SHSC.