

1 Revision 2

2 Zircon growth and recrystallization during progressive metamorphism, Barrovian zones,  
3 Scotland

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## ABSTRACT

26 The effect of progressive metamorphism (Grampian orogeny) and later tectonic activity on  
27 detrital zircon in the Barrovian zones of Scotland was studied using secondary ion mass  
28 spectrometry (SIMS) and backscattered electron imaging (BSE). Fifteen samples recording  
29 progressive metamorphism from the chlorite through the sillimanite–K-feldspar zones were  
30 investigated by: (1) SIMS U-Pb depth profiling into rims of unpolished zircon grains to analyze  
31 sub- $\mu\text{m}$ –scale features and (2) conventional spot analysis on sectioned and polished grains. Spot  
32 analyses of zircon interiors yield pre-metamorphic detrital ages for all metamorphic grades. Most  
33 are in the range ca. 600 to ca. 2000 Ma, but some stretch back to the Archean. Younger ages are  
34 recorded in zircon rims, but zircon rim alteration at lower metamorphic grades occurs over much  
35 shorter length scales (the outer  $\sim 80$  nm to  $\sim 1$   $\mu\text{m}$  of the grain) than in the upper amphibolite to  
36 granulite facies (rims of 10 to 30  $\mu\text{m}$ ). For example, Grampian ( $\sim 470$  Ma) metamorphism from  
37 the garnet and kyanite zones affected only the outermost rims ( $< 1$   $\mu\text{m}$ ) of detrital zircon grains.  
38 Thicker, 10 to 30  $\mu\text{m}$  rims that could be dated by conventional spot analysis developed only at  
39 high grades in the sillimanite and sillimanite–K-feldspar zones, probably in the presence of  
40 partial melt. The mean Grampian zircon age from spot and depth profile analyses is  $472 \pm 4$  Ma  
41 ( $n=19$ ). In addition to Grampian ages, the zircon depth profiles reveal ages related to five main  
42 events that postdate the Grampian Orogeny: decompression melting at ca. 450 Ma; subduction  
43 and I-type granite intrusion at ca. 420 Ma; granite intrusion at ca. 384 Ma; extension-related  
44 volcanism and vein mineralization at ca. 335 Ma, and further rifting and basaltic magmatism at  
45 ca. 250 Ma. These events are recorded only in the very narrow rims ( $< 1$   $\mu\text{m}$ ) of zircons, and are

46 thus undetectable with conventional spot analysis. We conclude that: (1) zircon interiors were  
47 able to retain detrital ages up to and including the highest grade of Barrovian metamorphism  
48 (sillimanite-K-feldspar zone) and (2) the <1  $\mu\text{m}$  thick zircon rims may preserve a rich history of  
49 metamorphic and post-metamorphic events that can be dated using SIMS U-Pb depth profiling  
50 techniques.

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52 **Keywords:** Barrovian, zircon, U-Pb geochronology, metamorphism, Dalradian

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## INTRODUCTION

55 The ability to accurately date both peak and post-peak metamorphic and fluid infiltration  
56 events is critical to understanding the geologic history of a metamorphic region. Zircon is an  
57 excellent U-Pb geochronometer due to its ability to substitute U and Th, but generally not Pb,  
58 into its structure when it crystallizes (e.g., Watson et al. 1997). Zircons are ubiquitous and  
59 extremely durable, allowing for their use in dating very old geologic material (e.g., Nemchin et  
60 al. 2006; Trail et al. 2007) and in studying provenance of zircon-containing sediments (e.g.,  
61 Cawood et al. 2003; Fedo et al. 2003; Rahl et al. 2003; Nelson and Gehrels 2007; Gehrels 2012).  
62 Zircons have been used to date metamorphism in myriad ways. Discordant ages from detrital  
63 zircons can be used to date Pb-loss events (e.g., Gastil et al. 1967; Gebauer and Grünenfelder  
64 1976), which was especially useful before the advent of techniques that allow for focused  
65 analysis of single domains within a zircon (Davis et al. 2003). Recently, much work has been  
66 done to understand the effect of metamorphism on zircons and to use zircons to better understand  
67 the evolution of mountain belts (e.g., Vavra et al. 1999; Rubatto et al. 2001; Breeding et al. 2004;

68 Hay and Dempster 2009). Age determination and geochemical analysis of detrital zircons found  
69 in metamorphic rocks have been used to constrain the timing and extent of peak metamorphism  
70 and fluid infiltration even when preserved as small-scale features at the rim or in zircon interiors  
71 (Carson et al. 2002; Mojzsis and Harrison 2002; Breeding et al. 2004).

72 In spite of these advances, much remains to be learned regarding which metamorphic  
73 grades and conditions will result in recrystallized or newly grown zircon, particularly during  
74 progressive, Barrovian-style metamorphism. Our focus is on the classic Grampian metamorphic  
75 rocks in the Scottish Highlands, including the Barrovian type locality of Glen Clova. These rocks  
76 underwent peak metamorphism at 470-465 Ma (Oliver et al. 2000; Baxter et al. 2002). Tectonic  
77 activity in the region, including extensive volcanism and hydrothermal vein formation, continued  
78 for approximately another 150 m.y. Due to the continued elevated heat flow in the region (Oliver  
79 et al. 2008) and the close proximity to the later igneous intrusions (Fig. 1), it is possible that the  
80 rocks in this study were affected by this later tectonic activity. We aim, therefore, to investigate  
81 the effects of both peak and post-peak metamorphic, magmatic, and tectonic activity on the  
82 zircons from the metamorphic rocks of the Scottish Highlands.

83 Conventional U-Th-Pb isotopic analysis of zircon by Secondary Ion Mass Spectrometry  
84 (SIMS) involves focusing the ion beam on interior domains of a sectioned and polished grain  
85 mounted in epoxy. Coupled with backscattered electron (BSE) or cathodoluminescence (CL)  
86 images of the sectioned zircons, this “spot” method allows for the analysis of specific domains  
87 within a zoned zircon as long as the domains are larger than the size of the beam. “Depth  
88 profiling” by SIMS, on the other hand, is a method for analysis of much smaller (tens to  
89 hundreds of nm-scale) domains at the unsectioned, unpolished rims of zircons and is uniquely  
90 suited to detect changes in the isotopic composition of grains with depth (Carson et al. 2002;

91 Mojzsis and Harrison 2002). This method has been used in one area in the Scottish Highlands, as  
92 discussed below, by Breeding et al. (2004).

93 We present SIMS data from both traditionally sectioned and polished zircons and from  
94 depth profiling into unpolished zircon rims. The 15 samples reported in this study are from the  
95 chlorite through the sillimanite–K-feldspar zones in the Grampian Highlands of Scotland, with  
96 locations ranging from the west to the east coasts (Fig. 1). The goals of the study are to: 1) assess  
97 the degree of zircon growth/recrystallization that took place during progressive metamorphism  
98 across all metamorphic zones; 2) correlate post-peak ages recorded in the zircons to the ages of  
99 the abundant igneous rocks in the region (Fig. 1); and 3) relate internal textural information from  
100 zircon grains to isotopic and age data from SIMS analysis.

## 101 **GEOLOGIC SETTING**

102 The samples are pelitic metasediments from the Dalradian Supergroup, which lies  
103 between the Highland Boundary Fault (HBF) and the Great Glen Fault (GGF) in Scotland (Fig.  
104 1). Following Vorhies and Ague (2011), the field area was subdivided into Regions I-III, from  
105 northeast to southwest. The original sediments were deposited along the edge of the Iapetus  
106 Ocean during an extended period of rifting and subsequent basin closure (Cawood et al. 2003;  
107 MacDonald and Fettes 2006). Based upon detrital zircon ages combined with paleocurrent data,  
108 the sediment source was mostly Laurentian (Grenvillian) with smaller Lewisian input and with  
109 possible input from Baltica (Cawood et al. 2003; Breeding et al. 2004; Banks et al. 2007).  
110 Deposition began around 800 Ma and continued until the basin closure around 530 Ma (Cawood  
111 et al. 2003).

## 112 **Grampian event**

113           The tectonic events associated with the closure of the Iapetus Ocean are responsible for  
114 the metamorphism of the rocks in this study. The orogeny resulted in an increase in metamorphic  
115 grade from the HBF northward (Fig. 1), providing the basis for Barrow's (1893; 1912) original  
116 study of metamorphic index minerals and the later work of Tilley (1925). Around 490 Ma the  
117 initial loading of the Dalradian sediments began with the obduction of the Highland Border  
118 Ophiolite (Chew et al. 2010). The Grampian Orogeny continued with the collision of Laurentia  
119 with the Midland Valley Arc, and possibly other outboard microcontinents (Oliver et al. 2008;  
120 Chew et al. 2010). Following the collisional event, there was slab break-off and a resulting slab  
121 window (Oliver et al. 2008) and/or lithospheric extension (Viète et al. 2010). The subsequent  
122 rising hot asthenosphere partially melted and rose up to form the Newer Gabbros in the northeast  
123 (Oliver et al. 2008). The associated increased heat flow also partially melted the lower crust to  
124 form the numerous syn-metamorphic granites in the northeast (Regions I and II).

125           The combination of collisional tectonics with igneous intrusions during the Grampian  
126 Orogeny shaped the pressure–temperature–time paths of the metamorphic rocks. Most of what  
127 follows is from Vorhies and Ague 2011 and references therein. After the loading and increase in  
128 pressure ( $P$ ) across the entire Grampian terrane, the metamorphic histories of different localities  
129 in the study area began to diverge. In the western half of the Highlands, what we refer to herein  
130 as Region III, peak  $P$  in the garnet zone was relatively high, 0.9-1.1 GPa, at temperatures ( $T$ )  
131 between 500 and 630 °C. Peak metamorphism in this region was followed by near-isothermal  
132 decompression. Around the Barrovian type locality of Glen Clova, in Region II, maximum  
133 pressures were also high (~0.8-0.9 GPa). Peak temperatures, however, were attained at lower  
134 pressures during exhumation. At ~0.6 GPa, temperatures increased rapidly as the result of a brief

135 thermal pulse or pulses lasting a total of a few hundred thousand years to a few million years  
136 (Ague and Baxter 2007; Vorhies and Ague 2011, Viete et al. 2011). Also in Region II, to the  
137 north of Glen Clova in Glen Muick, the rocks reached upper amphibolite–granulite facies  
138 conditions of 750-800 °C at ~0.9-1.0 GPa. Pressures at peak-*T* in Region I to the northeast are  
139 the lowest observed, at ~0.4-0.5 GPa. These rocks as well as the upper amphibolite–granulite  
140 facies rocks of Region II were probably affected by the same tectonometamorphic activity that  
141 caused the thermal pulses in Glen Clova.

142 Current geochronological constraints on the metamorphism consist of garnet–whole-rock  
143 Sm-Nd dating, ages of nearby syn-metamorphic igneous intrusions (Fig. 1, Table 1), and U-Pb  
144 zircon dating of metamorphic rocks. The Grampian Orogeny is thought to have been relatively  
145 rapid, lasting around 15 m.y. (Oliver et al. 2000; Dewey 2005), with garnet growth lasting ca. 8  
146 m.y., from ca. 473-465 Ma (Baxter et al. 2002). Garnet growth in the garnet, kyanite, and  
147 sillimanite zones occurred penecontemporaneously at 467-464 Ma (Baxter et al. 2002). In order  
148 to further constrain the age of metamorphism Breeding et al. (2004) carried out SIMS depth-  
149 profiling on zircons from a rock in our Region I. Ague (1997) previously determined that fluid  
150 flow through fractures had altered the chemistry and mineral assemblage of a selvage region  
151 adjacent to a quartz vein. Zircons from within the vein selvage had a U-Pb lower intercept age of  
152  $462 \pm 9$  Ma with isotopic alteration apparent in the outer 1.3  $\mu\text{m}$  of the zircon grain. The outer ~1  
153  $\mu\text{m}$  of the zircon is marked by an increase in U, Th, and non-radiogenic Pb compared to the more  
154 interior portions of the grain. Breeding et al. (2004) concluded that this age reflects zircon  
155 growth or recrystallization during the syn-metamorphic fluid infiltration.

156 The synmetamorphic gabbros are part of a group called the Newer Gabbros which are  
157 large, mantle-derived mafic intrusions (Pankhurst 1969; Dempster et al. 2002). The largest of

158 these are the Inch and Morven-Cabrach gabbros, but the group also includes the Haddo House  
159 and Portsoy Gabbros. The mafic magmatism was quite extensive at this time, with the thickness  
160 of the Inch estimated to be as much as 5,500 m (Clarke and Wadsworth 1970). Many studies  
161 have attempted to date different intrusions in the group and most put the age of intrusion at ca.  
162 470 Ma (e.g., Brown et al. 1965; Pankhurst 1970; Dempster et al. 2002). The synmetamorphic  
163 granites, such as the Auchlee and Aberdeen, include S-type granites that almost certainly contain  
164 a substantial component of partially-melted sedimentary crust (Harmon et al. 1984). Oliver et al.  
165 (2008) conclude that the melting was due to increased crustal heat flow caused by a slab window  
166 and the associated rising asthenosphere in Regions I and II. The added heat required for the  
167 thermal pulses during Grampian metamorphism was likely supplied by these synmetamorphic  
168 intrusions (Baxter et al. 2002; Ague and Baxter 2007; Vorhies and Ague 2011). Viète et al.  
169 (2011) further postulate that shear heating could also have played a role.

#### 170 **Post-Grampian tectonic events**

171 Following the Grampian event, exhumation continued with little igneous activity until ca.  
172 430 Ma (Fig. 1, Table 1). There is one intrusion dated at 457 Ma (in addition to others in other  
173 Scottish terranes) which has been attributed to decompression melting during exhumation of the  
174 Grampian crust (Oliver et al. 2008). Around 430 Ma the final Caledonian collision of Avalonia  
175 against the Highland terrane began and was followed first by subduction-related granitic rocks  
176 (e.g., Dewey 1971), then by unilateral (Atherton and Ghani 2002) or bilateral (Oliver et al. 2008)  
177 slab break-off and another period of asthenospheric upwelling. These events resulted in granitic  
178 magmatism between the ages of 430 and 400 Ma. Following these events there is one intrusion at



179 396 Ma (Skene) and another at 390 Ma (Glen Tilt) which are possibly related to the distant  
180 Acadian Orogeny (Oliver et al. 2008).

181 Following the Caledonian orogeny there are a few igneous rocks and vein complexes that  
182 may also have affected the rocks in this study. First there was extension-related volcanism to the  
183 south of Region III between about 335 and 343 Ma (Monaghan and Parrish 2006). Also in  
184 Region III are carbonate veins thought to have originally formed 10-30 m.y. after peak Grampian  
185 metamorphism, then reactivated and remineralized in the late Carboniferous or early Permian  
186 (Parnell et al. 2000; Anderson et al. 2004).

## 187 **METHODS**

### 188 **Sample preparation**

189 Samples were crushed and separated using standard density and magnetic separation  
190 techniques. An aliquot of zircon grains and the AS3 standard (Schmitz et al. 2003) were hand-  
191 picked and placed onto double-sided tape, then cast in epoxy. The epoxy mounts were sectioned  
192 to reveal grain interiors, polished using  $\frac{1}{4}$   $\mu\text{m}$  diamond paste, cleaned ultrasonically, rinsed with  
193 1 *N* HCl, and coated with  $\sim 10$  nm of Au using a sputter coater. Imaging of sectioned grains was  
194 done using backscattered electron imaging (BSE) on the JEOL JXA-8530F electron microprobe  
195 at Yale University. Additional zircons were hand-picked and pressed into indium (In) metal and  
196 left unpolished. Standard AS3 crystals were pressed into the mount and polished prior to the  
197 addition of the unknown zircons. The In mount was cleaned ultrasonically, rinsed with 1 *N* HCl,  
198 and coated with  $\sim 10$  nm of Au.

199 **SIMS analysis**

200 SIMS analysis of zircons was performed at the University of California, Los Angeles on  
201 the CAMECA ims 1270 using previously published techniques (Schmitt et al. 2003; Breeding et  
202 al. 2004). An  $^{16}\text{O}^-$  primary beam at 10 keV was focused to a  $\sim 15\ \mu\text{m}$  spot diameter. Oxygen  
203 flooding was used to increase  $\text{Pb}^+$  yields. Secondary ions of  $^{94}\text{Zr}_2^{16}\text{O}$  (counting time = 1 s),  $^{204}\text{Pb}$   
204 (3 s),  $^{206}\text{Pb}$  (6 s),  $^{207}\text{Pb}$  (7 s),  $^{208}\text{Pb}$  (4 s or 2 s, depending on session),  $^{232}\text{Th}$  (2 s),  $^{238}\text{U}$  (3 s),  
205  $^{238}\text{U}^{16}\text{O}$  (2 s) were measured in peak jumping mode with individual sweeps over this mass range  
206 constituting an analysis cycle. Three techniques were used in this study. First is the conventional  
207 analysis of sectioned zircons cast in epoxy. Hereafter called “spot” analyses, these took  $\sim 12$   
208 minutes each and resulted in a pit  $\sim 0.75\ \mu\text{m}$  deep. Two types of depth profiling were also used,  
209 wherein the analyses were done into the unpolished rims of whole zircon grains mounted in  
210 indium (In). “Short depth profiles” (SDPs) were done under the same conditions as the epoxy  
211 spots, resulting in  $\sim 15\ \mu\text{m}$  wide and  $\sim 0.75\ \mu\text{m}$  deep pits. Pit size and shape was measured using a  
212 MicroXAM-100 3D surface profiler and optical interferometer at UCLA. This information was  
213 used to estimate sputter rates. Analyses are separated into “blocks” which interpolate intensities  
214 between two consecutive magnet cycles. Each block represents  $\sim 75\ \text{nm}$  of depth. Since the SDPs  
215 were done without narrowing the field aperture with depth there is a degree of isotopic mixing  
216 that occurs during analysis. The relative contribution of surface-derived ions can be estimated by  
217 measuring the Au content at each cycle, as the samples are coated with Au. A depth profile done  
218 on standard grain 91500 (Wiedenbeck et al. 2004) shows that the surface Au signal decays  
219 approximately exponentially. After 7 cycles the Au signal is  $\sim 50\%$  of the original signal and  
220 after 9 cycles (the depth of the SDPs done in this study) the Au signal is  $\sim 40\%$  of the original.  
221 We can use this to algebraically estimate the actual age of the zircons at cycle 9 given the rim

222 age and the apparent age at cycle 9. For example, the  $^{206}\text{Pb}/^{238}\text{U}$  age of cycle 1 from analysis  
223 41A\_2\_2 (Fig. 7) is 477 Ma and the apparent  $^{206}\text{Pb}/^{238}\text{U}$  age of cycle 9 is 940 Ma. The actual age  
224 at cycle 9 will be closer to 1250 Ma, given a 40% contribution from cycle 1 and a 60%  
225 contribution from cycle 9. The larger the difference between cycle 1 and cycle 9, the larger the  
226 difference there will be between the cycle 9 actual and apparent ages. This is a simple model  
227 which assumes equal U concentrations and ignores the additional contributions from cycles 2-8.

228         Calculating the relative contribution of the surface layer for the interior part of short  
229 depth profiles (SDP) using Au does not depend on any assumption of similar ionization or  
230 collection efficiencies, only that these remain constant throughout the analysis. This is, to a large  
231 extent, justified in that data is only collected after an initial pre-sputter time during which sputter  
232 equilibrium is achieved. Also, the SDP duration is sufficiently short so that ionization effects  
233 remain largely constant throughout the analysis. In any case, we use this calculation as an  
234 illustration of the surface contribution effect, and do not aim to quantify interior ages in this way  
235 because of the complexities regarding U and age zonation in individual crystals. To be sure that  
236 the ages from SDP analyses represent the true rim age without any older, inner age domains  
237 mixing in, cycles from SDPs were graphed and carefully selected so that only the youngest  
238 cycles were used to calculate the age.

239         Finally, a “long depth profile” (LDP) was done, lasting ~90 minutes, with the field  
240 aperture set smaller than the secondary beam diameter to exclude secondary ions from the edges  
241 of the analysis pit and allow for better depth resolution in a deeper pit. Instrumental bias between  
242 conventionally sectioned zircon crystals in epoxy mounts, and unsectioned crystals pressed into  
243 In is absent based on excellent agreement between rim and interior analyses for rapidly  
244 crystallized volcanic zircon (e.g., Bindeman et al. 2006).

245 Concentrations of U and Th were estimated by comparing known values of  $U/^{94}Zr_2^{16}O$   
246 and  $Th/^{94}Zr_2^{16}O$  in the AS3 (Schmitz et al. 2003) and 91500 (Wiedenbeck et al. 2004) standard  
247 zircons to the same ratios in the unknowns. Corrections for common Pb were made using  $^{204}Pb$   
248 as a proxy for common Pb content. We used anthropogenic Pb ratios ( $^{206}Pb/^{204}Pb = 16.2$  and  
249  $^{207}Pb/^{204}Pb = 15.3$ ) from Sañudo-Wilhelmy and Flegal (1994). Overcorrection of  $^{207}Pb^*$  can  
250 occur by using  $^{204}Pb$  as a proxy for common Pb with unresolved minor interferences (e.g.,  
251  $^{186}W^{18}O$ ), resulting in apparent reverse discordance. We have tested this by using  $^{208}Pb$  as an  
252 alternative proxy for common Pb which mitigates reverse discordance, but concordia ages  
253 remain the same as with the  $^{204}Pb$  correction. However, because the  $^{208}Pb$  correction has higher  
254 uncertainties for high  $^{208}Pb^*$  analyses, and for consistency, we report the  $^{204}Pb$  corrected ages  
255 only. Data reduction and calculation of U-Pb ages were done using UCLA software (ZIPS v3.0.4  
256 written by C.D. Coath 2005) and Isoplot v4.11 (Ludwig 2008).

## 257 **Sample descriptions**

258 The zircons exhibit internal features imaged using BSE that are associated either with a  
259 detrital origin or with later growth/recrystallization. The detrital portions of the zircons—either  
260 the cores or the entire grain—may contain oscillatory zoning which indicates an igneous origin  
261 (e.g., Fig. 2J, L) or be completely homogeneous (e.g., Fig. 2A, B). They are often fractured (e.g.,  
262 Fig. 2B, C, E, F). They also yield ages that are older than the age of deposition (Cawood et al.  
263 2003). Three main features are possible indicators of metamorphic growth or recrystallization  
264 (Hoskin and Black 2000; Putnis 2002; Corfu et al. 2003; Harley et al. 2007; Hay and Dempster  
265 2009). (1) unzoned, nonporous rims ranging from sub-micrometer scale to tens of micrometers in  
266 width (e.g., Fig. 2I, K). (2) Rims containing micro-pores which terminate in irregular grain edges

267 (e.g., Fig. 2A). (3) Highly porous altered zones throughout the grain which may or may not  
268 follow pre-existing zoning (e.g., Fig. 2C, E, G, H, J). The nonporous rims may be either brighter  
269 or darker in BSE than the cores, however the porous portions are always brighter than the rest of  
270 the grain. The brighter BSE here is due to the higher levels of U in the domains, whereas the  
271 darker domains have lower U.

272 **Chlorite zone.** Three samples are from the chlorite zone in Region III; all are dominated  
273 by the mineral assemblage Qtz+albitic Pl+Ms+Chl (abbreviations from Kretz 1983). Sample  
274 120C is a metapsammite from an outcrop of metapsammitic and metapelitic rocks. The  
275 metapsammitic layers contain networks of quartz veins. Many of the zircon grains have <5  $\mu\text{m}$   
276 porous rim overgrowths, although the interiors of most grains are homogeneous (Fig. 2A).  
277 Samples 141A and 141B are from an outcrop containing a 1-2m wide quartz-rich vein which  
278 metasomatized the surrounding chlorite-rich phyllite. Sample 141A is from the altered portion of  
279 the outcrop and consists of large amounts of albitic plagioclase feldspar and altered inclusions of  
280 wall rock. The zircons have rim overgrowths as in sample 120C but with more cracks in their  
281 interiors (Fig. 2B). Sample 141B is a chlorite-rich phyllite and is inferred to have been the  
282 precursor to the metasomatized 141A. The zircons appear similar to those from 141A.

283 **Biotite zone.** Sample 4F is from the biotite zone in Region I. It is a coarse biotite schist  
284 containing mostly Qtz+Pl+Bt+Ms+Chl. Biotite porphyroblasts are largest (~2 mm) adjacent to  
285 large (5-20 cm) quartz veins which cross-cut pre-existing folds. The zircons are often cracked  
286 and have homogeneous to lightly-zoned interiors. They have rare porous interior and rim  
287 domains (Fig. 2C).

288 **Garnet zone.** In Region III, sample 150B1 is from a massive quartz and feldspar vein  
289 surrounded by selvage metapelite. The major silicate mineral assemblage is Qtz+Grt+albitic

290 Pl+Ms+Chl. A few of the zircons have distinct rim domains (Fig. 2D) and approximately half  
291 have interior porous areas (Fig. 2E). Sample 235A from Region II is a garnetiferous  
292 metapsammite with the prograde assemblage Qtz+Grt+Pl+Bt+Ms. The surrounding area contains  
293 quartz veins ~15-20 cm thick with a few locally smaller quartz veins. There are some rim  
294 domains and little interior porosity in the zircons (Fig. 2F).

295 **Staurolite zone.** The staurolite zone sample, 154J, is from an outcrop in Region I  
296 containing extensive veining with the abundance and size of the garnet and the staurolite  
297 increasing with proximity to veins (Masters and Ague 2005). The sample itself is from one of  
298 these near-vein reaction zones and contains Qtz+Grt+Pl+Bt+Ms+St±Chl. The zircons have  
299 occasional rim domains and interior porosity similar to those illustrated in Figures 2E and 2F

300 **Kyanite zone.** Sample 242B is from the Barrovian type locality in Region II. It is a  
301 garnetiferous schist layered within a primarily metapsammite outcrop. There is no nearby  
302 veining. The mineral assemblage is Qtz+Grt+Pl+Bt+Ms. More than half of the zircon grains in  
303 the separate have abundant interior porous domains (Fig. 2G).

304 **Sillimanite zone.** All three sillimanite zone samples are from the Barrovian type locality  
305 in Region II. Sample 66A is from a migmatitic, metapelitic gneiss containing  
306 Qtz+Grt+Pl+Bt+Ms+Sil+Ky. More than half of the zircons contain interior porous domains and  
307 one third have rim domains (Fig. 2H). Sample 263A is a veined, pelitic gneiss with large (0.5  
308 cm) garnets and the prograde mineral assemblage Qtz+Grt+Pl+Bt+Ms+Sil. Approximately half  
309 of the zircons from 263A have interior porous domains, sometimes along pre-existing zonation.  
310 Many zircons have distinct, nonporous rims (Fig. 2I). Sample 41A is from a massive, highly  
311 migmatitic psammitic gneiss cut by a 1 cm vein containing quartz, plagioclase, and muscovite.  
312 The mineral assemblage is Qtz+Grt+Pl+Bt+Ms+Sil+Ky . Almost all of the zircons have porous

313 domains; some of these follow possible original zoning and others cross the grain at seemingly  
314 random locations.

315 **Sillimanite–K-feldspar zone.** These samples were collected in Glen Muick (Baker and  
316 Droop 1983; Baker 1985) and in later sections may be referred to as a group rather than as  
317 individual samples. All of the samples contain Qtz+Grt+Kfs+Pl+Bt+Sil. The rocks from this area  
318 are the highest-grade in the sequence and the outcrops are migmatitic. Mafic granulites  
319 containing clinopyroxene+hornblende+garnet crop out nearby (Baker and Droop 1983; Baker  
320 1985). In our samples cordierite did not form because of the relatively high pressures (~0.9-1  
321 GPa). Sample 284A is a massive, coarse, unveined gneiss with bimodal garnet sizes (<1 mm and  
322 ~3 mm diameters). Samples 285A and 286B are gneissose and contain cm-scale quartz veins  
323 surrounded by coarse sillimanite. Sample 288B is an unveined, sillimanite-rich gneiss containing  
324 layers of quartz and ~4 mm garnets. The zircons from sample 284A all contain either oscillatory  
325 or irregular zoning. Approximately one third have porous domains along zones (Fig. 2J). Some  
326 also have distinct nonporous rims as in 263A (Fig. 2K). Sample 285A zircons have less porosity  
327 and none of the wide rims present in 284A. The zircons from 286B and 288B contain abundant  
328 cracks at the rims and through the centers of the grains. Most have oscillatory zonation and there  
329 is much less porosity than in the zircons from 284A (Fig. 2L).

### 330 **ZIRCON BEHAVIOR DURING PROGRESSIVE BARROVIAN METAMORPHISM**

331 Results of all spot analyses with radiogenic Pb content >90% (n=98) are shown on a  
332 concordia diagram in Fig. 3 and listed in Supplementary Data Table 1. Concordia ages from spot  
333 (n=63), SDP (n=53), and LDP (n=1) analyses are summarized in Fig. 4. All age results are  
334 quoted at 2 $\sigma$  uncertainty.

335 **Spot analyses**

336 The age results from spot analyses from all zones range from Grampian to ca. 2800 Ma  
337 (Fig. 4A). Importantly, in the chlorite through kyanite zones there are no ages from spot analysis  
338 younger than 500 Ma. In the sillimanite and sillimanite–K-feldspar zones, however, there are  
339 ages that reflect zircon growth or recrystallization during the Grampian Orogeny. The six  
340 measured Grampian rim domains result in a concordia age of  $468 \pm 7$  Ma (mean square of  
341 weighted deviates of concordance and equivalence, MSWD=0.9). This is identical, within error,  
342 to the age of fluid infiltration in the garnet zone of Region I found by Breeding et al. (2004) and  
343 to the age range of garnet growth found by Baxter et al. (2002). Additionally, despite the  
344 difference in metamorphic grade and the distance between the upper amphibolite–granulite  
345 facies rocks of Glen Muick and the rest of Region II, by separating the ages of the two areas  
346 there is no indication that they were metamorphosed at different times. The two spot analyses  
347 from the sillimanite zone give a combined age of  $476 \pm 28$  Ma (MSWD=0.28) and the four spot  
348 analyses from Glen Muick give a combined age of  $467 \pm 7$  Ma (MSWD=1.2).

349 We can also correlate the appearance of the spot analysis location to the isotopic results.  
350 Two main features are visible in BSE: (1) porous bands that are lighter in BSE intensity than the  
351 rest of the grain that are found in all zones (e.g., Fig. 2G, J), and (2) nonporous, lobate,  
352 homogeneous domains near the rims of grains that cut across pre-existing zonation that are found  
353 in the sillimanite and sillimanite–K-feldspar zones (Fig. 2I, K). The geochronological results of  
354 analysis of these two types of domains differ. Spots on porous bands, whether they are on the  
355 rim, following pre-existing zones, or cross-cutting the grain, result in discordant or nearly  
356 discordant ages. Ages that are nearly completely discordant have very high uncertainty. For  
357 example, the analysis spot shown in Fig. 2J has an imprecise concordia U-Pb age of  $596 \pm 220$



358 Ma due to the high discordance. All discordant analyses have  $^{206}\text{Pb}/^{238}\text{U}$  ages that are older than  
359 600 Ma and most are older than 1000 Ma, indicating that age resetting in these porous zones has  
360 not occurred during Grampian metamorphism and fluid infiltration. These results are not to be  
361 taken as actual ages, but are reported to show the high degree of isotopic discordance in these  
362 porous bands. In contrast, all concordant Grampian ages detected using spot analyses were  
363 performed on nonporous rim domains up to 30  $\mu\text{m}$  in width from the sillimanite and  
364 sillimanite–K-feldspar zones (Fig. 2I, K). The only exception to this is one zircon from Glen  
365 Muick which appears to be completely metamorphic (JAB288Brow1\_14). It is likely that the  
366 same type of alteration is responsible for the two different textures seen in BSE imaging: fluid-  
367 mediated dissolution-precipitation reactions (e.g., Putnis 2009). Other such zircons are seen in  
368 environments where fluid infiltration was important (e.g., Wayne and Sinha 1992; Hacker et al.  
369 1998; Hoskin and Schaltegger 2003; Hay et al. 2009). In this process the original zircon  
370 composition is metastable and soluble in the local fluid. Porosity develops, allowing the reaction  
371 interface to move further into the grain and the new zircon to reprecipitate. This process has been  
372 demonstrated in zircons and other minerals under experimental conditions (e.g., Harlov and  
373 Dunkley 2010). The rim areas on the zircons that gave concordant Grampian ages were likely  
374 affected by the same process. The rim porosity is either on the nanoscale and thus too small to  
375 image, or was destroyed by later recrystallization.

### 376 **Short depth profiles**

377 Short Depth Profile (SDP) ages obtained from the outermost micrometer of zircon rims  
378 span from ca. 250 Ma to ca. 2500 Ma (Fig. 4B, Supplementary Data Table 2). As a group, these  
379 results differ greatly from the spot analyses in that most of the SDP ages are younger than 800

380 Ma and more than half are of Grampian age or younger. The mean Grampian age derived from  
381 12 SDP analyses from Regions II and III is  $474 \pm 6$  Ma (MSWD=0.94), which is within error of  
382 the previously determined Grampian ages as well as the  $468 \pm 7$  Ma age derived from spot  
383 analyses in this study. These SDP results can be separated in order to compare the ages by  
384 geographical region. For Region I Breeding et al. (2004) determined that the age of fluid  
385 infiltration in the garnet zone was  $462 \pm 9$  Ma. In Region II there are eight analyses with ages  
386 corresponding to the peak Grampian event from kyanite zone sample 242B, sillimanite zone  
387 samples 41A and 263A, and from sillimanite–K-feldspar zone samples 284A and 286B. These  
388 are equivalent at  $473 \pm 7$  Ma (MSWD=0.61). In Region III there are four SDP analyses, all from  
389 garnet zone sample 150B1, that are equivalent at  $483 \pm 25$  Ma (MSWD=1.7). While it must be  
390 noted that the uncertainty of the combined age in Region III is admittedly high, these results are  
391 consistent with the hypothesis that the Grampian Orogeny was penecontemporaneous, within  
392 error, in all three regions across the Highlands. Furthermore, as with the spot analyses, we can  
393 separate out the upper amphibolite–granulite facies Glen Muick samples from the rest of the  
394 samples from Region II to show that these ages too are equivalent within error. The SDPs from  
395 the Glen Muick rocks have a combined age of  $477 \pm 8$  (n=3, MSWD=0.58) and the SDPs from  
396 the other samples from Region II have a combined age of  $467 \pm 11$  (n=5, MSWD=0.49).

397       There are 23 SDP results that indicate growth/recrystallization after the Grampian  
398 Orogeny. As summarized in Figure 5, these results fall into five distinct ages or groups of ages,  
399 four of which can be correlated to known local igneous or tectonic events. First there are six  
400 analyses that give a combined age of  $450 \pm 9$  Ma (MSWD=1.3). These are from samples 120C  
401 (chlorite zone, Region III), 150B1 (garnet zone, Region III), 242B (kyanite zone, Region II), and  
402 263A (sillimanite zone, Region II). This 450 Ma age is during a time recognized by Oliver et al.

403 (2008) to have been marked by decompression and erosion and numerous S-type granitic  
404 intrusions, likely the result of decompression melting. While there is only one known igneous  
405 intrusion of approximately this age illustrated in the current work (Kennethmont Granite, Fig. 1,  
406 Table 1), there are other intrusions of this age to the north of the study area that are not listed in  
407 this paper (Oliver et al. 2008). Next there are seven SDP analyses from samples 41A and 263A  
408 (sillimanite zone, Region II) that give a combined age of  $421 \pm 11$  Ma (MSWD=0.8). These two  
409 samples are close to each other and also very close to the Lochnagar and Glen Doll granites,  
410 which intruded at  $420 \pm 2$  Ma and  $419 \pm 5$  Ma, respectively (Table 1). The heat and/or fluid input  
411 from these proximal intrusions likely caused the growth/recrystallization seen in the outermost  
412 zircon rims from these samples. Five analyses from sillimanite zone samples 41A and 263A  
413 (Region II) and garnet zone sample 150B1 (Region I) give a combined age of  $384 \pm 13$  Ma  
414 (MSWD=1.4). This age is within error of the age of the Glen Tilt and Skene granites (Table 1).  
415 The zircons in this study were likely affected by the same tectonic activity that produced these  
416 granites—possibly the Acadian Orogeny (Oliver et al. 2008).

417       Following the grouping at ca. 385 Ma there are two more ages seen in the SDP rim  
418 analyses. Chlorite zone sample 120C from Region III and sillimanite zone sample 41A from  
419 Region II have one analysis each which combine to an age of  $339 \pm 16$  (MSWD=0.43). During  
420 this time there was extension-related volcanism in the Midland Valley Terrane to the south of the  
421 HBF. In particular, just to the south of sample 120C is the Renfrewshire Hills Block of the Clyde  
422 Plateau Lavas which was formed at 335 Ma (Monaghan and Parrish 2006). The volcanic rocks  
423 from the extension events to the east may have affected sample 41A as well. Finally there is an  
424 SDP age of  $252 \pm 17$  Ma from sample 263A (sillimanite zone, Region II). Following the  
425 Carboniferous extension, rifting continued and further volcanism occurred in the Midland Valley

426 Terrane. There was basaltic magmatism to the north and to the south of the study area dated as  
427 late as ca. 250 Ma and ca. 264 Ma, respectively (Upton et al. 2004). We conclude that the ca. 250  
428 Ma age in sample 263A is likely related to this continued activity in the region.

429 Interestingly, the SDP results reveal that only two samples below the sillimanite zone  
430 record Grampian ages on the outer ~1  $\mu\text{m}$  rims of zircons. Sample 242B is from the kyanite zone  
431 and has one Grampian age SDP and sample 150B1 is from the garnet zone and has four  
432 Grampian age SDPs. Sample 150B1 is heavily veined and, thus, metamorphic vein-related fluids  
433 may have helped mediate Grampian zircon growth and/or recrystallization (e.g., Breeding et al.,  
434 2004). Post-Grampian rim domains apparently developed more pervasively in the rocks. It is  
435 possible that earlier, Grampian-age domains were destroyed by later growth and/or  
436 recrystallization, or that such domains never formed in the first place.

437 Retrograde metamorphism is common throughout the Highlands; small degrees of  
438 retrogression are present in most samples studied herein (e.g., chlorite after garnet, biotite, or  
439 staurolite; fine-grained muscovite aggregates after aluminosilicates). The SDP results show that  
440 post-peak Grampian activity spanned 100 Ma or more, and was associated with pulses of  
441 magmatism and/or tectonism. Thus, retrogression had multiple causes, and multiple events are  
442 recorded in zircons from one sample, such as in 41A and 263A as described above. Resolution of  
443 these tiny age domains is impossible with conventional SIMS techniques, and instead requires  
444 depth profiling.

445 U-Pb internal systematics do not allow us to distinguish between Pb-loss and new  
446 growth/recrystallization for this age range. We therefore use compositional evidence and the  
447 consistent behavior of samples (i.e. absence of distinct rim regions in some samples, presence in  
448 others) as indications to dismiss Pb-loss. Most of the peak and post-peak ages reported have

449 Th/U < 0.1, used as qualitative evidence of metamorphic growth or recrystallization, as opposed  
450 to the older, detrital ages reported which usually have higher Th/U. As described previously, the  
451 spot analyses of Grampian age are located on homogeneous, rim regions, which we conclude are  
452 areas of metamorphic growth/recrystallization. Figure 3B shows the spot analyses on the  
453 concordia diagram from 0-600 Ma. The ellipses cluster around ca. 470 Ma and do not extend into  
454 younger ages, indicating that recent Pb-loss is not responsible for the ages. Additionally, the  
455 post-peak ages that we have reported are in distinct clusters and are the same age as known  
456 tectonic events, whereas Pb-loss would produce more of a continuous range of ages approaching  
457 the present.

458         In this context, it is interesting to note that zircon crystals from the sillimanite–K-feldspar  
459 zone rocks of Glen Muick lack post-peak rim ages; however this data set only includes six SDP  
460 analyses. A recent study of zircons from the Valle D’Arbedo in the Swiss Alps found that rims  
461 which had been recrystallized during an earlier episode of metamorphism were not affected by  
462 the later Alpine event because recrystallized zircon was more stable (Vonlanthen et al. 2012).  
463 Therefore it is possible that the Grampian recrystallization protected the upper amphibolite–  
464 granulite facies zircons of Glen Muick from the later events. However, there are many post-peak  
465 ages in zircons from the sillimanite zone, which also have wide Grampian rim domains. Zircon  
466 stabilization during high-temperature metamorphic recrystallization therefore appears possible,  
467 but additional investigation is required to further corroborate this.

#### 468 **Long depth profile**

469         A long depth profile (~5.5  $\mu\text{m}$ ) from sample 286B provides evidence constraining the  
470 timing of the upper amphibolite–granulite facies metamorphism and partial melting of the rocks

471 in Glen Muick. Fig. 6 shows the  $^{206}\text{Pb}/^{238}\text{U}$  and  $^{207}\text{Pb}/^{235}\text{U}$  ages as well as the Th/U content of the  
472 zircon plotted against depth (see also Supplementary Data Table 3). The isotope data from the  
473 outer  $\sim 1.15\ \mu\text{m}$  of the zircon gives a concordia age of  $475 \pm 10\ \text{Ma}$ . In the inner  $\sim 2.5\ \mu\text{m}$  of the  
474 depth profile the Th/U is  $\sim 0.01$  then begins to rise, peaking at  $\sim 0.1$  at  $\sim 2\ \mu\text{m}$  depth. Th/U then  
475 drops again and in the outer  $\sim 1.15\ \mu\text{m}$  is  $\sim 0.017$ . Elevated Th/U in zircon (above 0.1) is often  
476 taken as a sign of melt-present growth because Th is mobilized in melt, with lower values  
477 indicating metamorphic growth (Harley et al. 2007). The ages from  $\sim 5.1\ \mu\text{m}$  to  $\sim 2\ \mu\text{m}$  (the  
478 deepest part of the profile) reveal an old inherited core, likely of metamorphic origin. The spike  
479 in Th/U corresponds to the sharp drop in zircon age from the older interior to the outer rim age of  
480  $475 \pm 10\ \text{Ma}$ . The spike almost certainly represents mobilization of Th during partial melting  
481 during the Grampian event. The reduced Th/U ratio after the spike indicates either that a high-Th  
482 accessory mineral (e.g., monazite) incorporated much of the available Th during the initial  
483 melting stage and the zircon grew in the presence of that melt, or that the rim growth occurred in  
484 the absence of melt.

485         The long depth profile shows that the Glen Muick rocks underwent metamorphism and  
486 partial melting during the Grampian orogeny, a conclusion that is also supported by the spot and  
487 SDP data: By combining spot and SDP results from Glen Muick we get an age of  $471 \pm 6\ \text{Ma}$   
488 ( $n=7$ ,  $\text{MSWD}=1.13$ ). Comparably, the seven spot and SDP results of the other rocks from  
489 Region II, not including the Glen Muick samples, combine to give an age of  $468 \pm 10\ \text{Ma}$   
490 ( $\text{MSWD}=0.43$ ). Finally, combining all Grampian-age spot SDP, and LDP data from all samples  
491 results in an age of  $472 \pm 4\ \text{Ma}$  ( $n=19$ ,  $\text{MSWD}=0.5$ ).

## 492 **Spatial variation in SDP ages**

493           Depth profiles in the Barrovian zones reveal that zircon alteration during a given event  
494 certainly does not affect every grain in the sample and, moreover, it can affect different parts of  
495 the same grain differently. For example, three SDPs were performed on one grain from sample  
496 41A (Fig. 7). The outermost cycle of analysis 2\_1 is concordant at  $642 \pm 110$  Ma and cycles 2  
497 and 3 are discordant, but appear to be approximately the same age. Cycles 1 and 2 of analysis  
498 2\_2 are concordant at  $462 \pm 31$  Ma. Cycle 1 of analysis 2\_3 is discordant with a  $^{206}\text{Pb}/^{238}\text{U}$  age of  
499  $713 \pm 56$  Ma. For all three analyses the innermost cycles (approximately 700 nm depth) are  
500 discordant. The gentle slope of results from the youngest to the oldest ages should not be taken  
501 as actual ages and represent isotopic mixing during analysis. There may be step functions along  
502 the depth profile that have been smoothed out due to age mixing (see discussion in Methods,  
503 above).

504           On the other hand, in some cases multiple depth profiles from the same grain such as  
505 those from 263A have the same ages (Fig. 7). All of the cycles from both analysis 1\_1 and 1\_2  
506 are statistically identical and concordant at  $414 \pm 24$  Ma and  $425 \pm 27$  Ma, respectively.  
507 Furthermore, the two analyses together have a concordia age of  $419 \pm 18$  Ma. Since the analysis  
508 stops at  $\sim 750$  nm there is no way to know how deep into the grain this age domain penetrates;  
509 however, given the lack of any spot analysis this young it is likely not deeper than a few  
510 micrometers.

511           The variation in extent or depth of alteration across zircon grains likely reflects the  
512 locations of pre-existing surface features that allowed for fluid infiltration and nanometer-scale  
513 recrystallization (Breeding et al. 2004). The isotopic variation between grains in the same sample  
514 can probably be attributed to the location of the zircon in the rock. In this study, as we have not

515 performed analyses *in situ*, we cannot tell if a zircon was located on a grain boundary or enclosed  
516 within a larger mineral. Whether or not a zircon is affected by thermal or fluid infiltration events  
517 likely depends on whether the heat or fluid has access to the zircon during the event.

#### 518 **CONCLUDING REMARKS**

519 Documentation and interpretation of zircon growth and recrystallization during Barrovian  
520 metamorphism in Scotland have been obtained using three different methods: conventional spot  
521 analysis of polished grains, short depth profiles (SDP) and a long depth profile (LDP).  
522 Conventional spot analyses of sectioned and polished grains demonstrates that zircon interiors  
523 retain inherited detrital ages from the Barrovian chlorite zone through the highest grades  
524 recorded in the sillimanite-K-feldspar zone. These ages are mostly between ca. 600 Ma and ca.  
525 2000 Ma, but Archean examples were also found. Metamorphism from chlorite to kyanite zone  
526 affected only the outer tens to hundreds of nm of zircon rims, if they were affected at all. Rims  
527 large enough for spot analysis—at least 20  $\mu\text{m}$  wide—only formed at and above the sillimanite  
528 zone.

529 In order to investigate the ages of smaller rim domains a different approach is necessary.  
530 The results of short depth profile (SDP) analysis in this study show that zircons commonly grow  
531 or recrystallize during metamorphism at lower grades, however the scale of the age domains is  
532 very small (the smallest resolution in this study is  $\sim 75$  nm). Some SDPs in zircons from the  
533 garnet through the sillimanite–K-feldspar zones revealed Grampian ages; however younger ages  
534 indicating post-peak growth/recrystallization as young as the Permian were also found.  
535 Importantly, the SDPs show that age domains on zircon rims are discontinuous even on the tens  
536 of nanometers scale.



537 Fig. 8 shows a summary cartoon of relevant zircon textures discussed in this paper.  
538 Interior porous domains that may or may not follow pre-existing zonation are seen at all  
539 metamorphic grades. In the sillimanite and sillimanite–K-feldspar zones, wide rim domains can  
540 be found that record the age of the peak metamorphism. The porous domains and wide rims  
541 probably formed by coupled dissolution-reprecipitation. In all metamorphic zones thin,  
542 discontinuous rim domains may be present that allow for age determinations by depth profiling  
543 but are too thin for spot analysis. These rims may preserve the ages of peak or post-peak activity.

544 Finally, whereas SDPs lead to some degree of isotopic mixing, long depth profiles  
545 (LDPs) can be used to resolve different ages with increasing depth. One grain in particular  
546 illustrates the utility of LDP analyses. By graphing the Th/U content along with the age, we are  
547 able to constrain the age of partial melting in the rocks from Glen Muick in the northern portion  
548 of Region II and conclude that the upper amphibolite–granulite facies metamorphism occurred  
549 during the Grampian Orogeny.

550 Results show growth/recrystallization during the Grampian event and during post-peak  
551 tectonic or magmatic events (Fig. 5). To summarize, the combined Grampian ages are:

- 552 • Grampian spot and SDP analyses from Region II, not including the Glen Muick  
553 samples (n=7):  $468 \pm 10$  Ma
- 554 • Grampian spot and SDP analyses from Glen Muick (n=8):  $472 \pm 5$  Ma
- 555 • Grampian SDP analyses from Region III: (n=4):  $483 \pm 25$  Ma
- 556 • All Grampian spot, SDP, and LDP analyses (n=19):  $472 \pm 4$  Ma

557 The post-peak ages and their likely causes are:

- 558 • ca. 450 Ma: Post-orogenic decompression melting, intrusion of S-type granites

- 559 • ca. 417 Ma: Subduction and intrusion of I-type granites such as the Lochnagar  
560 granite and Glen Doll diorite
- 561 • ca. 384 Ma: Possible effect of the Acadian Orogeny and intrusion of the Glen Tilt  
562 and Skene Granites
- 563 • ca. 335 Ma: Extension-related volcanism and vein remineralization; formation of  
564 the Clyde Plateau Basalts
- 565 • ca. 252 Ma: Rifting and basaltic magmatism

566 Most geochronological studies of Barrovian metamorphism have focused on the  
567 northeastern part of the sequence (Regions I and II herein). Our results are consistent with the  
568 interpretation that the entire Barrovian sequence, including the sillimanite-K-feldspar rocks of  
569 Glen Muick, underwent prograde metamorphism penecontemporaneously, further illustrating  
570 that the Grampian orogeny was geologically rapid (Oliver et al. 2000; Baxter et al. 2002; Dewey  
571 2005). Determining how the rates of critical geologic processes, including burial, exhumation,  
572 and thermal pulse activity, contributed to this rapid orogenesis poses an exciting set of  
573 challenges for future tectono-metamorphic studies.

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575

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## FIGURE CAPTIONS

779 **TABLE 1.** Ages of relevant igneous rocks within the study area including abbreviations

780 found in Fig. 1.

781

782 **FIGURE 1.** A. Map of the Scottish Highlands indicating metamorphic zones, locations of

783 igneous rocks, and western sample locations. The three major regions are I: along the eastern

784 coast, II: Glen Clova and surrounding glens, and III: the western half of the Highlands. B. Detail

785 of northeastern portion of study area showing sample locations from Regions I and II. All

786 samples include the prefix JAB, which has been omitted throughout the text. Key to

787 abbreviations and more information on each formation is given in Table 1. Modified from

788 Atherton (1977); Ague and Baxter (2007); BGS (2007); and Vorhies and Ague (2011).

789

790 **FIGURE 2.** BSE images of zircons from this study showing A. smooth, homogeneous core

791 with minor porous overgrowth, B. cracked grain, C. occasional porous grain interior, D. thin rim

792 domain, E. minor interior porosity, F. thin rim domain, G. interior porosity that does not appear

793 to follow pre-existing zonation, H. interior porosity, thin rim domain, I. nonporous Grampian age

794 rim domain wide enough for ion beam spot, J. porosity that follows zoning which resulted in a

795 highly uncertain, nearly discordant age, K. nonporous Grampian rim domain, and L. pre-existing

796 oscillatory zoning. Black ovals indicate analysis spots, where appropriate.

797

798 **FIGURE 3.** A. Concordia diagram showing all polished-grain spot analyses with

799 radiogenic Pb > 90%. Error ellipses are plotted at 1 $\sigma$ . Concordia ages indicated in Ma. Box

800 indicates extent of part B of this figure. B. An enlargement of the box on concordia diagram in  
801 A, focused on the area from 0-600 Ma.

802

803 **FIGURE 4.** A. Ages of spot analyses plotted against metamorphic zone. Grey line  
804 indicates approximate age of the Grampian Event. B. Ages of short depth profile (SDP) and long  
805 depth profile (LDP) analyses plotted against metamorphic zone. Grey line indicates approximate  
806 age of the Grampian Event.

807

808 **FIGURE 5.** Schematic of SDP and spot results. Grampian results illustrate the  
809 synchronicity of orogenic activity across the study area. Zircon ages from this study indicate that  
810 the rocks have been affected by four post-peak tectonic events. Error bars and width of grey bar  
811 indicate  $2\sigma$  uncertainty.

812

813 **FIGURE 6.** Long depth profile (LDP) from sillimanite–K-feldspar zone sample 286B  
814 (Glen Muick).  $^{206}\text{Pb}/^{238}\text{U}$  and  $^{207}\text{Pb}/^{235}\text{U}$  ages and Th/U content plotted against profile depth into  
815 grain.

816

817 **FIGURE 7.** Examples of depth profiles showing the differences in results from three  
818 analyses on a single grain from sample 41A and the identical results from two analyses on the  
819 same grain from sample 263A.

820

821           **FIGURE 8.** Cartoon illustrating the difference between rim domains in the chlorite-kyanite  
822 zones and sillimanite-sillimanite–K-feldspar zones. Zircon interiors are of pre-Grampian age.  
823 Thin, discontinuous overgrowths may be of Grampian or post-Grampian age.  
824



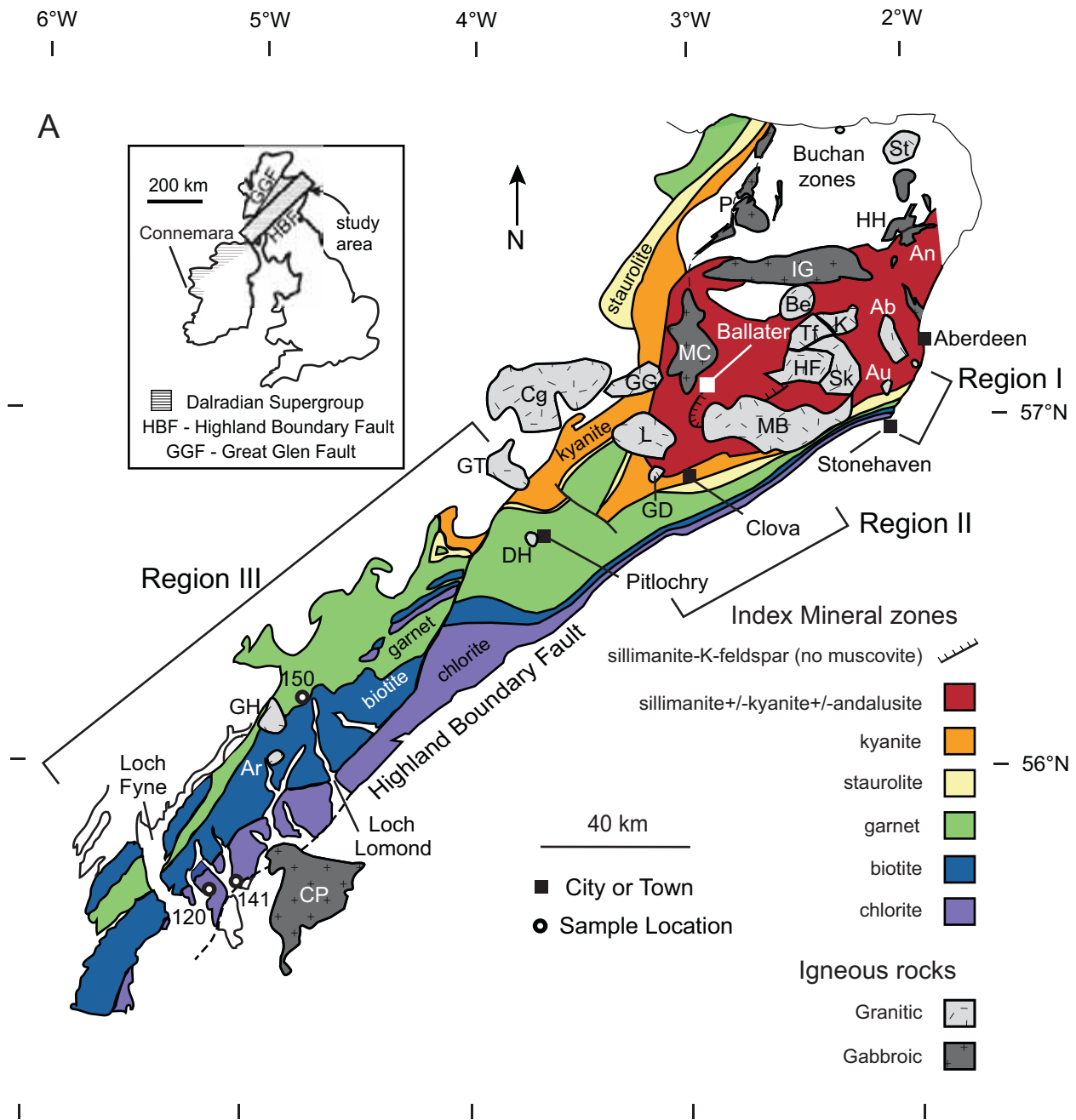


Fig. 1 A

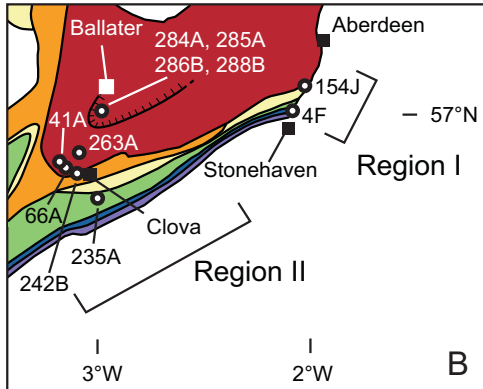


Fig. 1B

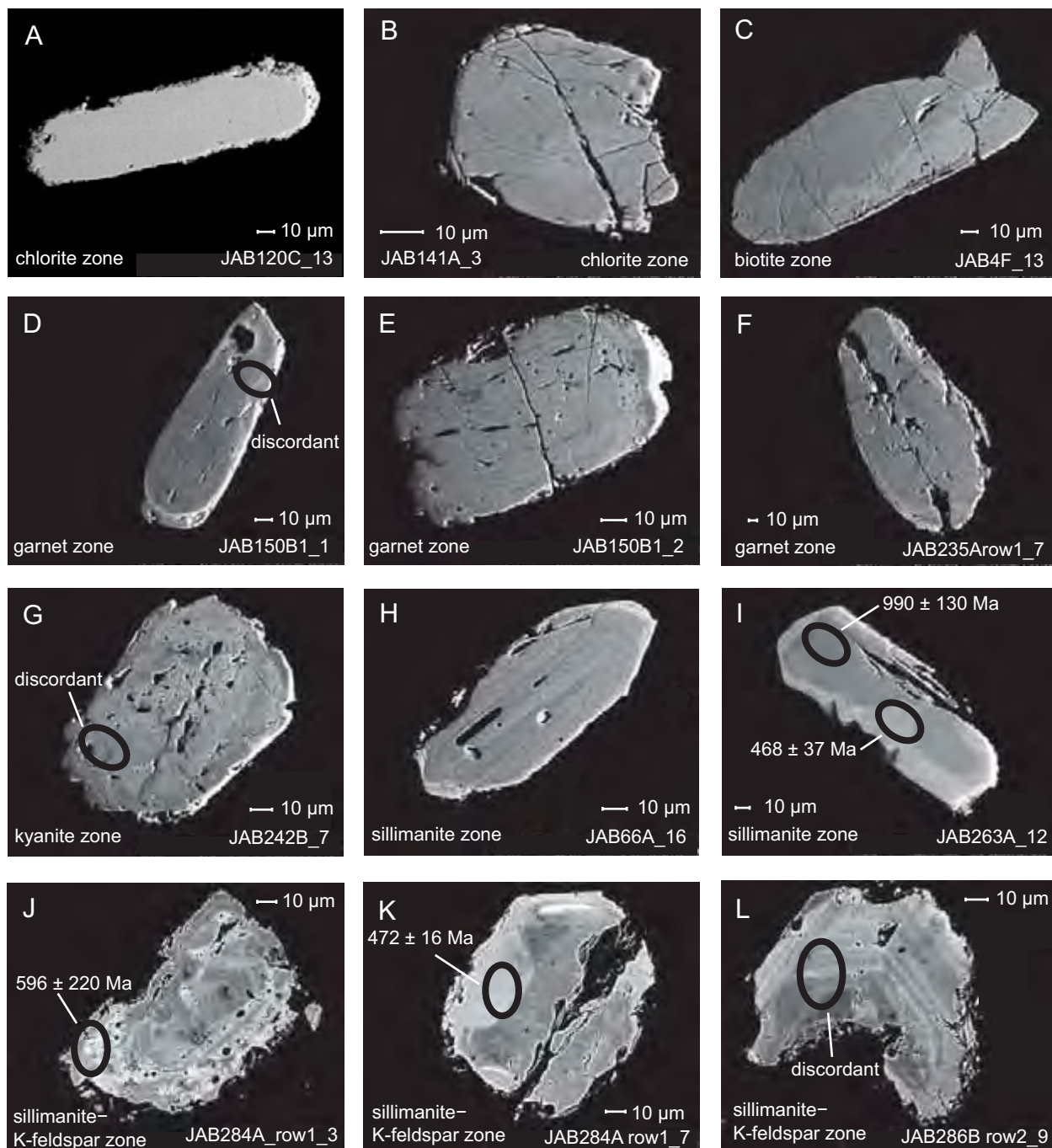


Fig. 2

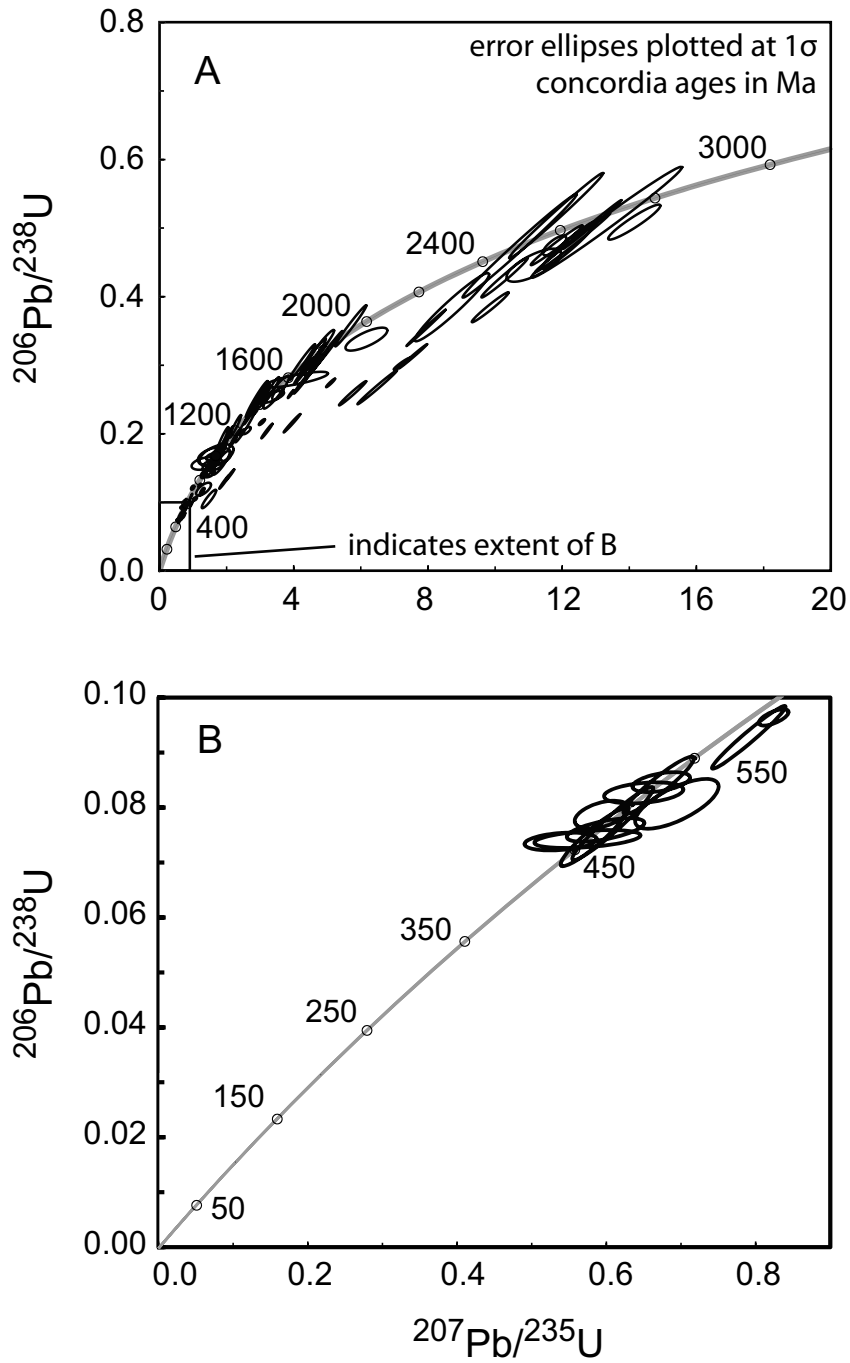


Fig. 3

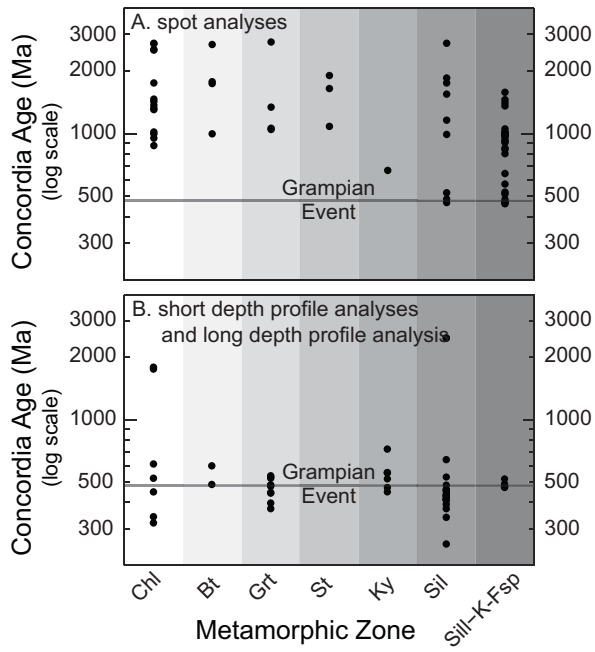


Fig. 4

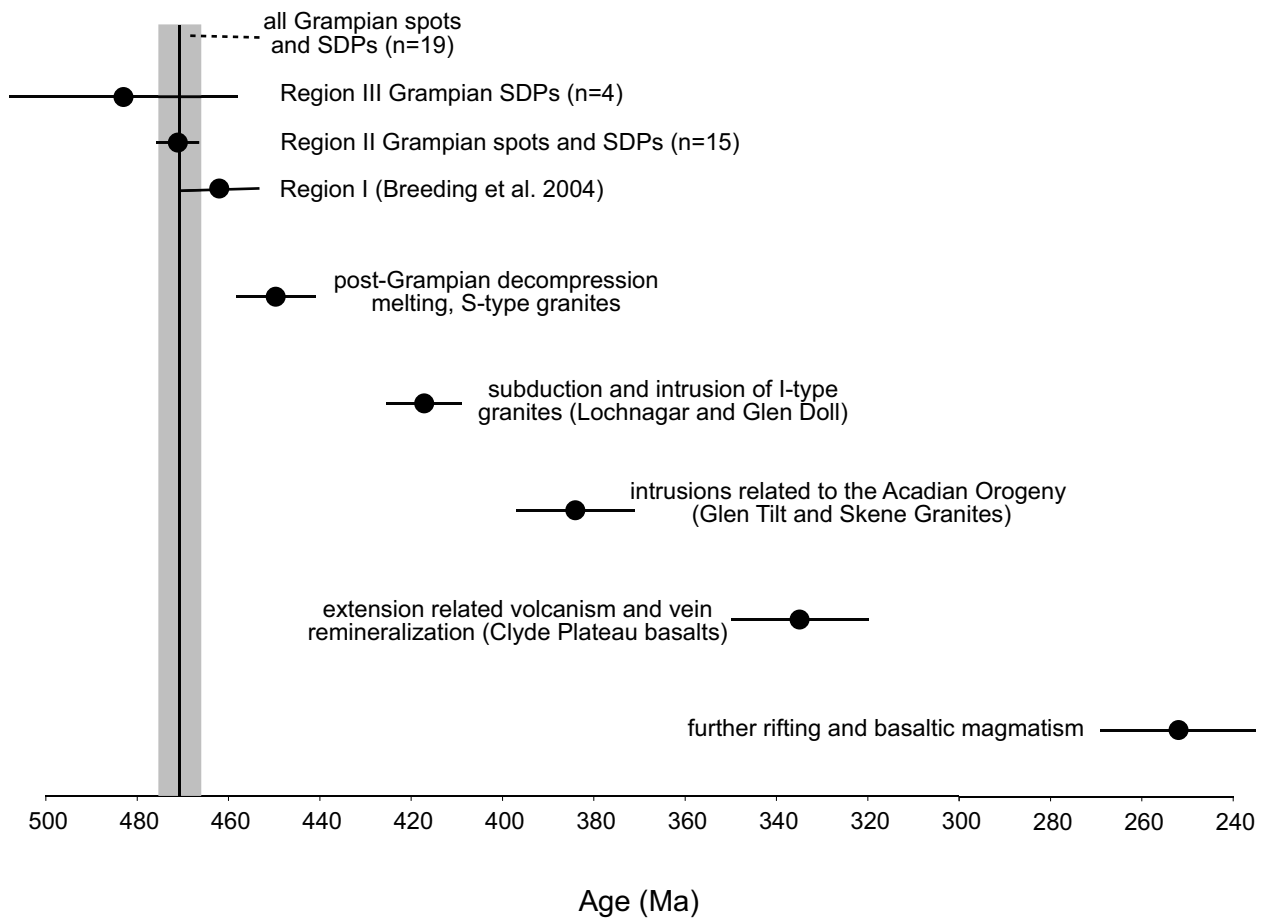


Fig. 5

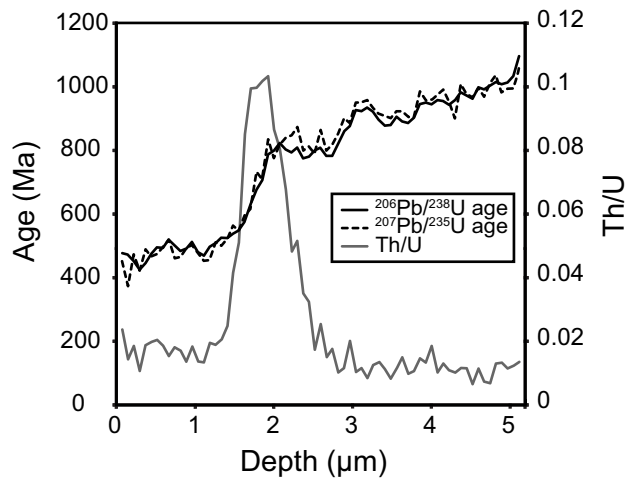


Fig. 6

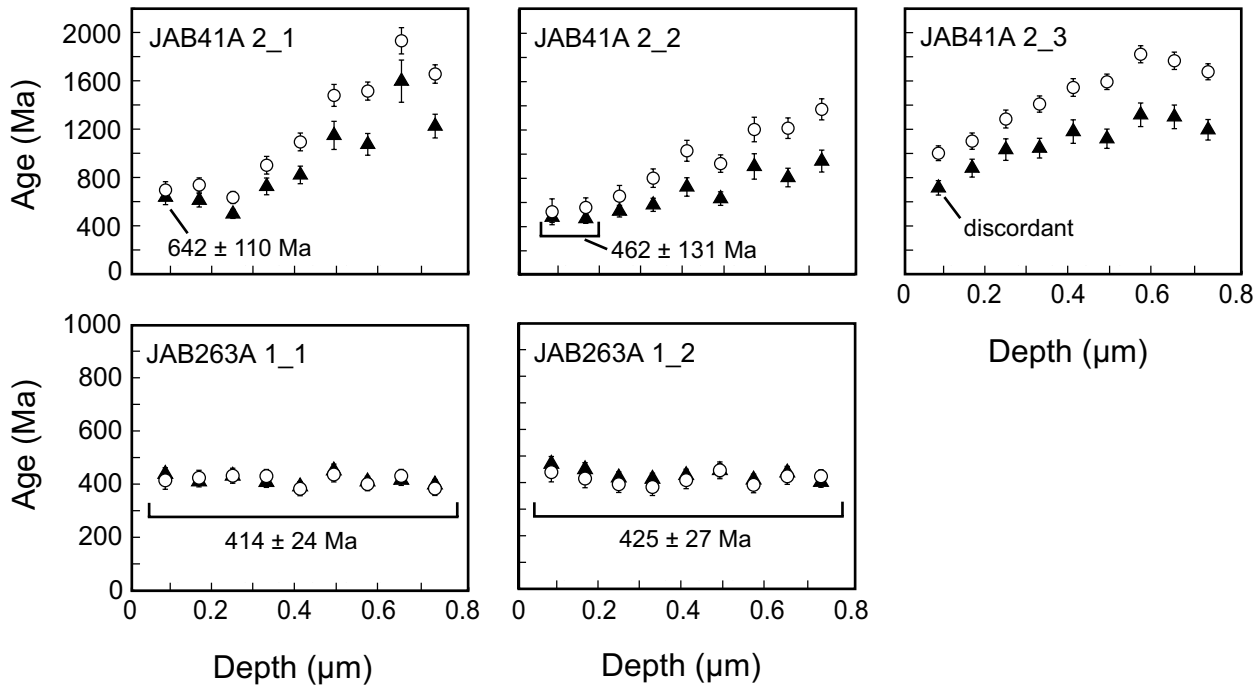
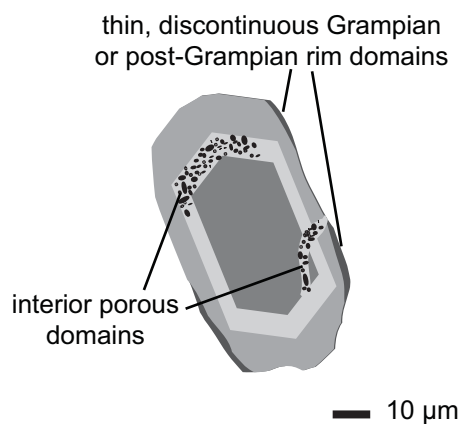
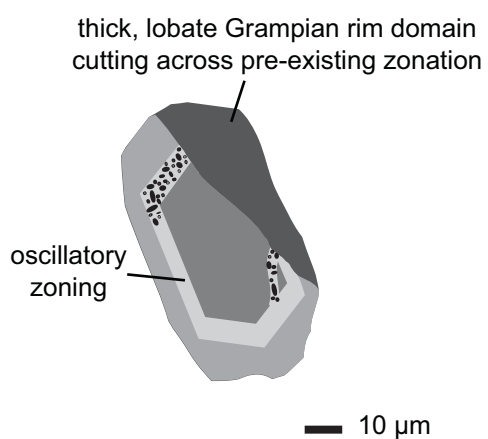


Fig. 7





chlorite - kyanite zones



sillimanite - sillimanite-K-feldspar zones

Fig. 8

**TABLE 1.** Ages of relevant igneous rocks within the study area

Age (Ma)	±	Name/Location	Fig. 2 Abbr.	Rock Type	Technique	Reference
491	15	Dunfallandy Hill	DH	granite	Rb-Sr whole rock	Pankhurst and Pidgeon, 1976
487	23	Haddo House	HH	gabbro	Rb-Sr whole rock on metamorphic aureole	Pankhurst, 1970
482	12	Arnage	An	granites /gneisses	Rb-Sr whole rock	Pankhurst, 1970
475	12	Auchlee	Au	granite	U-Pb zircon	Oliver et al., 2008
474	2	Portsoy	P	gabbro	U-Pb zircon	Martin and Condon in Oliver et al., 2008
472	<i>n.d</i>	Morven Cabrach	MC	gabbro	U-Pb zircon	Rogers et al., 1994
471	12	Tillyfourie	Tf	granite	U-Pb zircon	Oliver et al., 2008
470	1	Aberdeen	Ab	granite	U-Pb monazite	Kneller and Aftalion, 1987
470	9	Insch	IG	gabbro	U-Pb zircon	Dempster et al., 2002
468	<i>n.d</i>	Insch	IG	gabbro	U-Pb zircon	Rogers et al., 1994
467	6	Strichen	St	granite	U-Pb zircon	Oliver et al., 2000
457	1	Kennethmont	K	granite	U-Pb zircon	Oliver et al., 2000
429	2	Garabal Hill	GH	appinite	U-Pb zircon	Rogers and Dunning, 1992
426	3	Arrocher	Ar	appinite	U-Pb titanite	Rogers and Dunning, 1991
420	2	Lochnagar	L	granite	U-Pb zircon	Abbleby (pers.comm.) from Oliver et al., 2008
419	5	Glen Doll	GD	diorite	U-Pb zircon	Oliver et al., 2008
415	1	Glen Gairn	GG	granite	U-Pb monazite	Parry (pers. comm.) from Oliver et al., 2008
408	5	Bennachie	Be	granite	U-Pb zircon	Oliver et al., 2008
406	5	Mount Battock	MB	granite	U-Pb zircon	Oliver et al., 2008
404	18	Cairngorn	Cg	granite	U-Pb zircon	Oliver et al., 2008
403	8	Hill of Fare	HF	granite	U-Pb zircon	Oliver et al., 2008
396	6	Skene	Sk	granite	U-Pb zircon	Oliver et al., 2008
390	5	Glen Tilt	GT	granite	U-Pb zircon	Oliver et al., 2008
335	1	Clyde Plateau	CP	trachyandesite lava	U-Pb zircon	Monaghan and Parrish, 2006

*Note* : Abbreviations indicate locations on Fig. 2.

*n.d*: indicates that no uncertainty data was provided