

1 **Provenance of the Highland Border Complex: constraints on Laurentian margin**  
2 **accretion in the Scottish Caledonides**

3 **P.A. Cawood<sup>1,2</sup>, R.E. Merle<sup>2,3</sup>, R.A. Strachan<sup>4</sup> and P.W.G. Tanner<sup>5</sup>**

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6 <sup>1</sup> Department of Earth Sciences, University of St. Andrews, North Street, St. Andrews  
7 KY16 9AL, UK ([peter.cawood@st-andrews.ac.uk](mailto:peter.cawood@st-andrews.ac.uk))

8 <sup>2</sup> School of Earth and Environment, University of Western Australia, WA 6009,  
9 Australia

10 <sup>3</sup> Dipartimento di Geoscienze, Universita di Padova, 35100 Padova, Italy

11 <sup>4</sup> School of Earth & Environmental Sciences, University of Portsmouth, Portsmouth,  
12 PO1 3QL, UK.

13 <sup>5</sup> Department of Geographical and Earth Sciences, University of Glasgow, Glasgow,  
14 G12 8QQ.  
15

16 **Abstract**

17 Contrasting tectonic models for the Highland Border Complex in the Scottish  
18 Caledonides view it either as part of the rifted Laurentian margin of the Iapetus Ocean  
19 or as an oceanic terrane. Detrital zircon data from sandstones of the complex yield age  
20 peaks at 2.8-2.6, 1.3 and 1.1-1.0 Ga and minor peaks of ~1.7-1.9 Ga. These  
21 characteristics compare closely with the upper Dalradian Supergroup of the adjacent  
22 Grampian terrane, and with the record of eastern Laurentia. The data are also  
23 consistent with the Laurentian provenance indicated by palaeontological evidence  
24 from the complex, and field evidence for continuity with the Dalradian Supergroup.  
25 Detrital ages for the Cambrian Salterella Grit of the Caledonian foreland compare  
26 with that for approximately age-equivalent sandstone from the Highland Border  
27 Complex. Both were contemporaneous with the regressive Hawke Bay event,  
28 accounting for similarities in provenance, and further linking the Highland Border  
29 Complex to Laurentia. The Grampian terrane was being uplifted and shedding  
30 detritus throughout the Ordovician and Silurian. The absence of this event from the

31 detrital zircon records of either the Midland Valley or Southern Upland terranes,  
32 suggests that these blocks cannot have been in their current location relative to the  
33 Grampian terrane before the end Silurian.

#### 34 **Introduction**

35 Establishing the existence and origin of lithotectonic successions (terrane) outboard  
36 of the continental margin within orogenic belts requires a variety of data sets,  
37 including stratigraphic, biogeographic, geochronological, geochemical and  
38 paleomagnetic. Increasingly, detrital zircon data, for which tens of thousands of  
39 analyses are now available worldwide, are being used to characterize the nature of the  
40 foreland source regions, which then provides a basis for comparison with the detrital  
41 zircon records of internal terranes. Such comparisons provide a benchmark to  
42 constrain the potential degree of allochthoneity of terranes and the paleogeography of  
43 lithotectonic elements of the orogen (e.g. Gehrels *et al.* 1995, 1996; Colpron &  
44 Nelson 2009).

45         In this paper detrital zircon data are used to assess the provenance and tectonic  
46 setting of the Highland Border Complex, a controversial sequence of sedimentary and  
47 igneous rocks within the Scottish Caledonides. The Highland Border Complex in  
48 Scotland occurs along the junction between the Grampian and Midland Valley  
49 terranes (Fig. 1; Bluck 1983, 1984 and references therein), and correlative lithologies  
50 may also be exposed along strike in the same structural setting in west Ireland (Chew  
51 2003). The Grampian terrane is underlain mainly by a thick sequence of deformed and  
52 metamorphosed mid-Neoproterozoic to early Ordovician shallow- and deep-water  
53 sedimentary and volcanic rocks, the Dalradian Supergroup. Deposition occurred on  
54 the eastern margin of Laurentia during the breakup of Rodinia and evolution of the  
55 Iapetus Ocean (Anderton 1985; Harris *et al.* 1994; Strachan *et al.* 2002; Leslie *et al.*

56 2008). In contrast, the Midland Valley terrane to the southeast is thought to be  
57 underlain by a volcanic arc that developed in the Iapetus Ocean and collided with the  
58 Laurentian margin during the mid-Ordovician Grampian orogenic event (Lambert &  
59 McKerrow 1976; Dewey & Shackleton 1984; Dewey & Ryan 1990; Bluck 2002). The  
60 intervening Highland Boundary Fault is currently interpreted as a steep reverse fault  
61 that formed during the Devonian, and presumably obscures an older Ordovician  
62 suture between Laurentia and the oceanic terranes (Tanner 2008; cf. Soper & Hutton  
63 1984; Dewey & Strachan 2003). The low-grade Ordovician to Silurian sedimentary  
64 rocks of the Southern Uplands terrane farther to the southeast represent an  
65 accretionary prism developed above a NW-dipping subduction zone (McKerrow *et al.*  
66 1977; Leggett *et al.* 1979; Stone & Merriman 2004).

67         The Highland Border Complex occupies a zone <1.3 km wide and 250 km  
68 long, located adjacent to the Highland Boundary Fault and between the Dalradian  
69 Supergroup to the northwest and an unconformable cover of late Silurian-Devonian  
70 strata (Old Red Sandstone facies) to the southeast (Fig. 1). It consists of a diverse  
71 sequence of weakly deformed, low-grade metasedimentary rocks of Cambrian-  
72 Ordovician age, associated locally with pillow lavas and with ultramafic rocks of the  
73 Highland Border Ophiolite. Two contrasting tectonic models have been proposed for  
74 the Highland Border Complex. One views it as an exotic oceanic terrane that was  
75 once considerably separated from the Dalradian Supergroup (Curry *et al.* 1984; Bluck  
76 2002), whereas the other divides it into two parts: a newly defined Trossachs Group,  
77 viewed as the youngest part of the Dalradian Supergroup, and the allochthonous  
78 Highland Border Ophiolite (Tanner & Sutherland 2007).

79         In order to test these conflicting models, we present analyses of detrital  
80 zircons obtained from four samples from the Highland Border Complex and compare

81 them with published data-sets from the Dalradian Supergroup (Cawood *et al.* 2003).  
82 In addition, we present new detrital zircon data from a sample of the Cambro-  
83 Ordovician shallow-water sedimentary succession that accumulated on the Hebridean  
84 foreland of the Scottish Caledonides (Fig. 1). This succession is broadly time-  
85 equivalent to upper deep-water parts of the Dalradian Supergroup as well as the  
86 Highland Border Complex. Its provenance is relevant to the origins of the latter as  
87 well as informing wider debate on basin evolution and sedimentary drainage patterns  
88 on the Laurentian margin (Cawood *et al.* 2007a, b). We then integrate our results into  
89 a brief overview of the assembly and accretion of terranes onto the margin of  
90 Laurentia.

91

## 92 **Geological setting of the Highland Border Complex**

93 The stratigraphical succession and age of the Highland Border Complex and its  
94 relationship to the Dalradian Supergroup are highly controversial issues (Curry *et al.*  
95 1984; Bluck 1983, 1984, 1985, 1990; 2002, 2010, 2011; Bluck *et al.* 1992; Tanner  
96 1995, 1997, 1998, 2011; Bluck & Ingham 1997; Tanner & Pringle 1999; Tanner &  
97 Sutherland 2007). The rationale behind the view that the Highland Border Complex  
98 developed as an exotic oceanic terrane some distance from the Laurentian margin can  
99 be summarised as follows (Tanner & Sutherland 2007). Radiometric and  
100 palaeontological data available in the 1980s showed that the Dalradian Supergroup  
101 was undergoing post-orogenic uplift and rapidly shedding detritus at the same time as  
102 the (now contiguous) Dounans Limestone in the Highland Border Complex at  
103 Aberfoyle with an Arenig (~470 Ma) fauna, was being deposited (Curry *et al.* 1984;  
104 Bluck 1985, 1990; Bluck *et al.* 1992). The Highland Border Complex was thought to  
105 be underlain by an ophiolite, and as the structurally overlying Cambrian-Upper

106 Ordovician sequence contained no Dalradian detritus, it was concluded that these  
107 sediments were deposited far from the Laurentian margin. According to this model,  
108 the Highland Border Complex was first proximal to the Dalradian Supergroup in late  
109 Silurian-early Devonian times after the former had been deformed and  
110 metamorphosed during the mid-Ordovician Grampian orogenic event. The spatial  
111 distribution of stratigraphical ages across the Highland Border Complex outcrop,  
112 mainly from chitinozoa, appeared to show that it youngs towards the Dalradian  
113 (Bluck 2002).

114 In contrast to the exotic terrane model, the possibility of stratigraphical (and  
115 structural) continuity between undoubted Dalradian rocks and the Highland Border  
116 Complex was first recognised by Clough (cited by Geikie 1897) and developed by  
117 Johnson & Harris (1967). The case for this interpretation was strengthened further as  
118 follows.

119 1) Way-up structures, although uncommon, invariably show that the Highland  
120 Border Complex youngs away from the Dalradian Supergroup (e.g. Tanner  
121 1995, see however Bluck 2002, 2010, 2011). For example, Tanner (1995)  
122 demonstrated that at Keltie Water, the Ben Ledi Grits (Southern Highland  
123 Group) are in stratigraphic continuity, albeit with a possible cryptic non-  
124 conformity, with the Keltie Water Grit Formation (formerly Highland Border  
125 Complex), which includes the Lower Cambrian Leny Limestone. There is also  
126 evidence for stratigraphical continuity between the Dalradian rocks and the  
127 Highland Border Complex at Glen Sannox, Isle of Arran and the River North  
128 Esk (Fig. 2; Tanner 1995).

129 2) Throughout the Highland Border Complex and the adjacent Dalradian rocks of  
130 the 'steep belt', the early penetrative cleavage has the same relationship to

131 bedding: S1 faces down irrespective of whether the rocks are right-way-up or  
132 have been inverted by later D4 folding (Tanner 1995, see also Johnson &  
133 Harris 1967). It follows from this that the Highland Border Ophiolite is  
134 located at the *structural top* of the Highland Border Complex. .

135 3) Re-assessment of the supposed chitinozoa featured in publications on the  
136 Highland Border Complex (Burton et al. 1983), has concluded that this  
137 evidence is unreliable (Tanner & Sutherland 2007). It remains the case that  
138 precisely constrained stratigraphical ages are available for only three localities,  
139 at each of which the fossil faunas have Laurentian affinities. Trilobites from  
140 the Leny Limestone at Callander (Fig. 2) yielded a late Early Cambrian (~515  
141 Ma) age; trilobites and conodonts from the Dounans Limestone at Aberfoyle  
142 (Fig. 2) gave a middle Arenig (~470 Ma) age (Rushton & Owen 1999); and  
143 the conodonts from the Margie Limestone of the River North Esk gave a late  
144 Tremadocian to lowermost Arenig age (Ethington 2008). Consequently, there  
145 are no sedimentary rocks of proven age younger than Arenig in the Highland  
146 Border Complex (Tanner & Sutherland 2007).

147 4) The results of modern isotopic dating indicate that the Grampian orogenic  
148 event was relatively short-lived and occurred at 475-465 Ma (Oliver et al.  
149 2000; Chew *et al.* 2010 and references therein). This is consistent with the  
150 palaeontological evidence that the Trossachs Group does not include any strata  
151 younger than the Arenig. Published Cambrian to early Ordovician K-Ar and  
152 Rb-Sr mineral ages obtained from the Dalradian Supergroup (Dempster 1985;  
153 Dempster *et al.* 1995) are likely unreliable and of uncertain significance  
154 (Evans & Soper 1997; Tanner & Pringle 1999).

155 Given the unreliability of the chitinozoan ages, recognition that the Highland  
156 Border Ophiolite lies at the top of the Highland Border Complex, and tighter  
157 constraints on the age of the Grampian orogenic event, a radically different  
158 stratigraphical template of the sequence has emerged (Fig 3; Tanner & Sutherland  
159 2007). In this model, the Highland Border Complex is now divided into two parts  
160 (Figs 2, 3): an older Trossachs Group in continuity with, and part of, the Dalradian but  
161 probably containing undetected unconformities and/or non-conformities, and an upper  
162 sequence comprising the obducted ophiolite.

163 Recent reappraisal of the Highland Border Ophiolite has concluded that much of it  
164 comprises serpentized continental mantle and associated sediments, which formed  
165 part of the floor to an extended Dalradian basin, and was thrust on to the Trossachs  
166 Group immediately prior to the start of the Grampian orogeny (Tanner 2007:  
167 Henderson *et al.* 2009). The ophiolite is thus likely to have formed at the rift-drift  
168 transition of the Iapetan rifting cycle, which on the basis of regional considerations  
169 probably occurred at ~540 Ma (Cawood *et al.* 2001). However, this new view of the  
170 'ophiolite' is not critical to the current debate concerning the affinities of the  
171 sedimentary rocks assigned to the Trossachs Group by Tanner & Sutherland (2007).

172

### 173 **Geology of the Cambro-Ordovician succession on the Laurentian foreland in** 174 **Scotland**

175 Passive margin strata related to the final break-up of Rodinia are represented by the  
176 Cambrian to Ordovician sedimentary rocks in the Hebridean (Laurentian) foreland of  
177 Scotland, northwest of the Moine Thrust (Fig. 1; Park *et al.* 2002 and references  
178 therein). The succession ranges in age from Early Cambrian to Middle Ordovician. It  
179 can be divided into a lower, mainly clastic part, the Ardvreck Group (~250 m thick),

180 and an upper carbonate sequence, the Durness Group (~750 m thick). The Ardvreck  
181 Group is Early Cambrian and composed of the Eriboll and An t-Sron formations (Fig.  
182 3), each of which consists of two members. The oldest unit, the 75-125 m thick Basal  
183 Quartzite Member of the Eriboll Formation, is a feldspathic to quartzitic sandstone  
184 with locally abundant cross-bedding (McKie 1990). The overlying 75-100 m thick  
185 Pipe Rock Member is a quartz arenite characterised by abundant *Skolithos* burrows.  
186 The An t-Sron Formation consists of the lower dolomitic siltstones of the Furoid  
187 Beds, 12-27 m thick, and the upper quartz arenites of the Salterella Grit, up to 20 m  
188 thick. The Furoid Beds contain Early Cambrian *Olenellus* trilobites and brachiopods  
189 which indicate an absolute age of ~520 Ma (Prigmore & Rushton 1999). The  
190 Ardvreck Group has been interpreted by McKie (1990) as a transgressive sequence of  
191 barrier island to tidal shelf arenites (Eriboll Formation) overlain by storm-dominated  
192 clastic carbonate sediments (Furoid Bed) and regressive sandsheets (Salterella Grit).

193       The Ardvreck Group is interpreted to be time-equivalent to the deep-water  
194 (meta) sedimentary rocks of parts of the Southern Highland Group and the Highland  
195 Border Complex. In particular, the Salterella Grit is essentially the same late Lower  
196 Cambrian age (~515 Ma; Smith & Rasmussen 2008) as the Leny Limestone (Fig. 3).  
197 Strachan & Holdsworth (2000) suggested that the foreland succession and the upper  
198 parts of the Southern Highland Group and the Highland Border Complex represent,  
199 respectively, shallower and deeper parts of the same passive margin basin. Detrital  
200 zircon data from the Ardvreck Group has so far been published only from the Eriboll  
201 Formation (Cawood *et al.* 2007b).

202

203 **Sample descriptions**



204 Five samples were collected for detrital zircon analysis: three from the Highland  
205 Border Complex (Trossachs Group); one from the Highland Border Ophiolite; and  
206 one from the Cambrian succession of NW Scotland. In order of inferred decreasing  
207 ages they are as follows:

208         Sample **A266** is a reddish-brown sub-litharenite sampled from the Highland  
209 Border Ophiolite in the Loch Ard Forest, near Aberfoyle (Fig. 2) [NN 46808 96083].  
210 This sample was taken from a stratified arenite that lies between the banded  
211 amphibolite at the base of the ophiolite, and a thick unit of serpentine conglomerate  
212 that constitutes the main body of the ophiolite. An approximate age of ~540 Ma is  
213 inferred on the basis of the suggested age of formation of the ophiolite (see above),  
214 which formed part of the floor of the Dalradian basin. The sample consists of  
215 irregularly shaped, unsorted, matrix-supported quartz and lithic clasts (to 1.5 mm) set  
216 in a matrix of sericite, with stringers and patches of indeterminate opaque material,  
217 some of which appears to be oxidized serpentinite. The lithic clasts consist largely of  
218 mosaic or multi-grain quartz or of dark, unidentified material.

219         Sample **A265** is a grey psammite from the transition between the Southern  
220 Highland Group and the Trossachs Group in the Loch Ard Forest, near Aberfoyle  
221 (Figs. 2, 3) [NN 45579 96150]. This sample is inferred to be slightly older than the  
222 Lower Cambrian Leny Limestone, and is included because it further extends the  
223 vertical range of Dalradian strata from which detrital zircon analyses have been  
224 obtained (Cawood *et al.* 2003). This rock consists of quartz clasts (0.1 – 0.2 mm  
225 across) with some plagioclase grains, set in a matrix of sericite, chlorite, quartz,  
226 plagioclase and opaque minerals. Also present are numerous flakes of detrital white  
227 mica, of the same size as the quartz grains.

228           Sample **A472** is an arenite from the topmost part of the Lower Keltie Water  
229 Formation. It lies stratigraphically immediately below the late Lower Cambrian  
230 (~515 Ma) Leny Limestone of the Trossachs Group in Leny Quarry near Callander  
231 (Figs. 2, 3) [NN 6157 0988]. This sample is clast-supported and consists of quartz  
232 and plagioclase (to 0.5 mm across) accompanied by detrital white mica of the same  
233 size and set in a matrix that consists largely of sericite, quartz, plagioclase and opaque  
234 grains.

235           Sample **SG07** was collected from the middle of the late Lower Cambrian  
236 Salterella Grit (~515 Ma) on the east shore of Loch Eriboll (Fig. 1) [NC 4416 5807].  
237 The sample is a grey, medium-grained, equigranular quartz sandstone. It is clast-  
238 supported and consists mainly of sub-rounded grains of quartz with interstitial  
239 carbonate cement.

240           Sample **NE148** is a quartz arenite from the Lower Margie Grit in the River  
241 North Esk, near Edzell (Fig. 2) [NO 58663 73310]. The sample was taken from  
242 immediately southeast of, and stratigraphically above, the Margie Limestone from  
243 which a conodont assemblage of uppermost Tremadoc to lowermost Floian (~478  
244 Ma) was reported by Ethington (2008). This sample consists of sub-angular quartz  
245 clasts (to 0.5mm) and detrital white micas of similar size, set in a carbonate-  
246 dominated matrix. There are some lithic grains, mainly of composite quartz.

247

#### 248 **Analytical procedures**

249           For each sample, 1 to 5 kg of rock was crushed and the zircons were extracted  
250 using LST heavy liquid and Frantz magnetic separator. From the non-magnetic, heavy  
251 fraction of each sample, approximately 100 zircons were selected by hand picking  
252 under binocular and mounted in an epoxy resin. The mounts were polished to expose

253 the interior of the zircons. All the grains were investigated to characterize the possible  
254 internal structures by cathodoluminescence imaging (CL) using a Philips XL 30  
255 scanning electron microscope at Curtin University. The acceleration voltage applied  
256 during the several sessions, ranged between 12 and 20 KV. The U-Pb dating work  
257 was performed with a high-resolution ion microprobe (Shrimp II instrument) at Curtin  
258 University. The analytical conditions were identical to those described by Compston  
259 *et al.* (1984), Nelson (1997) and Williams (1998). The samples were analysed over  
260 several sessions during which the intensity of the O<sup>2-</sup> primary beam was set between  
261 0.8 nA and 5.3 nA. Pb/U and Pb/Th calibration was performed relative to the 556 Ma-  
262 old CZ3 zircon standard (Pidgeon *et al.* 1994) which was analyzed after every five  
263 unknowns throughout the analytical session. The raw data were reduced using the  
264 Squid macro (Ludwig, 2001) and the plots made with Isoplot (Ludwig, 2003).  
265 Common Pb was corrected for by using <sup>204</sup>Pb using the present-day terrestrial ratios  
266 from Stacey & Kramer (1975). Although less precise than corrections based on <sup>207</sup>Pb  
267 or <sup>208</sup>Pb for young zircons, this correction can be applied to both old and young grains  
268 and hence results in consistency of data treatment. The errors were calculated by  
269 propagating analytical uncertainties and are shown as 1-sigma level.

270 The number of scans of each mass for a single analysis was reduced from  
271 seven to five in order to investigate approximately 50-60 grains during each session.  
272 This number of analysed grains provides a 95% probability of finding any population  
273 that makes up 5% of the total (Dodson *et al.* 1988). Considering that the main purpose  
274 of this work is to decipher the provenance of the zircons, the cores of the grains were  
275 preferentially investigated.

276

277 **Results**

278 Concordia and frequency distribution plots for the individual samples are presented in  
279 Figures 4 and 5. The complete analytical data set can be obtained from the Society  
280 Library or the British Library Document Supply Centre, Boston Spa, Wetherby, West  
281 Yorkshire LS23 7BQ, UK as Supplementary Publication No. SUPxxx (yy pp.). It is  
282 also available online at <http://www.geolsoc.org.uk/SUPxxx>. The probability density  
283 distribution diagrams of the ages have been constructed following the methodology of  
284 Nemchin & Cawood (2005). This involves double weighting of the data based on the  
285 probability of concordance and errors highlighting the most concordant data.

286         The grains extracted from the samples have an average size of 150  $\mu\text{m}$ , with  
287 occasional grains exceeding 200  $\mu\text{m}$  in length. The grains display smoothed faces  
288 with a length-width ratio of 2:1. Rounded morphologies are common excepting in  
289 sample SG07 where they are dominant. The cathodoluminescence images revealed a  
290 large range of internal structures. Oscillatory zoning indicative of a magmatic origin  
291 is common but complex growth patterns reflecting local resorption also occur. Faint  
292 zoning and multi-stage dissolution and growth zoning features are widely observed.  
293 Homogeneous, dark, zoned grains are rare.

294

295 *Sample A266 (arenite from Highland Border ophiolite)*

296 55 analyses were made in 53 grains with ages ranging from  $962 \pm 17 \text{ Ma}$  ( $^{206}\text{Pb}/^{238}\text{U}$   
297 age,  $1\sigma$ ) to  $3208 \pm 16 \text{ Ma}$  ( $^{207}\text{Pb}/^{206}\text{Pb}$  age,  $1\sigma$ ). The uranium and thorium content and  
298 the Th/U ratio are generally in the range of igneous zircons (U = 24-608 ppm, Th = 1-  
299 235 ppm, Th/U = 0.23-1.89 with only one analysis having a Th/U = 0.01) and  
300 variations are not related to the age of the grains.

301 Six analyses have discordance higher than 10%. On the frequency distribution  
302 diagram (Fig. 5), two main peaks are centred at ~1050 Ma and ~2700 Ma.  
303 Subordinate peaks occur at 1350 Ma, 2350 Ma, 2950 Ma and 3200 Ma.

304

305 *Sample A265 (psammite from transition between Southern Highland and Trossachs*  
306 *groups)*

307 42 analyses were made on 38 grains. The uranium content of the grains ranges from  
308 25 to 965 ppm, the thorium content from 13 to 808 ppm and the Th/U ratio is  
309 generally from 0.28 to 1.31, which overlaps the range of expected values for  
310 magmatic zircons. Three analyses with a Th/U lower than 0.28 (~ 0.055) display very  
311 high discordance. No correlation between the chemical composition and age was  
312 observed.

313 Ages of detrital zircon grains range from  $621 \pm 16$  Ma ( $^{206}\text{Pb}/^{238}\text{U}$  age,  $1\sigma$ ) to  
314  $2901 \pm 5$  Ma ( $^{207}\text{Pb}/^{206}\text{Pb}$  age,  $1\sigma$ ). Nineteen analyses display discordance higher than  
315 10%. Following the procedure of Nemchin & Cawood (2005), the frequency diagram  
316 (Fig. 5) shows four main peaks at ca. 600 Ma, 1050 Ma, 1400 Ma and 2700 Ma. Two  
317 minor peaks are observable at 1200 Ma and 1700 Ma.

318

319 *Sample A472 (arenite adjacent to Leny Limestone)*

320 55 analyses were made on 53 grains from sample A472. Uranium and thorium  
321 contents (16-546 ppm and 5-294 ppm respectively) and their Th/U ratios (main range:  
322 0.23-1.88 with three outliers) are similar to those of the other samples.

323 The ages range from  $1158 \pm 15$  Ma ( $^{206}\text{Pb}/^{238}\text{U}$  age,  $1\sigma$ ) to  $2973 \pm 9$  Ma  
324 ( $^{207}\text{Pb}/^{206}\text{Pb}$  age,  $1\sigma$ ) with only seven analyses having discordance higher than 10%.

325 On a Concordia diagram (Fig. 4), the analyses plot close the curve in three groups at

326 ~1200 Ma, ~2000 Ma and ~2800 Ma. On the frequency plot two main populations are  
327 present at ~2000 Ma and ~2700 Ma but with ages spread over 400 Ma (Fig. 5). Three  
328 minor peaks can be distinguished at ~1050 Ma, ~1450 Ma and ~1700 Ma.

329

330 *Sample SG07 (Salterella Grit sandstone)*

331 60 analyses were performed in 55 grains from sample SG07 and yielded ages ranging  
332 from  $1758 \pm 27$  Ma ( $^{207}\text{Pb}/^{206}\text{Pb}$ ,  $1\sigma$ ) to  $2917 \pm 26$  Ma ( $^{207}\text{Pb}/^{206}\text{Pb}$ ,  $1\sigma$ ). Uranium and  
333 thorium contents and Th/U ratios (13-308 ppm, 7-262 ppm and 0.12-2.20,  
334 respectively) are similar to those from the other samples analysed. Only one sample  
335 displays discordance higher than 10 %. The remaining analyses plot on the Concordia  
336 curve forming a spread from 1800 Ma to 2900 Ma (Fig. 4). On the probability density  
337 distribution diagram, analyses show a main peak at 1800 Ma with a series of minor  
338 peaks between 2100 Ma and 2850 Ma (2100 Ma, 2400 Ma, 2550 Ma, 2700, 2850  
339 Ma), each corresponding to only a few analyses (Fig. 5).

340

341 *Sample NE148 (Margie Grit sandstone)*

342 Uranium concentrations of the 65 analyses from 63 grains largely range from 20 to  
343 280 ppm, with one analysis having a concentration reaching 600 ppm, and thorium  
344 content ranges from 4 to 262 ppm. The Th/U ratio varies from 0.24 to 2.90 which  
345 overlaps the range of expected values for magmatic zircons. No obvious variations of  
346 the U and Th content and Th/U ratio correlated to the calculated age are observed,  
347 which range from  $1004 \pm 34$  Ma ( $^{206}\text{Pb}/^{238}\text{U}$  age,  $1\sigma$ ) to  $3650 \pm 7$  Ma ( $^{207}\text{Pb}/^{206}\text{Pb}$  age,  
348  $1\sigma$ ). Plotted on a Concordia diagram (Fig. 4), the analyses plot close to the Concordia  
349 curve, displaying 2 groups (Fig. 4). Only eight analyses display discordance higher

350 than 10%. On the frequency diagram, there are two main age populations at ~1100  
351 Ma and ~2600 Ma with subordinate peaks occurring at 600 Ma and 1400 Ma (Fig. 5).

352

### 353 **Discussion**

354 The detrital zircon age spectra obtained for the Trossachs Group have a number of  
355 features in common. Overall they are characterized by age peaks at 2.8-2.6, 1.3 and  
356 1.1-1.0 Ga along with variable amplitude peaks in the range 1.7-1.9 Ga (Fig. 5). Rare  
357 Palaeoarchaeon grains are present in some samples. These gross characteristics  
358 compare closely to those of the upper Dalradian Supergroup, notably the Green Beds  
359 sample from the Southern Highland Group (Fig. 6), and with major periods of crust  
360 generation and orogenic activity within eastern Laurentia (e.g. Hoffman 1989). The  
361 age spectrum is entirely distinct from that associated with early Palaeozoic  
362 successions derived from Gondwana that occur within the Caledonian-Appalachian  
363 orogen elsewhere (Schofield & Waldron 2011 and references therein). The data is  
364 therefore consistent with a Laurentian provenance for the Trossachs Group, as  
365 indicated by the palaeontological evidence, and with the interpretation that this lies in  
366 stratigraphic continuity with the Southern Highland Group, and is therefore part of the  
367 Dalradian Supergroup (Tanner & Sutherland 2007).

368       There is no published palaeocurrent data available for the Trossachs Group.  
369 However, evidence from some of the underlying Dalradian Supergroup shows that it  
370 was supplied by sustained flow parallel to the Laurentian shoreline, along the basin  
371 axis from the southwest (Anderton 1985). In this context, the overall age range of  
372 detritus is consistent with derivation from the Laurentian foreland, especially the  
373 Labrador-Greenland region (Cawood *et al.* 2003). Archaean detritus overlaps with  
374 that of Archaean cratons, notably the Superior, and the variable proportions of

375 Palaeoproterozoic detritus corresponds to the timing of suturing of Archaean cratons  
376 by a series of orogenic belts (Ketilidian-Makkovik, New Quebec, Nagsugtoqidian,  
377 Torngat belts). Mesoproterozoic detritus is consistent with derivation from the  
378 Grenville Orogen. The presence of detrital age peaks in the late Palaeoproterozoic and  
379 early Mesoproterozoic (1.8-1.5 Ga) and the evidence for a tectonothermal event  
380 between 1.2-1.0 Ga is typical of the Labrador-southern Greenland region of eastern  
381 Laurentia (Cawood *et al.* 2003). The few grains with late Neoproterozoic ages in two  
382 samples (A265 & NE148) were probably derived from magmatism associated with  
383 rifting along the east Laurentian margin associated with opening of the Iapetus ocean  
384 (Cawood *et al.* 2001, 2003).

385         The detrital age spectrum for the sample of Salterella Grit (SG07) obtained  
386 from the Caledonian foreland in Scotland contains minor Neoproterozoic peaks between  
387 2.9 and 2.5 Ga, a significant Palaeoproterozoic peak at 1.8 Ga, as well as minor peaks  
388 at 2.4, 2.2 and 2.0 Ga (Fig. 5). The new data continues the broad upward trends  
389 evident within the Eriboll Formation, *viz* a gradual reduction in the proportion of  
390 Archaean detritus, and a corresponding increase in the amount of Palaeoproterozoic  
391 material (Cawood *et al.* 2007b). Palaeocurrent data indicate an overall southeasterly  
392 palaeoflow with some evidence for bimodal northeasterly and southeasterly flow  
393 (McKie 1990). In this context, the overall age range of detritus is consistent with  
394 derivation from the west Greenland segment of the North Atlantic craton (Cawood *et*  
395 *al.* 2007b).

396         The detrital age spectra obtained for the Trossachs Group differ from those  
397 available for the broadly coeval Cambrian strata on the Hebridean foreland in one  
398 important respect: 1.2-1.0 Ga detritus is entirely absent in the latter, which rules out a  
399 common (Labrador-Superior) provenance. None the less, the sample of sandstone



400 adjacent to the Leny Limestone (A472) is distinctive from the other Trossachs Group  
401 samples in that it contains a much smaller proportion of Mesoproterozoic (particularly  
402 1.2-1.0 Ga) detritus and a rather higher proportion of Palaeoproterozoic (2.2-1.8 Ga)  
403 detritus (Fig. 6). We suggest that the age spectrum for A472 bears comparison with  
404 that for the sample of the approximately age-equivalent Salterella Grit (SG07),  
405 implying a common provenance. The Salterella Grit was deposited during a global  
406 regressive eustatic event known as the Hawke Bay event (Palmer & James 1979).  
407 This event would presumably have resulted in the transport of significant amounts of  
408 sand offshore into the deep-water Trossachs Group basin, thus accounting for the  
409 broad similarities in provenance exhibited by these two samples. If correct, this  
410 correlation is critical to the current debate concerning the status of the Highland  
411 Border Complex as it ties the latter to the margin of Laurentia and does not support  
412 the exotic terrane model.

413         We emphasise that detrital zircon data on their own cannot provide  
414 unambiguous solutions to controversies such as that concerning the Highland Border  
415 Complex. The question of whether or not the Midland Valley terrane is underlain by  
416 (unexposed) Laurentian basement has been long debated (Bluck 2002 and references  
417 therein) and it remains a possibility that it originated as a microcontinental ribbon that  
418 rifted from the margin of eastern Laurentia during the development of the Iapetus  
419 Ocean. A similar origin has been proposed for the 'Sliswood Division' in NW  
420 Ireland (Chew *et al.* 2010). If the sedimentary rocks of the Highland Border Complex  
421 were deposited on such a microcontinental ribbon, there might be little in their  
422 provenance to distinguish them from sedimentary rocks deposited on autochthonous  
423 Laurentian basement. In that light, possible linkage of the samples of age-equivalent

424 sandstone deposited on the Caledonian foreland and in the Trossachs Group assume  
425 particular importance.

426         A relative probability plot comparing detrital zircons from the Trossachs  
427 Group (including the Keltie Water Grit Formation analysis of Cawood *et al.* 2003)  
428 with data from the Ardvreck Group of the Hebridean foreland (Cawood *et al.* 2007b,  
429 and herein), the upper Dalradian Argyll Group (Cawood *et al.* 2003), the Southern  
430 Highland Group (Cawood *et al.* 2003), the Midland Valley terrane (Phillips *et al.*  
431 2009) and the Southern Uplands terrane (Waldron *et al.* 2008) is presented in Figure  
432 7. A time-space plot highlighting the age relations between these sample sets is given  
433 in Figure 8. The samples display a series of common peaks, the relative proportions of  
434 which vary between the different groupings, which are characteristic of Laurentia  
435 (e.g. Hoffman 1989). Neoarchaean detritus is present in all groups, notably in the age  
436 range 2800-2600 Ma, corresponding with final stabilization of the cratonic nuclei of  
437 Laurentia (Hoffman 1989). Neoarchaean detritus only constitutes a minor proportion  
438 of analysed samples from the Midland Valley and Southern Uplands terranes. Late  
439 Palaeoproterozoic age zircon detritus corresponds with assembly of Laurentia and the  
440 supercontinent of Nuna along a series of collisional orogenic belts at around 2000-  
441 1800 Ma (Hoffman, 1996; Zhao *et al.* 2002). Detritus of this age constitutes a  
442 dominant component of the Ardvreck and Argyll groups. There is a marked decrease  
443 in the proportion of late Palaeoproterozoic in the Southern Highland Group and this  
444 trend continues into the Trossachs Group and detritus of this age are only a minor  
445 component of the Mildand Valley and Southern Uplands terranes (Fig. 7). The late  
446 Mesoproterozoic to earliest Neoproterozoic (1250-950 Ma) corresponds with the  
447 timing of the Grenville Orogeny and assembly of the supercontinent of Rodinia.  
448 Detritus of this age form a dominant proportion of all units except the Ardvreck

449 Group. Samples with detritus of this age also contain zircons in the range 1500-1300  
450 Ma, which corresponds with earlier accretionary margin activity along eastern  
451 Laurentia (Cawood et al. 2003). The late Neoproterozoic to early Palaeozoic Argyll,  
452 Southern Highland and Trossachs groups of the Dalradian Supergroup all contain a  
453 minor component of grains in the range 650-550 Ma, interpreted here to relate to  
454 input of rift-related magmatism associated with Rodinia breakup and opening of the  
455 Iapetus Ocean (see Cawood et al. 2001). The Midland Valley and Southern Highland  
456 Group contain early Paleozoic zircon detritus (490-460 Ma) related to magmatic arc  
457 activity generated during closure of Iapetus Ocean (Phillips et al. 2009).

458         Despite the strong points of comparison between the different data sets  
459 presented in Figure 7, there are also some notable differences that are difficult to  
460 reconcile with published models for sedimentary drainage patterns linking the terranes  
461 north of the Iapetus suture (Fig. 1) following the Grampian orogenic event. Given the  
462 Ordovician and Silurian cooling ages obtained from mineral phases in the Dalradian  
463 rocks, it is likely that they were being exhumed and eroded at this time and were thus  
464 shedding sediments into adjacent basins. Hutchison & Oliver (1998) proposed that  
465 detrital garnet within the Southern Uplands accretionary prism could be matched  
466 directly with *in situ* Dalradian garnets. It was envisaged that river systems flowed  
467 from an actively uplifting Grampian terrane, transporting detritus derived from the  
468 Dalradian Supergroup across the Midland Valley and into the accretionary prism. In  
469 this case, it would be expected that the detrital zircon record of the Midland Valley  
470 and Southern Uplands samples would match that of the Dalradian samples, in  
471 particular that of the Argyll Group, the likely source of much of the detrital garnet.  
472 All the Dalradian and Trossachs Group samples contain much higher proportions of  
473 detrital Archaean grains than the Midland Valley and Southern Uplands samples (Fig.

474 7). In addition, the Argyll Group samples are characterised by a large proportion of  
475 Palaeoproterozoic detritus, which only exists as a small proportion in the Midland  
476 Valley and Southern Uplands samples (Fig. 7). Consequently, it seems unlikely that  
477 the Ordovician-Silurian sedimentary rocks in the Midland Valley terrane were derived  
478 from erosion of the Grampian terrane (see also Phillips *et al.* 2009) and we suggest  
479 that the same conclusion can be applied to the Southern Uplands terrane. The  
480 distinctive geochemistry of the Southern Uplands terrane also argues against its  
481 derivation by erosion of the Grampian terrane (Stone *et al.* 1999). The absence of a  
482 comparable detrital zircon record in the Midland Valley and Southern Upland terranes  
483 suggests that they cannot have been in their current location relative to the Grampian  
484 terrane until at least the end Silurian.

485         These discrepancies in the detrital zircon records of now adjacent terranes  
486 support models for significant sinistral strike-slip movement on the bounding  
487 Highland Boundary and Southern Upland faults during the late Silurian to early  
488 Devonian closure of the Iapetus Ocean (Pickering *et al.* 1988; Soper *et al.* 1992;  
489 Dewey & Strachan 2003). It is difficult to be specific about the sources of the detrital  
490 zircons within the Midland Valley and Southern Uplands terranes, other than they  
491 were derived from Laurentia. Some of the Midland Valley detritus could have been  
492 derived from local basement (Phillips *et al.* 2009). Presumably an Ordovician  
493 Barrovian-style metamorphic belt extended along the Laurentian margin between  
494 west Ireland and Newfoundland, and this could have been the source of the detrital  
495 garnet and zircon within the Southern Uplands terrane if the latter was formerly  
496 located along strike further to the southwest.

497

## 498 **Conclusions**

- 499 1. Detrital zircon age spectra for the Highland Border Complex in Scotland are  
500 characterized by age peaks at 2.8-2.6, 1.3 and 1.1-1.0 Ga along with variable  
501 amplitude peaks in the range 1.7-1.9 Ga. These age peaks compare with the  
502 major periods of crust generation and orogenic activity in eastern Laurentia.  
503 The data is therefore consistent with the Laurentian provenance of the  
504 complex indicated by palaeontological evidence.
- 505 2. The detrital zircon age spectra for the Highland Border Complex compare  
506 closely to those of the upper parts of the Dalradian Supergroup of the adjacent  
507 Grampian terrane. This supports the interpretation that the two successions lie  
508 in stratigraphic continuity and questions an exotic status for the Highland  
509 Border Complex.
- 510 3. The detrital age spectrum for a sample of Cambrian Salterella Grit of the  
511 Hebridean foreland is distinguished from the Highland Border Complex by  
512 the complete absence of 1.2-1.0 Ga detritus. However, it compares with that  
513 for an approximately age-equivalent sandstone from the Leny Limestone of  
514 the Highland Border Complex. Both were contemporaneous with the  
515 regressive Hawke Bay event which may account for the broad similarities in  
516 provenance. This further ties the Highland Border Complex to the margin of  
517 Laurentia.
- 518 4. The detrital zircon age spectra for the Dalradian Supergroup and the Highland  
519 Border Complex show some similarities but also some significant differences  
520 with published data sets for Ordovician-Silurian sedimentary rocks of the  
521 adjacent Midland Valley terrane and also the Southern Upland terrane to the  
522 southeast. This is incompatible with current models that view these  
523 sedimentary rocks as derived from the erosion of the Grampian terrane. These

524 discrepancies in the detrital zircon records of now adjacent terranes support  
525 models for significant sinistral strike-slip movement on the bounding  
526 Highland Boundary and Southern Upland faults during the late Silurian to  
527 early Devonian closure of the Iapetus Ocean.

528

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534

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774 **Figure Captions**

775 Fig. 1. Lithotectonic divisions of the Caledonian Orogen in Scotland and northern

776 England. Box outlines area of Figure 2. Salterella Grit sample, SG07 occurs

777 on shores of Loch Eriboll in the Hebridean foreland, close to the Moine thrust.

778 Fig. 2. Geologic map in the vicinity of the Highland Border Complex, Scotland,

779 showing location of analysed samples, A265, A266, A472 and NE148.

780 Fig. 3. Stratigraphic columns for the Cambro-Ordovician succession in the Hebridean

781 foreland and for the Trossachs Group and Highland Border Ophiolite along

782 the Highland Boundary Fault. Analysed samples shown by numbered

783 triangles. Unnumbered triangles refer to stratigraphic position of samples from

784 the Hebridean foreland analysed by Cawood *et al.* (2007b). Stars show fossil

785 locations in the Trossachs Group and Highland Border Ophiolite.

786 Fig. 4. Concordia diagrams based on  $^{204}\text{Pb}$ -corrected data for analysed samples. Error

787 ellipses are shown at 2 sigma level;

788 Fig. 5. Probability density distribution diagrams of Concordia ages for analyzed

789 samples, which were constructed using the method of Nemchin and Cawood

790 (2005). Grey shaded bars delineate the age ranges 2800-2600 Ma, 2000-1750

791 Ma and 1250-950 Ma. *n*—number of analyses; *s*—number of samples.

792 Fig. 6. Probability density distribution diagram of siliciclastic sequences from the the

793 Caledonian Orogen in Scotland northern England. Sources of data: Ardvreck

794 Group, Hebridean foreland – Cawood *et al.* 2007b and this paper (SG07);

795 Argyll Group – Cawood *et al.* (2003); Southern Highlands Group – Cawood *et*

796 *al.* (2003); Trossachs Group – this paper (samples A265, A472, NE148) and

797 Cawood *et al.* (2003); Highland Border Ophiolite – this paper; Midland Valley

798 terrane – Phillips *et al.* (2009); Southern Uplands terrane – Waldron *et al.*  
799 (2008). Abbreviations: *n*—number of analyses; *s*—number of samples.

800 Figure 7. Late Neoproterozoic to early Paleozoic lithostratigraphic sequences in the  
801 Hebridean foreland and Grampian, Midland Valley and Southern Uplands  
802 terranes showing position of analysed samples plotted on figure 6. Tie scale  
803 from Gradstein *et al.* (2004). Abbreviations: Neoprot. – Neoproterozoic; E –  
804 Early; M – Middle; L – Late; T – Tremadoc; A – Arenig.

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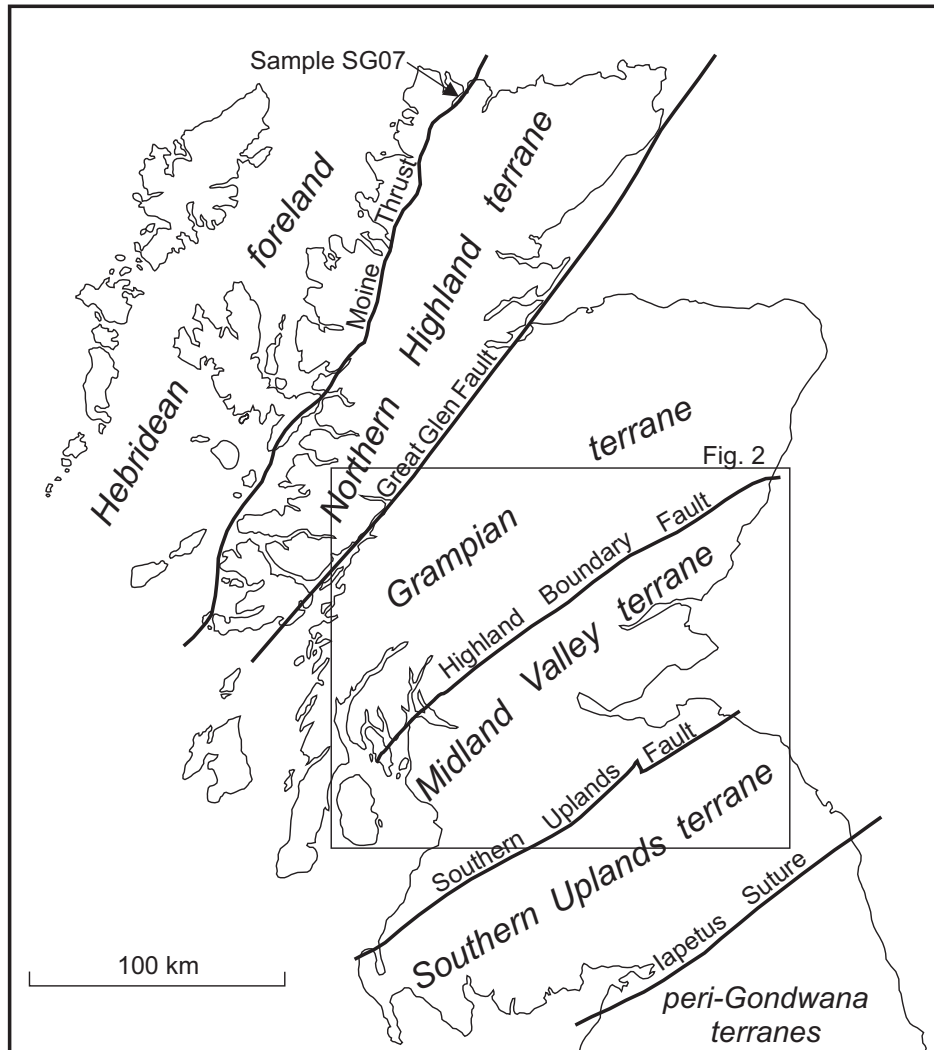
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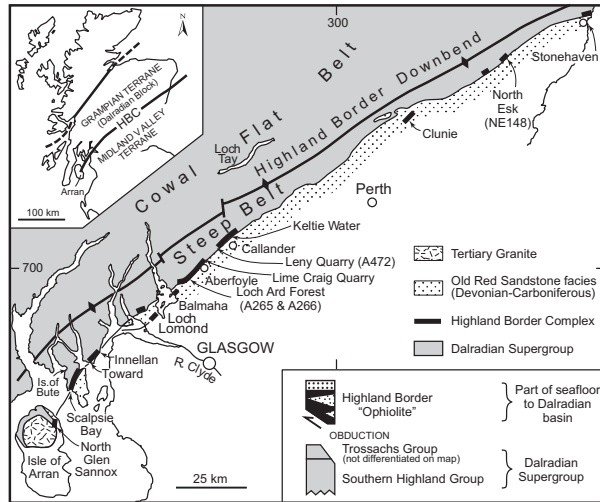
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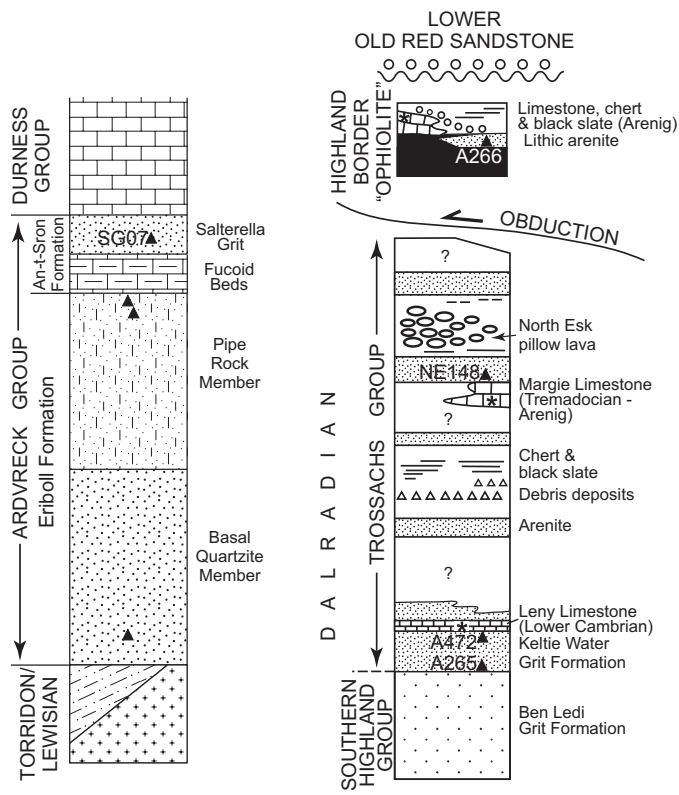
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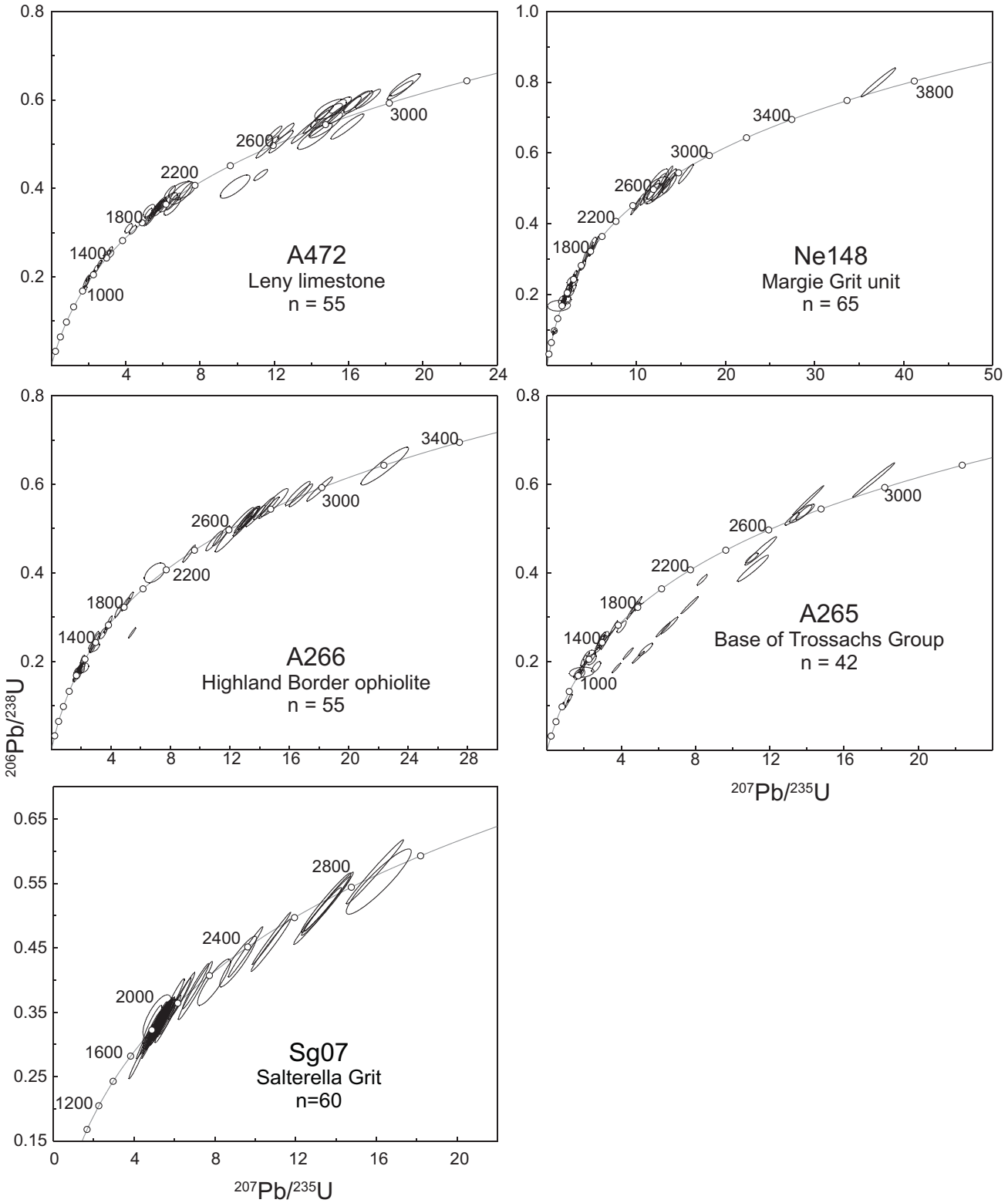
Cawood et al. Fig. 2



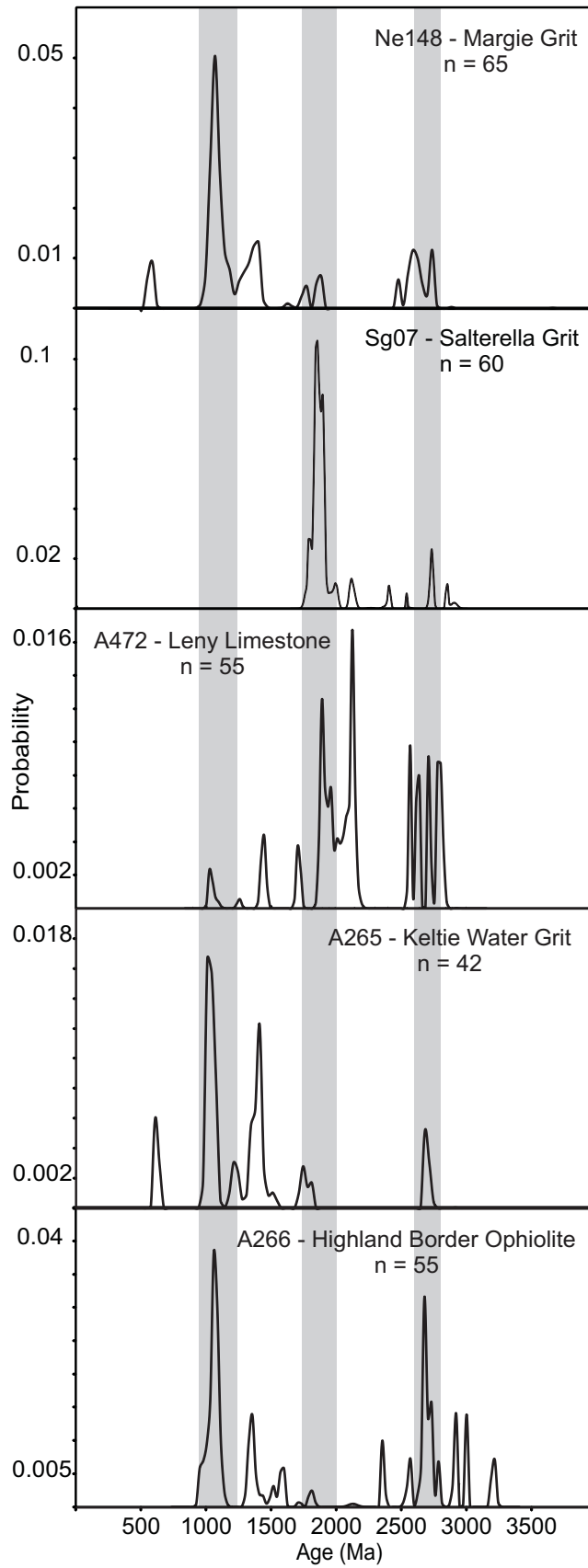
Cawood et al Fig. 3



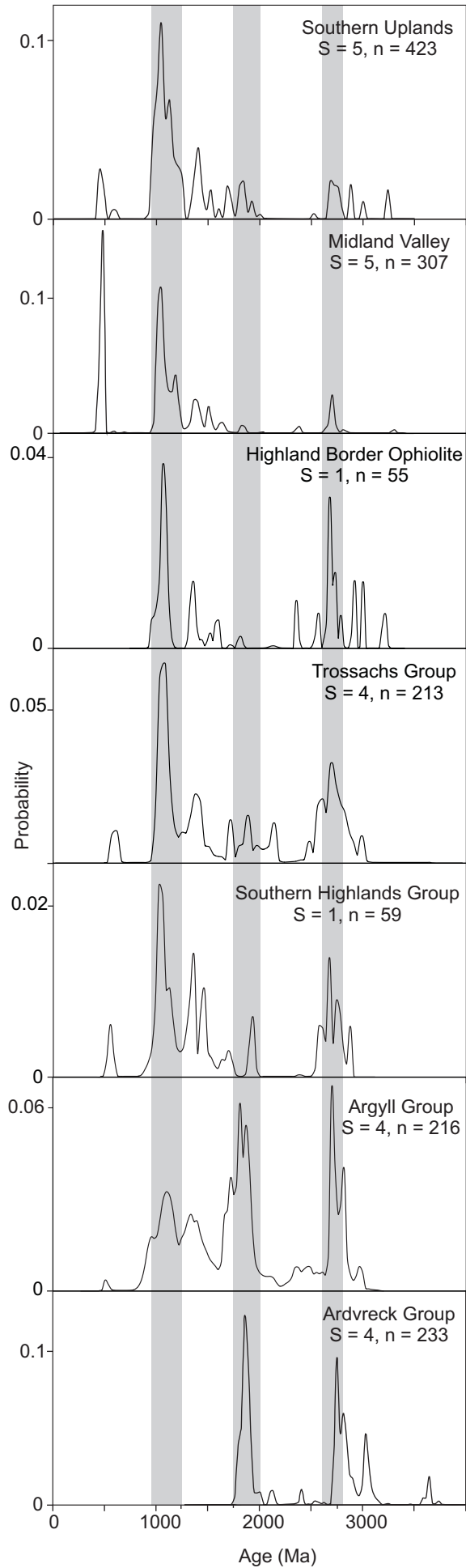
Cawood et al Fig. 4



Cawood et al Fig. 5



Cawood et al Fig. 6





Cawood et al. Fig. 7

