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Evidence, or not, for the late Tonian break-up of Rodinia? The Dalradian Supergroup, Scotland

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Abstract: The Tonian–Cambrian Dalradian Supergroup in Scotland is a siliciclastic–carbonate succession that can be up to 10 km thick. The consensus view is that its lower part, the mid- to late Tonian Grampian and Appin groups, formed in rift basins: the deep marine turbidites of the Grampian Group infilled rift depocentres, whereas the shallow marine strata of the Appin Group mark basin-bounding palaeohighs. This scenario is used as a key line of evidence to infer the onset of the break-up of Rodinia between Laurentia and Baltica. However, deformation during the mid-Ordovician Caledonian Orogeny obscured the original depositional frameworks. Reconstructing these frameworks (and hypothesized rift basins) has relied on the trace and major element log-ratio geochemistry of minor carbonate rocks to assign the units to either the Grampian or Appin group – that is, to rift depocentres or basin-bounding palaeohighs, respectively. We report new carbon and oxygen isotope and geochemical data and use these to create a revised stratigraphic framework for the Grampian and Appin groups. Our findings show that the previous geochemical-based correlations are unreliable and that there is no evidence for palaeohighs or rift basins. Instead, the Grampian–Appin groups are a deeper marine flysch to a shallower marine molasse succession formed in response to the mid-Tonian Knoydartian Orogeny. From a Scottish perspective, evidence for the break-up of Rodinia is recorded higher in the Dalradian succession during the deposition of the early Cryogenian Argyll Group.

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The Tonian–Cambrian Dalradian Supergroup in Scotland and Ireland formed during a pivotal period in Earth history. It spans the Cryogenian climatic extremes of the Sturtian and Marinoan Snowball Earth (Brasier and Shields 2000; McCay *et al.* 2006; Prave *et al.* 2009*a*, *b*; Ali *et al.* 2018; Fairchild *et al.* 2018), the break-up of the Rodinia supercontinent (Dalziel 1997; Cawood *et al.* 2007, 2010, 2016) and the subsequent late Ediacaran opening of the Iapetus Ocean (e.g. Robert *et al.* 2021). As such, it figures prominently in scenarios aimed at reconstructing the Neoproterozoic geological evolution of the North Atlantic realm. Here, we assess a concept that is one of the most enduring about the Dalradian Supergroup – namely, that it was initiated in response to a phase of episodic, but prolonged, extensional tectonism that persisted throughout its entire depositional history (Soper and Anderton 1984; Anderton 1985; Harris *et al.* 1994; Glover *et al.* 1995; Dalziel and Soper 2001; Stephenson *et al.* 2013). This concept is central to many ideas regarding the tectonic fragmentation of Rodinia and the ultimate formation of the Iapetus Ocean. But, is this concept correct?

We focus on the geology of the northern Grampian Highlands of Scotland (Figs 1 and 2a), where the idea was first framed that Dalradian sedimentation began in rift basins bounded by long-lived palaeohighs (Fig. 2b; Glover *et al.* 1995; Smith *et al.* 1999). This tectonic scenario hinges on lithostratigraphic correlations contingent on the trace element and major element (Fe–Mg–Ca) log-ratio compositions of carbonate rocks as a correlation tool to help identify stratigraphic position within the Dalradian succession (Rock 1985; Thomas 1989; Thomas *et al.* 2004; Thomas and Aitchison 2006). Here, we report new carbon ($\delta^{13}C_{carb}$) and oxygen ($\delta^{18}O_{carb}$) isotope and geochemical data from the same carbonate rocks. Our results question the correctness of some of the geochemical-based correlations and the plate tectonic inferences drawn from them. Accordingly, we propose a revised stratigraphic framework compatible with our new and previously published carbon–oxygen isotope and compositional data. This framework enables us to construct a geological scenario more in line with our findings and the insights obtained from across the North Atlantic realm since the rift-model concept was first advanced more than three decades ago.

Dalradian Supergroup

The Dalradian Supergroup is exposed in the Grampian Highlands of Scotland, several inner Hebridean islands and Shetland, in addition to various inliers in western and northern Ireland. Our focus is on the exposures that occur across mainland Scotland (Fig. 1). The Dalradian Supergroup is divided into five main units, from oldest to youngest, the Glenshirra, Grampian, Appin, Argyll and Southern Highland groups; our interest is in the Grampian and Appin groups. On mainland Scotland, the Dalradian rocks are bounded by the Great Glen and Highland Boundary faults and were variably metamorphosed and deformed at greenschist to amphibolite facies during the mid-Ordovician Grampian phase of the Caledonian Orogeny.

Few units capable of constraining depositional ages have been found in the Dalradian succession. The only unequivocal depositional age is a U–Pb zircon date of *c.* 601 Ma from a tuff near the

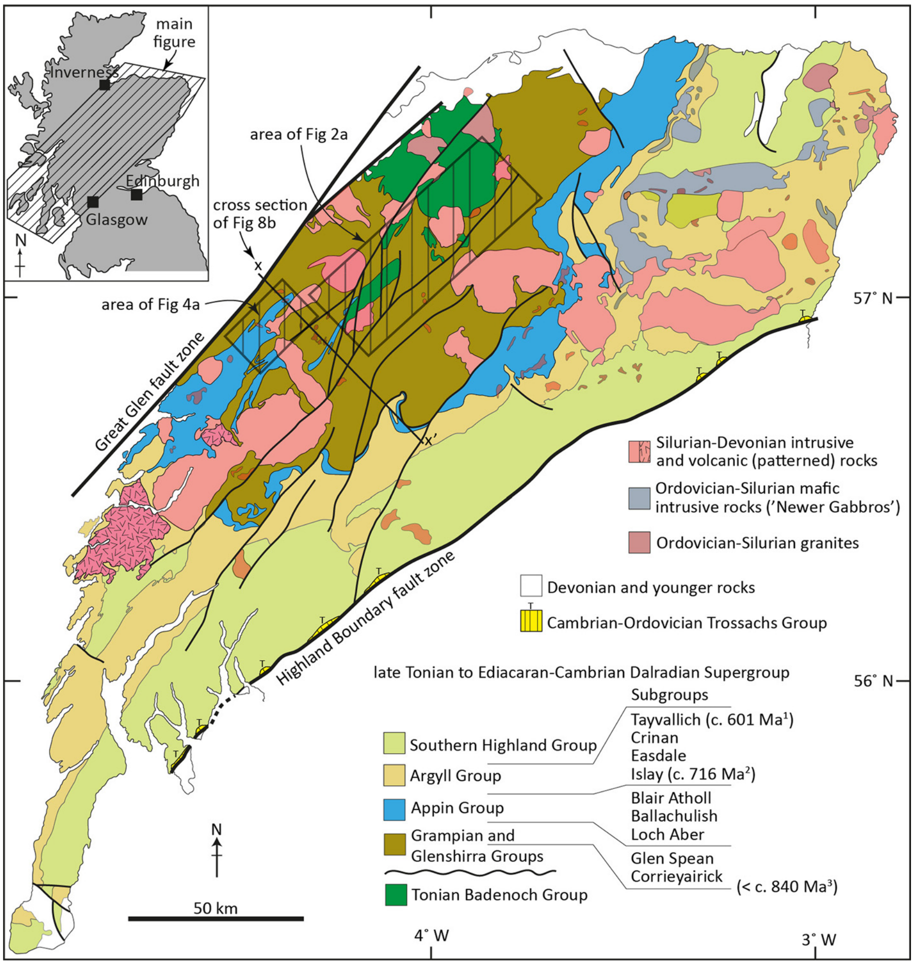

Fig. 1. Simplified map and stratigraphic nomenclature of the Dalradian Supergroup in mainland Scotland. Ages from (1) Dempster *et al.* (2002), (2) Fairchild *et al.* (2018) and (3) this study. Source: simplified after British Geological Survey (2007).

base of the Southern Highland Group (Dempster *et al.* 2002); consequently, the timing of initiation of Dalradian deposition is uncertain. The inferred oldest unit, the Glenshirra Group, occurs in several small (tens of square kilometres), structurally isolated exposures; it has no known base and mylonitized shear zones separate it from all other Dalradian units. Objective assessment of the geological data leads to the conclusion that the stratigraphic position of the Glenshirra Group is currently unknown.

For the Grampian and Appin groups, two other age constraints are key. The first is that the Grampian Group must be younger than the *c.* 840 Ma amphibolite facies metamorphism that affected the gneissose rocks of the unconformably underlying Badenoch Group (Piasecki 1980; Piasecki and Temperley 1988*a*, *b*; Highton *et al.* 1999; Hyslop and Piasecki 1999; Phillips *et al.* 1999). Some workers have suggested that Dalradian deposition began after *c.* 806 Ma, the U–Pb monazite age (Noble *et al.* 1996) for granitic pegmatites associated with the mylonites that cut the Badenoch Group rocks (see discussion in Stephenson *et al.* 2013). However, no direct cross-cutting age relationship between the mylonites and Grampian Group rocks has been confirmed. Hence the initial deposits of the Grampian Group could either pre- or post-date the timing of mylonitization. The second constraint is linked to the glaciogenic Port Askaig Formation (Spencer 1971). The base of this formation marks the contact between the Appin Group and the overlying Argyll Group and the formation is interpreted to be correlative with the *c.* 716–660 Ma Sturtian glaciation (Brasier and Shields 2000; McCay *et al.* 2006; Prave *et al.* 2009*a*; Ali *et al.* 2018; Fairchild *et al.* 2018). These constraints place the deposition of the Grampian and Appin groups as occurring at some time between *c.* 840 and 716 Ma.

Grampian and Appin groups

The Grampian Group is divided into the older Corrieyairick and younger Glen Spean subgroups. The former is a monotonous succession, 5 km or more in thickness, of centimetre to decimetre thick graded beds of micaceous psammite, semipelite, pelite and minor quartzite. The proportions of these lithologies vary from area to area (Fig. 3) and, in places near the base of the Corrieyairick Subgroup, there are thin, typically 1–10 m thick, laterally discontinuous (in part, likely due to structural boudinage) carbonate rock bodies. The entirety of the Corrieyairick Subgroup is interpreted as a deep marine turbidite fan system (Glover and Winchester 1989; Glover 1993; Robertson and Smith 1999; Smith *et al.* 1999). The overlying Glen Spean Subgroup is a several kilometre thick succession of interbedded psammite and semipelite marked by a diverse suite of sedimentary features, including planar and trough cross-bedding, current and combined-flow ripples, flaser bedding and herringbone cross-lamination. It has been interpreted as a tide- and storm-influenced marine shelf and shoreline system (Glover 1993; Glover *et al.* 1995; Banks 2007).

The Appin Group is 3–5 km thick and is divided into, from the base upwards, the Lochaber, Ballachulish and Blair Atholl subgroups. These subgroups are, respectively, a feldspathic and quartzitic psammite–pelite unit, a unit of interbedded psammite and pelitic–graphitic schist with minor carbonate rocks, and a unit marked by limestone–dolomite bodies many hundreds of metres thick. All are interpreted as having formed on a shallow marine shelf (Hickman 1975; Litherland 1980; Anderton 1985), with the additional aspect that the Blair Atholl Subgroup exhibits

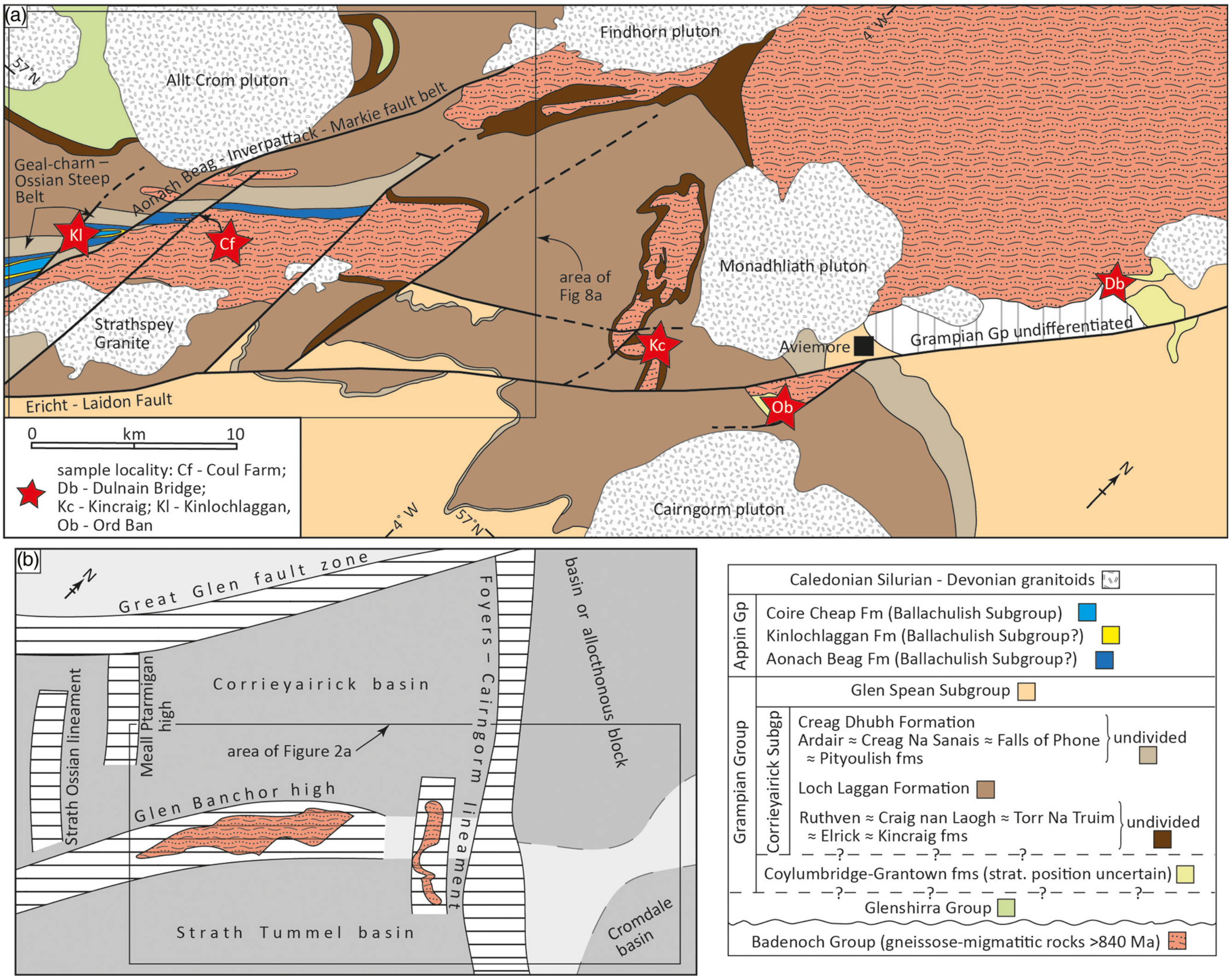

Fig. 2. (**a**) Generalized map of the northern Grampian Highlands. (**b**) Postulated palaeogeography of the Grampian Group basins. Source: map simplified after British Geological Survey (2007); postulated palaeogeography after Smith *et al.* (1999).

northeastward facies changes into carbonate rhythmite and mudstone deposits. In areas where the sedimentology of both groups has been studied (Glover and Winchester 1989; Glover 1993; Glover *et al.* 1995), their composite stratigraphic framework defines a progradational succession of deeper marine turbidites upwards into shallow marine and coastal deposits (Fig. 4a, b).

Northern Grampian Highlands: the case against rift basins

The most widely held view of the Dalradian Supergroup is that it was deposited at the start of a prolonged phase of extensional tectonism associated with the break-up of Rodinia and the formation of the Iapetus Ocean (Soper and Anderton 1984; Anderton 1985;

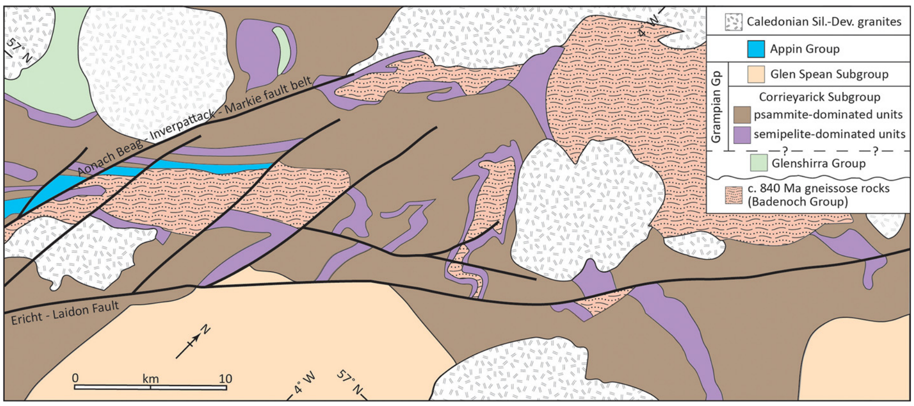

Fig. 3. Rock distribution map showing the occurrence of psammite- and semipelite/pelite-dominated units in the northern Grampian Highlands (same map as in Fig. 2a). Note that the finer grained facies (pelites/semipelites) commonly occur above the contact with the Badenoch Group rocks; see text for discussion. Source: after Robertson and Smith (1999).

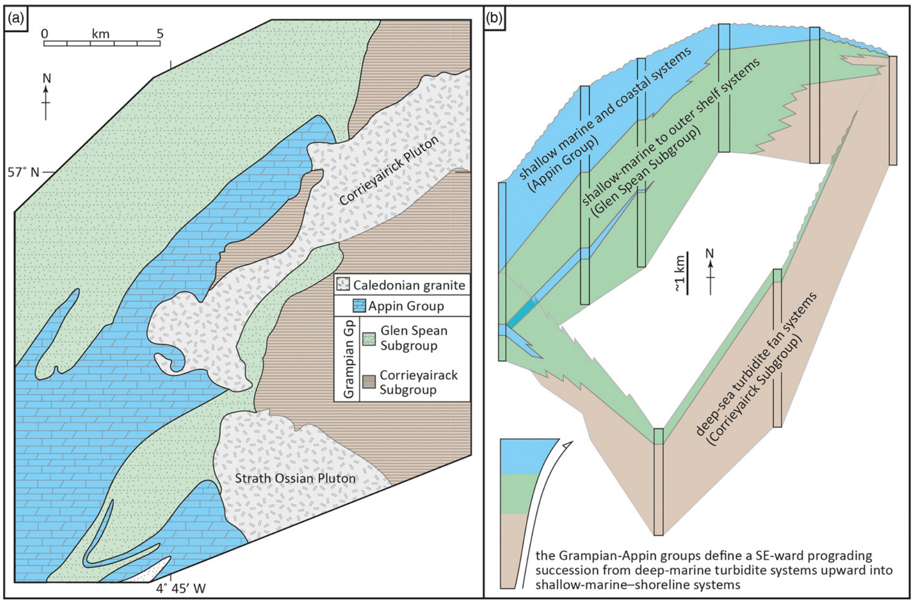

Fig. 4. (**a**) Simplified geological map of part of the north-central Grampian Highlands (see Fig. 1 for location). (**b**) Fence diagram showing the overall progradational shallowing-upwards depositional trend of the Grampian–Appin groups for the north-central Grampian Highlands. Source: after Glover *et al.* (1995).

Harris *et al.* 1994; Dalziel and Soper 2001; Cawood *et al.* 2007; Stephenson *et al.* 2013). This view relies on the interpretation that Dalradian sedimentation began in rift basins bounded by postulated palaeohighs comprised of the gneissose rocks of the Badenoch Group, the most notable examples being the Corrieyairick and Strath Tummel sub-basins and Glen Banchor High (Fig. 2b; Glover and Winchester 1989; Glover *et al.* 1995; Robertson and Smith 1999; Smith *et al.* 1999; Banks and Winchester 2004; Banks *et al.* 2007; Leslie *et al.* 2013; Stephenson *et al.* 2013). Establishing the existence of these features relied on using the trace element and Fe–Mg–Ca log-ratio compositions of carbonate rocks as a correlation tool (Thomas 1989; Thomas and Aitchison 2006). This led to the conclusion that carbonate rocks adjacent to the Glen Banchor High belonged in the Appin Group. This conclusion was then used as evidence to infer that the Badenoch Group rocks represented long-lived, high-standing basement blocks (i.e. horsts) that were onlapped and eventually buried during the deposition of the Ballachulish Subgroup of the Appin Group (Smith *et al.* 1999; Leslie *et al.* 2013).

However, many features of the Grampian and Appin groups are difficult to reconcile with the stratigraphic architectures and sedimentological features that identify rift basins formed in continental crust (e.g. Wernicke 1990; Purser and Bosence 1998; Ravnås *et al.* 2000; Withjack *et al.* 2013). The infill of such basins is typified by strata that exhibit rapid lateral and vertical facies changes. The associated sediment is mostly locally derived, commonly poorly sorted and coarse grained, particularly in areas proximal to basin-bounding highs/horsts. The Grampian and Appin groups have none of these characteristics. Provenance studies show that the sediments were sourced mostly from far-distant Mesoproterozoic terranes on the Laurentian craton (Cawood *et al.* 2003). Further, the Grampian Group rocks in the Corrieyairick and Strath Tummel sub-basins are fine-grained turbidites that show no indication of sedimentation influenced by intra-basinal highs. In fact, the inferred palaeohighs are in many instances encased by formations comprised of finer grained semipelite/pelite facies rather than coarser grained psammite (Fig. 3).

If we include the Glenshirra Group in the rift basin models, then the contrast between it and sediments related to continental rifts is even more striking. The Glenshirra rocks are the coarsest facies in the region, but, rather than being locally derived, poorly sorted sedimentary breccias and conglomerates, they are mostly quartzo-feldspathic psammites with dispersed, rounded granitoid clasts. The clasts were derived from the *c.* 1.7 Ga meta-igneous Rhinns Complex, of which the nearest present day exposures are 200 km to the west on Islay (Banks *et al.* 2007, 2013).

In summary, the Glenshirra–Grampian–Appin groups are devoid of the typical lithological features that characterize the infill of continental rift basins. By contrast, starting with the early Cryogenian Argyll Group, it and the overlying units of the Dalradian Supergroup contain much evidence for deposition during extensional tectonism. This evidence includes coarse clastic facies exhibiting significant variations in lateral and vertical facies and thickness (Anderton 1985), synsedimentary normal faults (Soper and Anderton 1984; Levell *et al.* 2020) and increasingly widespread mafic and felsic igneous activity (Fettes *et al.* 2011; Strachan *et al.* 2013). These aspects warrant reconsideration of the interpretation that the lower part of the Dalradian Supergroup (the Glenshirra–Grampian–Appin groups) formed in discrete rift basins separated by basin-bounding palaeohighs. They also highlight the importance of robustly assessing the use of geochemical data derived from carbonate rocks before using them to formulate tectonic and sedimentary basin models.

Carbonate rock units in the Grampian and Appin groups

Although carbonate rocks are only minor components of the dominantly siliciclastic successions, their correlation is crucial in assigning stratigraphic affinity and postulating the existence of rift basins and palaeohighs. As noted earlier, the key criterion in making such inferences has been the trace element and major element log-ratio compositions of these rocks. An alternative methodology that has proved successful in constructing intra-basinal correlations in Neoproterozoic successions worldwide has been combining stratigraphic and structural mapping with detailed sedimentology and carbon isotope chemostratigraphy. The Dalradian Supergroup is one such example in which this methodology has been used with great success (Brasier and Shields 2000; Condon and Prave 2000; McCay *et al.* 2006; Prave *et al.* 2009*a*, *b*; Ali *et al.* 2018; Fairchild *et al.* 2018). This past success gave us confidence in applying this approach to test the correlations reliant on element log-ratio data. It also provides an independent means of assessing the proposed linkages of carbonate rocks to the Grampian and Appin groups in the three key areas central to establishing the rift basin model (Fig. 2a) – Coul Farm, Kincraig and Kinlochlaggan – and two additional related areas, Dulnain Bridge and Ord Ban.

In the present stratigraphic framework of the Dalradian Supergroup, the carbonate rocks at Kinlochlaggan and Coul Farm are in the Appin Group (Ballachulish Subgroup), whereas those at Kincraig are in the lower part (Corrieyairick Subgroup) of the Grampian Group (Smith *et al.* 1999; Thomas and Aitchison 2006; see discussion in Leslie *et al.* 2013). The stratigraphic position of the Dulnain Bridge and Ord Ban carbonate rocks (the Grantown and Coylumbridge formations, respectively) is uncertain. However, the former is thought to occur within the lower part of the Grampian Group, whereas the latter has been variously assigned to either the Grampian or Appin groups (Thomas and Aitchison 2006; see discussions in Stephenson *et al.* 2013 and Leslie *et al.* 2013).

As we discuss later, we interpret both the Dulnain Bridge and Ord Ban carbonate-rock-bearing localities to be part of the Grampian Group. To re-emphasize what was highlighted previously, the carbonate rocks at all these localities occur near to gneissose basement rocks (the Badenoch Group). Accordingly, those at Kincraig, which occur in the lower part of the Dalradian succession (the Grampian Group), were denoted as marking a rift basin depocentre. By contrast, the inferred much higher stratigraphic position of the carbonate rocks at Coul Farm and Kinlochlaggan (Appin Group) was interpreted as outlining a basin-bounding palaeohigh.

Carbonate rock units: new samples and analyses

The carbonate rocks in the five areas noted in the previous section of this paper are dominantly calcite marbles. They range in thickness from *c.* 1 to 10 m and are encased in psammitic to pelitic units many hundreds of metres thick. The marbles were sampled at *c.* 1 m spacings with 21 new samples being collected. The carbon and oxygen isotope compositions of these samples were measured following standard protocols at the mass spectrometry facility in the Department of Geology, University of Tartu, Estonia. These data were combined with previously reported carbon and oxygen isotope data (Prave *et al.* 2009*a*) for rocks of the Grampian and Appin groups obtained at the Scottish Universities Environmental Research Centre, East Kilbride, Scotland, also following standard protocols. Stable isotope data are reported as $\delta^{13}C_{carb}$‰ and $\delta^{18}O_{carb}$‰ relative to the V-PDB standard and the analytical uncertainty for both laboratories was better than ±0.1‰. In total, we report 96 carbonate carbon and oxygen isotope analyses for Grampian and Appin group strata (Table 1). We also obtained major and trace element compositions following a HNO_3–$HClO_4$–HF digestion protocol for the 21 new samples (reported in the Supplementary material) using the inductively couple plasma mass spectrometry facility in the Department of Geology, University of Tartu.

Carbonate rock units: analytical results integrated with field-based observations

Three key findings can be drawn from our carbon isotope data (Fig. 5a–c): (1) there is clear differentiation between the calcite marbles at Kinlochlaggan and those at Coul Farm and Kincraig (the three areas that form the keystone of the rift basin model); (2) the Kinlochlaggan data overlap considerably with those for the Appin Group limestones, whereas the Coul Farm–Kincraig data form an isotopically distinct cluster; and (3) the Ord Ban and Dulnain Bridge data form a coherent cluster different from that of the Coul Farm–Kincraig data. Given these findings, we use the carbon isotopic composition of each of the limestones to postulate a chemostratigraphically consistent trend (Fig. 5c) in which the Kinlochlaggan (Coire Cheap Formation) data occupy a carbon isotope space intermediate between the Ballachulish and Appin limestones and those of the Ord Ban and Dulnain Bridge marbles (the Coylumbridge and Grantown formations, respectively) are intermediate between the Grampian Group and the lowermost Appin Group.

The carbon isotope data for the calcite marbles at Kinlochlaggan are compatible with these rocks belonging in the Appin Group, whereas those at Coul Farm are not. However, the Coul Farm data match well with the Kincraig data (i.e. the lower Grampian Group). Thus, the carbon isotope data allow us to consider the marble units at Coul Farm and Kincraig as correlative. Examination of the siliciclastic rocks at these two localities, as well as in the intervening areas, shows that there are only minor variations in the thickness and proportions of psammitic, quartzitic and pelitic units between Coul Farm and Kincraig. Such facies changes are commonplace in both ancient and modern turbidite fan systems. Hence both the siliciclastic rocks and the marbles at the two localities can be interpreted as lateral equivalents.

It is also noteworthy that, although the rocks at Coul Farm were placed in the Appin Group based on Fe–Mg–Ca log-ratios (Thomas and Aitchison 2006; see discussion in Leslie *et al.* 2013), the Coul Farm locality lacks the well-defined quartzite–limestone and limestone–graphitic schist couplets that are such characteristic features of the Appin Group elsewhere across the Dalradian outcrop belt. Thus, combining these findings and observations and, given that all researchers agree that the marbles at Kincraig belong in the lower Grampian Group, the most parsimonious conclusion to reach is that the marbles and associated strata at Coul Farm are also part of that group. As explained in the following, removing the rocks at Coul Farm from the Appin Group and placing them in the Grampian Group has far-reaching implications.

Given the previous reliance on the trace element and Fe–Mg–Ca log-ratio compositions of the carbonate rocks as evidence to differentiate units and construct correlations from area to area across the northern Grampian Highlands (Rock 1985; Thomas 1989; Thomas and Aitchison 2006; see discussion in Leslie *et al.* 2013), we tested these correlations with our compositional data (Fig. 6). Our results raise severe doubts about using such a method to assign stratigraphic affinity. Rare earth element spider diagrams and immobile element data, such as Sc–Th–Zr plots (Fig. 6a), are unable to differentiate between the units assigned to the Grampian and Appin groups by other workers (Thomas and Aitchison 2006 found likewise). Fe–Mg–Ca log-ratio plots also generate ambiguous results. Our data (Fig. 6b, c) show the purported Appin Group marbles (Coul Farm) clustering with Grampian Group marbles (Kincraig).

The marbles at Ord Ban and Dulnain Bridge are another interesting case study. Thomas and Aitchison (2006) concluded that the marbles at Ord Ban and Kincraig are correlative and therefore part of the Grampian Group, whereas the marbles at Dulnain Bridge (the Grantown Formation in Figs 5 and 6 and Table 2) belong in the Appin Group. By contrast, our carbon isotope data show the Ord Ban marbles to be distinct from the marbles at Kincraig, but similar to the marbles at Dulnain Bridge (Fig. 5c). Elemental ratios that can be used to assess the provenance of terrigenous material in carbonate rocks, such as Th/Sc and Ti/Al (Fig. 6a), also hint at the plausibility of a Coul Farm–Kincraig linkage, as well as an Ord Ban–Dulnain Bridge linkage. Further, the siliciclastic rocks associated with the marbles at Ord Ban and Dulnain Bridge are similar-appearing psammitic–pelitic schist and paragneiss, hence are permissive of correlating these localities lithostratigraphically.

In summary, the trace element and Fe–Mg–Ca log-ratio data can be used to differentiate the correlations between Grampian and Appin group carbonate rocks. What can be stated with certainty is that the marbles at Coul Farm and Kincraig, and those at Ord Ban and Dulnain Bridge, share four traits: (1) each grouping has internal carbon isotope compatibility; (2) each grouping displays siliciclastic and carbonate lithofacies similarity; (3) the encasing psammite–pelite units are comparable in all four areas; and (4) all four areas lack the well-developed quartzite–carbonate and graphitic schist–

Table 1. *Carbonate carbon and oxygen isotopic data*

Grampian Group: Corrieyairick Subgroup			Appin Group: Ballachulish Limestone			Appin Group: Appin Limestone		
Locality	$\delta^{13}C_{carb}$	$\delta^{18}O_{carb}$	Locality	$\delta^{13}C_{carb}$	$\delta^{18}O_{carb}$	Locality	$\delta^{13}C_{carb}$	$\delta^{18}O_{carb}$
Coul Farm*			Spean River			Onich		
CF-1	7.36	−14.40	BLH-1	−0.57	−11.57	APP-1	2.10	−14.88
CF-2	3.33	−14.03	BLH-2	−0.47	−11.80	APP-2	3.96	−14.43
CF-3	5.36	−13.19	BLH-3	−0.82	−11.67	APP-4	3.75	−10.97
CF-4	5.86	−12.58	BLH-4	−0.38	−12.90	APP-5	3.72	−12.10
CF-5	6.90	−12.90	BLH-5	−2.58	−12.34	APP-6	1.90	−13.43
CF-6	8.04	−13.51	BLH-6	−4.03	−12.74	APP-7	3.67	−11.82
CF-7	8.20	−12.71	BLH-7	−5.28	−13.44	APP-8	4.20	−10.22
Dulnain Bridge*			BLH-8	−5.22	−13.13	APP-9	4.13	−10.49
DB-1	1.44	−12.31	BLH-9	−7.04	−12.82	APP-10	3.76	−11.80
DB-2	0.50	−17.28	Port Appin			APP-11	3.60	−11.93
DB-3	−1.61	−16.95	BC-0	−3.89	−10.23	APP-12	3.82	−12.56
DB-4	0.53	−17.46	BC-2	−4.59	−10.76	APP-13	4.07	−13.34
Kincraig West*			BC-3	−4.43	−9.55	APP-14	3.91	−14.03
KC-1	9.00	−10.29	BC-4	−4.11	−10.17	APP-15	3.36	−14.89
KC-2	8.89	−11.73	BC-5	−4.17	−10.25	APP-16	2.51	−16.00
KC-3	2.66	−18.01	Benderloch			APP-17	2.72	−14.60
KC-4	5.63	−16.28	BD-19	1.12	−14.15	APP-18	2.64	−14.75
KC-5	3.76	−18.55	BD-18	1.17	−14.43	APP-19	1.71	−16.91
KC-6	7.83	−11.20	BD-17	0.88	−14.43	APP-20	2.71	−13.90
Kincraig East			BD-16	1.15	−14.32	APP-21	2.60	−15.51
KC-1a	8.24	−10.14	BD-15	1.01	−14.06	Sandend Bay West		
KC-2a	9.27	−10.1	BD-14	1.09	−13.83	FD-A	3.89	−9.26
KC-3a	4.03	−14.89	BD-13	0.07	−14.10	FD-B	4.98	−10.95
KC-4a	7.65	−12.19	BD-12	1.06	−13.99	FD-C	5.44	−10.44
KC-5a	7.73	−14.37	BD-11	0.80	−12.76	FD-D	4.84	−8.79
KC-6a	3.93	−18.51	BD-10	1.47	−13.18	FD-1	4.37	−8.96
KC-7a	8.84	−11.54	BD-9	2.06	−12.98	FD-2	4.45	−10.55
Ord Ban*			BD-8	1.46	−12.95	FD-3	3.76	−9.42
OB-1	0.57	−16.19	BD-7	1.68	−13.20			
OB-2	0.77	−17.23	BD-6	−2.23	−11.81			
OB-3	2.81	−10.90	BD-5	0.75	−14.15			
OB-4	2.16	−11.62	BD-4	−1.41	−10.29			
Kinlochlaggan*			BD-3	−1.67	−17.53			
KLL-1	0.06	−13.88	BD-2	−1.21	−18.08			
KLL-2	2.41	−11.89	BD-1	−0.94	−17.59			
KLL-3	2.12	−14.19						
KLL-4	2.03	−13.49						
KLL-5	1.74	−11.97						
KLL-6	1.24	−12.93						
KLL-7	3.38	−8.63						
KLL-8	2.51	−8.91						

*Data analysed at the University of Tartu, Estonia; all other data analysed at the Scottish Universities Environmental Research Centre, East Kilbride, UK.
Sample locations (positions given as latitude–longitude): Benderloch 56.48610−5.45150; Coul Farm 57.02704−4.32235; Dulnain Bridge 57.31308−3.66339; Kincraig East 57.13316−3.94946; Kincraig West 57.13266−3.94971; Kinlochlaggan 56.97610−4.39337; Onich 56.70118−5.22178; Ord Ban 57.15524−3.82829; Port Appin 56.56315−5.38524; Sandend Bay West 57.68565−2.74642; Spean River 56.89142−4.88391.

carbonate couplets that characterize Appin Group strata in the nearby Geal Charn–Ossian Steep Belt (Kinlochlaggan) and everywhere else along the Dalradian outcrop belt (Fig. 2a).

Taken in total, these lines of evidence infer that the marbles at all four localities are part of the lower Grampian Group. Crucially, placing the Coul Farm rocks into the Grampian Group eliminates the stratigraphic evidence for the Glen Banchor High. This finding, along with the data and observations highlighted earlier, also eliminates the stratigraphic basis for rift basins. Such a dramatic change in tectonic scenario emphasizes rather sharply the requirement to integrate as thoroughly as possible geochemical data based on carbonate rocks with sedimentological and stratigraphic observations before relying on the former as a correlation tool or as a criterion for assigning stratigraphic affinity.

An alternative basin scenario: the Grampian–Appin groups as a Knoydartian flysch–molasse

More than two decades ago, Prave (1999) speculated that the combined Grampian Group and Appin Group stratigraphic framework exhibited facies characteristics comparable with the flysch–molasse successions of foreland basins (e.g. the Taconic–Acadian–Alleghenian clastic wedges of the Appalachian Basin; Allen and Homewood 2009; Busby and Azor 2012; Miall and Blakey 2019). In this scenario, the deeper marine turbiditic Corrieyairick Subgroup was defined as flysch and the shallow marine Glen Spean–Lochaber subgroups constituted molasse. The Ballachulish and Blair Atholl subgroups were attributed to an overstepping successor basin marked by a transition from mixed siliciclastic–carbonate facies into carbonate-dominated strata. The

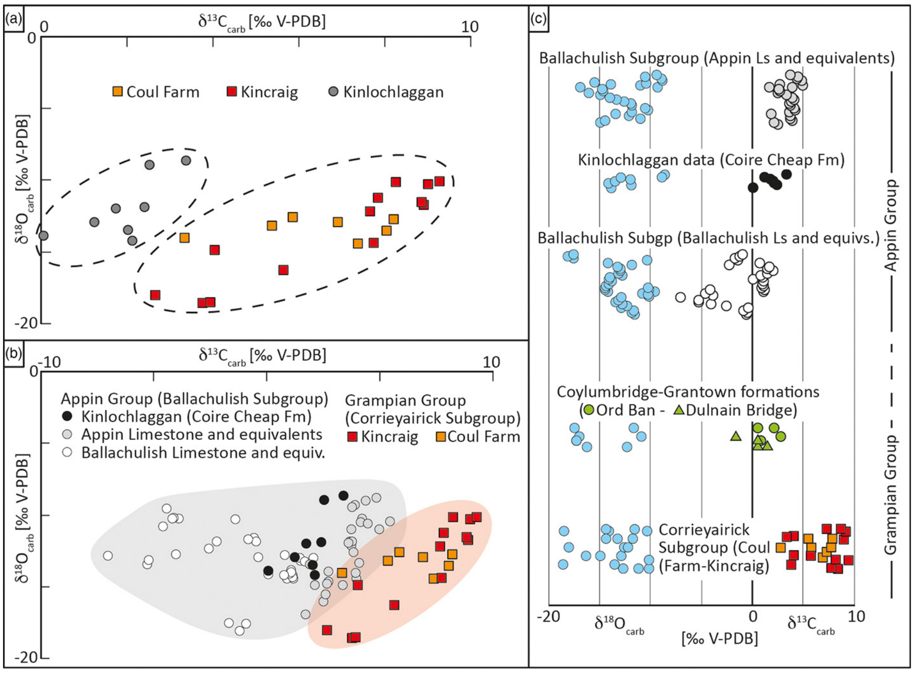

Fig. 5. (**a**) Carbon–oxygen isotope cross-plot showing clear clustering of data for marbles from Kincraig and Coul Farm v. those from Kinlochlaggan. (**b**) Carbon–oxygen isotope cross-plot showing the same data as in part (a), but with additional data for carbonate rocks of the Appin Group (Ballachulish Subgroup). (**c**) Relative age stratigraphic plot of carbon and oxygen isotope data for the Grampian Group marbles (Corrieyairick Subgroup, Coylumbridge–Grantown formations) and Appin Group limestones (Ballachulish Subgroup, Coire Cheap Formation). See text for discussion, Figure 2a for locations and Table 1 for the raw data.

depositional framework established by previous workers for the Grampian–Appin succession (Fig. 4b) is entirely compatible with this scenario, which is now strengthened by our carbon isotope and geochemical data.

What was lacking for this earlier hypothesis to be considered viable was a well-documented Neoproterozoic orogeny to generate flysch-to-molasse sedimentation. This is no longer the case. The careful application of modern, high-precision techniques to age-date minerals and ascertain their isotopic compositions has confirmed that the Scottish Highlands experienced two phases of Tonian-age, prograde amphibolite facies metamorphism and migmitization, one spanning *c.* 840–780 Ma and another *c.* 740–725 Ma (Piasecki and van Breeman 1993; Noble *et al.* 1996; Rogers *et al.* 1998; Highton *et al.* 1999; Hyslop and Piasecki 1999; Tanner and Evans 2003; Cawood *et al.* 2015; Mazza *et al.* 2018). These findings were from Sm–Nd ages on zoned garnets and U–Pb mineral ages combined with *P–T* studies showing that the peak metamorphic conditions reached *c.* 650–700°C and 6–9 kbar (Vance *et al.* 1998; Cutts *et al.* 2009, 2010). This period of metamorphism and deformation is termed the Knoydartian Orogeny. First defined by Bowes (1968), this orogeny is now recognized across the circum-North Atlantic region (Tanner and Evans 2003; Cawood *et al.* 2010; Krabbendam *et al.* 2022).

In the tectonic scenario we propose here (Fig. 7), the Knoydartian Orogeny can be thought of as part of a prolonged phase of collisional tectonics between northern Laurentia and Baltica during Tonian time, the so-called Valhalla Orogeny (Cawood *et al.* 2010).

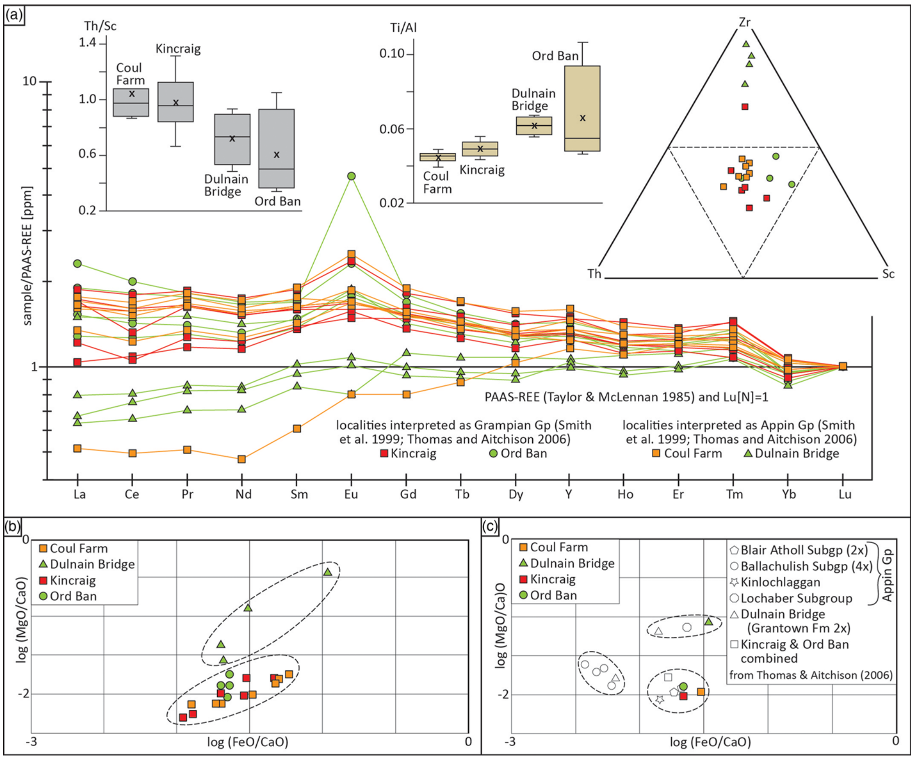

Fig. 6. Geochemical data for samples interpreted as belonging in the Grampian and Appin groups. (**a**) Rare earth element spider plot doubly normalized to PAAS and Lu; Th/Sc and Ti/Al box-and-whisker plots (x denotes median value, horizontal line marks mean value); and ternary diagram plot of Sc–Th–Zr. (**b**) Fe–Mg–Ca log-ratio plot of same data as in part (a). (**c**) Plot showing average Fe–Ca–Mg log-ratios for the data in part (b) and including Grampian Group and Appin Group data from Thomas and Aitchison (2006). See text for discussion and Table 2 and Supplementary material for the raw data.

Table 2. *Fe–Mg–Ca log-ratio data (average values for n samples)*

	log(FeO/CaO)	log(MgO/CaO)
Appin Group – Blair Atholl Subgroup		
Inchrory limestones (n = 160)*	−1.68	−1.91
Schiehallion limestones (n = 14)*	−1.73	−1.97
Appin Group – Ballachulish Subgroup		
Findhorn limestones (n = 7)*	−2.42	−1.61
Kyllachy limestones (n = 6)*	−2.28	−1.66
Torulian Formation (n = 16)*	−2.22	−1.88
Dufftown Formation (n = 49)*	−2.33	−1.70
Appin Group – Lochaber Subgroup		
Pitlurg Formation (n = 11)*	−1.63	−1.13
Marble localities discussed in the text		
Grantown A (n = 6)*	−2.19	−1.78
Grantown B (n = 8)*	−1.85	−1.18
Kinlochlaggan (n = 27)*	−1.84	−2.07
Ord Ban and Kincraig (n = 26)*	−1.78	−1.78
Coul Farm (n = 7)**	−1.52	−1.97
Dulnain Bridge (n = 4)**	−1.47	−1.07
Kincraig (n = 6)**	−1.66	−2.03
Ord Ban (n = 4)**	−1.66	−1.90

*Data from Thomas and Aitchison (2006); **data obtained in this study. See text for discussion and Supplementary material for raw data.

Given that the Dalradian Supergroup occurs south of the Great Glen Fault, which is thought to have had many hundreds of kilometres of left-lateral displacement during the Silurian–Devonian (Soper *et al.* 1992; Dewey and Strachan 2003; although see Searle 2021 for a contrary view), Dalradian basins would have been distal to the main areas affected by the Knoydartian Orogeny. Accordingly, the Knoydartian orogen and Laurentian craton would have been sources of detritus for the Grampian–Appin flysch-to-molasse succession, as indicated by detrital zircon provenance matching (e.g. Cawood *et al.* 2003). It needs to be noted that a Knoydartian-age peak (*c.* 840–725 Ma) is largely absent in the detrital zircon record of the Grampian–Appin rocks. We speculate two explanations: deeper

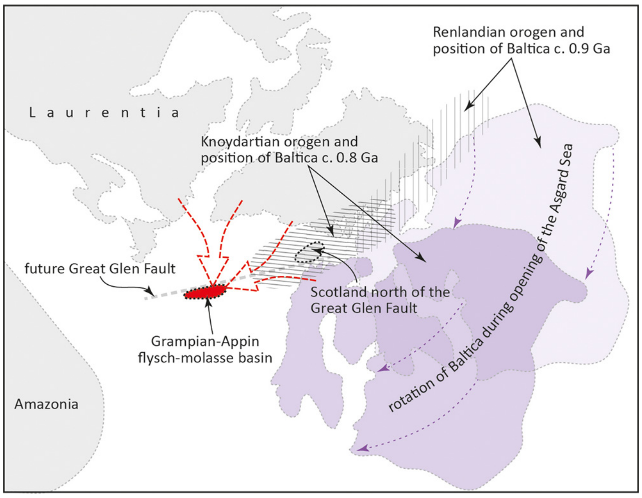

Fig. 7. Tectonic reconstruction of the North Atlantic realm between *c.* 1.0 and 0.8 Ga. Baltica–Laurentia interaction generates the Valhalla Orogen comprised of the older Renlandian and younger Knoydartian orogenies. Baltica is hypothesized to have rotated clockwise relative to Laurentia to create the Asgard Sea and, sequentially, these two orogenic episodes. Erosion of the Knoydartian orogen and cratonal Laurentia were the sediment sources for the Grampian–Appin flysch–molasse succession (dashed red arrows denote generalized sediment transport pathways). Source: after Cawood *et al.* (2010).

crustal levels of the orogen had not yet been exhumed during the time of Grampian–Appin deposition, or there was a dearth of zircon-bearing magmatic bodies in the sector of the orogen being eroded.

In summary, four independent datasets (carbon–oxygen isotope chemostratigraphy, lithofacies similarity, a many kilometre thick progradational depositional framework and detrital zircon age spectra) are supportive of interpreting the Grampian–Appin groups as a flysch-to-molasse transition linked to the Knoydartian Orogeny. However, as compelling as this interpretation is, it must also be compatible with the structural geology established for the northern Grampian Highlands via many decades of detailed mapping (Piasecki 1980; Haselock *et al.* 1982; Okonkwo 1988; Piasecki and Temperley 1988*a*, *b*; Glover and Winchester 1989; Phillips *et al.* 1999; Glover *et al.* 1995; Highton *et al.* 1999; Hyslop and Piasecki 1999; Robertson and Smith 1999; Smith *et al.* 1999; Banks and Winchester 2004; Leslie *et al.* 2013). We offer the following as a permissive interpretation of that geology.

Compatibility between structural and stratigraphic datasets

Since the efforts of Thomas (1979) and Piasecki (1980), all workers have recognized that the Aonach Beag–Inverpattack–Markie fault zones (see Figs 2a and 8) demarcate two regions of contrasting structural styles: NW of those faults are the tight, upright folds of the Geal Charn–Ossian Steep Belt, whereas kilometre-scale recumbent folds typify areas to the SE. Crucially, and acknowledging that kinematic indicators across these areas record various senses of displacement, structures with top-to-the-west/NW displacements (i.e. reverse faulting) can be observed in many places along the juncture between these two structurally distinct regions (see discussions in Leslie *et al.* 2013).

In step with those observations, we propose that the Aonach Beag–Inverpattack–Markie shear zones incorporate west/NW directed reverse (or thrust) faults (Fig. 8a–c). Such an interpretation was suggested previously (e.g. Piasecki 1980; Piasecki and Temperley 1988*b*), but rejected by later workers for two reasons (see discussion in Leslie *et al.* 2013). The first reason was the minor differences in the thickness and colour of psammitic and pelitic beds between the two key localities of Coul Farm and Kincraig. The second was the belief that the Coul Farm locality contained rocks belonging to the Appin Group. As we noted earlier, textural and compositional variation is a natural component of turbidite fan depositional systems, hence the variability of siliciclastic strata between localities is to be expected. Our findings reported herein also refute the postulated linkage of the rocks at Coul Farm to the Appin Group. These two criticisms are therefore no longer valid.

Our revised stratigraphy and structural framework offer a straightforward explanation of the geological relationships in the northern Grampian Highlands (Fig. 8a–c). The Aonach Beag–Inverpattack–Markie and related shear zones are, in part, reverse faults with older rocks, such as the Badenoch Group, in their hanging walls juxtaposed against younger rocks (Appin Group) in the Geal Charn–Ossian Steep Belt as footwall synclines. Such an interpretation can be accommodated with relatively minor modifications of the interpretative geophysical cross-section for the Grampian region (Fig. 8c; Smith *et al.* 1999). Further, comparing that cross-section with the nearby, well-documented region of extended continental crust of the North Sea shows how dissimilar the structural styles are between these two settings (Fig. 8d). The most notable divergence is the lack of normal fault displacements along the base of the Grampian Group. In fact, unfolding the Grampian Group rocks would result in the base of that succession becoming a relatively flat to low-amplitude undulatory surface. Above this surface, the Grampian Group forms an overall wedged-shaped depositional geometry, devoid of normal faults and lacking horst–graben style rift basins.

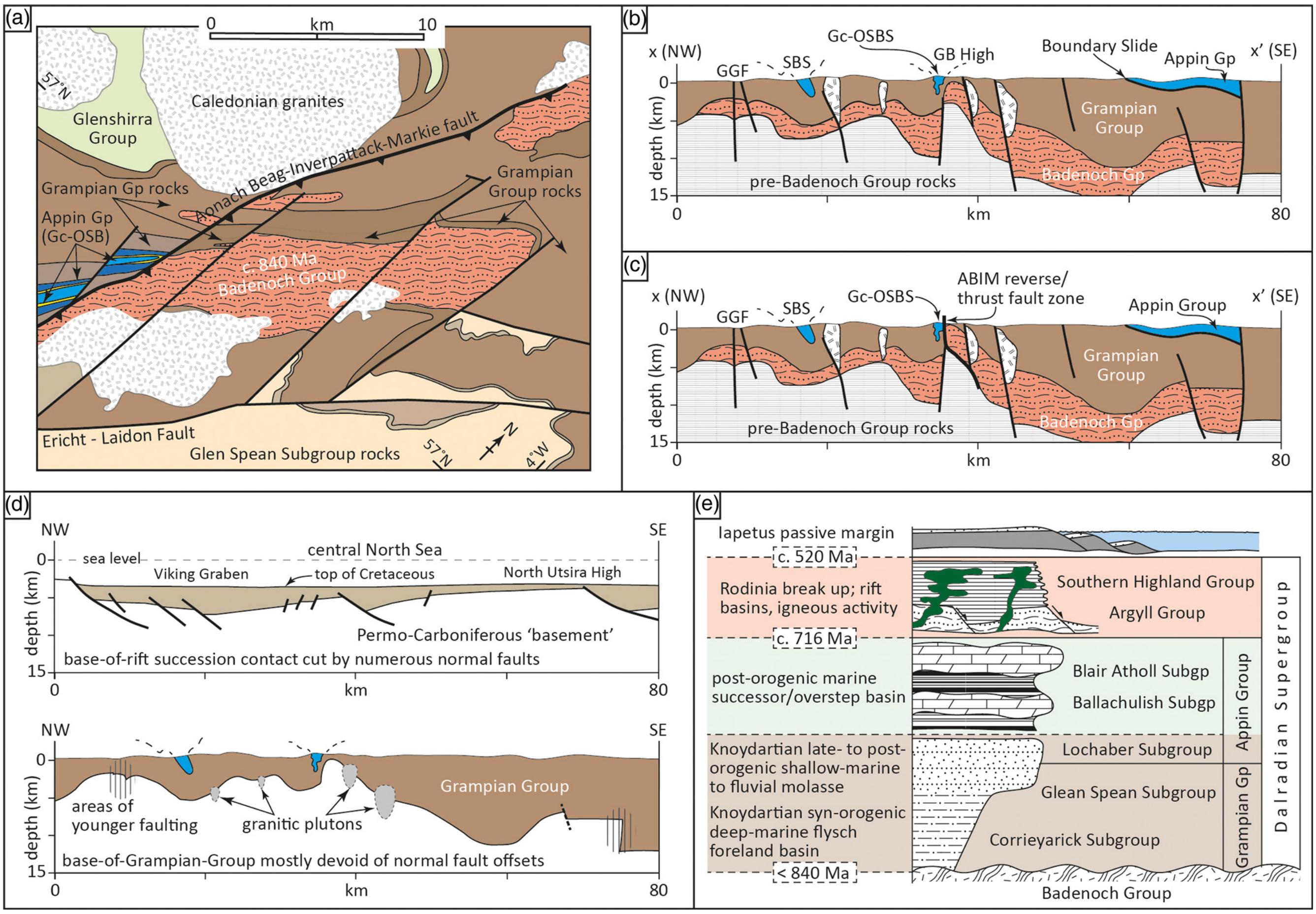

Fig. 8. (**a**) Revised stratigraphy and generalized map of the northern Grampian Highlands in which Grampian Group rocks envelope the gneissose rocks of the Badenoch Group. The Aonach Beag–Inverpattack–Markie fault belt is interpreted as an original reverse/thrust fault system with Appin Group rocks in the footwall synclines. (**b**) Geophysical cross-section. (**c**) Reinterpretation of cross-section in part (b) showing the Aonach Beag–Inverpattack–Markie as a reverse fault system juxtaposing Badenoch Group rocks in the hanging wall against Appin Group rocks in the footwall. (**d**) Comparison of central North Sea rift basins (after Phillips *et al.* 2019) and Grampian Group 'basins' redrawn and simplified from part (b). (**e**) Proposed tectonostratigraphic framework of the Dalradian Supergroup. ABIM, Aonach Beag–Inverpattack–Markie fault zone; GB, Glen Banchor; Gc-OSBS, Geal Charn–Ossian Steep Belt syncline; GGF, Great Glen Fault; SBS, Stob Ban synform. Source: map in part (a) simplified after British Geological Survey (2007); geophysical cross-section in part (d) from Smith *et al.* (1999).

Summary and conclusions

The Dalradian Supergroup has played a central part in tectonic models aimed at understanding the Neoproterozoic geological evolution of the North Atlantic region, particularly regarding the break-up of the supercontinent of Rodinia. Underpinning these models is the interpretation of the Grampian and Appin groups as having formed in rift basins – for example, the Corrieyairick–Strath Tummel sub-basins separated by the Glen Banchor High. This palaeogeography was predicated on using Fe–Mg–Ca log-ratios as a correlation tool to place the carbonate rocks adjacent to the purported Glen Banchor High into the Appin Group. Our carbon–oxygen isotope and geochemical compositional data for carbonate rock units from across the area of putative rift basins, combined with stratigraphic and sedimentological reassessments, show that correlation to be incorrect. Instead, our findings permit the placing of many of these carbonate rocks into the Grampian Group. Based on our revised stratigraphy, we interpret the Aonach Beag–Inverpattack–Markie shear zones as, in part, a belt of original reverse faults, an interpretation mooted previously by other workers (e.g. Piasecki 1980; Piasecki and Temperley 1988*b*). The Dalradian rocks SE of this belt belong to the Grampian Group, which unconformably overlies the Badenoch Group. Appin Group rocks are present NW of the belt in tightly folded synclinal structures (the Geal Charn–Ossian Steep Belt).

Viewing the Dalradian succession in its entirety, there is no evidence to infer that the deposition of the Grampian and Appin groups occurred in rift basins. An alternative interpretation, one compatible with our stratigraphic observations and new geochemical and carbon–oxygen isotope data, is that these groups record a deep marine turbiditic flysch that passes upwards into a shallow marine–shoreline molasse and, subsequently, evolves into an overstepping or successor basin (Fig. 8e). We attribute the genesis of that succession as a response to foreland flexing and loading during the Tonian Knoydartian Orogeny, not extensional tectonism. The late Tonian Grampian–Appin groups are the record of the erosion of the Knoydartian orogen and the adjacent cratonal hinterland of Laurentia.

It was during deposition of the overlying parts of the Dalradian Supergroup, the Cryogenian Argyll and Ediacaran Southern Highlands groups, that extensional tectonism came to the fore and where abundant geological evidence exists to infer the onset of rifting in Laurentia and the break-up of Rodinia, leading to the opening of the Iapetus Ocean during Ediacaran–Cambrian time. This evidence includes the characteristic stratigraphic patterns of abrupt lateral and vertical facies and thickness changes, synsedimentary normal faulting and the onset and expansion of mafic (and bimodal) volcanism. Consequently, geological scenarios for the mid- to late Neoproterozoic tectonic evolution of the North Atlantic region and the break-up of Rodinia need to be revised and refined regarding the timing of the onset of separation of Baltica from Laurentia.

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Data availability All data generated or analysed during this study are included in this published article (and if present, its Supplementary information files).

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