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Timing and heat sources for the Barrovian metamorphism, Scotland

Daniel R. Viete [a,b,*], Grahame J.H. Oliver [c,d], Geoff L. Fraser [e], Marnie A. Forster [b], Gordon S. Lister [b]

[a] Department of Civil Engineering, Monash University, Melbourne VIC 3800, Australia
[b] Research School of Earth Sciences, Australian National University, Canberra ACT 0200, Australia
[c] School of Geography and Geosciences, University of St Andrews, St Andrews KY16 9AL Fife, Scotland, United Kingdom
[d] Department of Geography, National University of Singapore, Singapore 119077, Singapore
[e] Minerals and Natural Hazards Division, Geoscience Australia, Canberra ACT 2609, Australia

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ABSTRACT

New SHRIMP U/Pb zircon ages of 472.2 ± 5.8 Ma and 471.2 ± 5.9 Ma are presented for the age of peak metamorphism of Barrovian migmatites. $^{40}Ar/^{39}Ar$ ages for white mica from the Barrovian metamorphic series are presented, and are recalculated using recently-proposed revisions to the ^{40}K decay constants to allow more precise and accurate comparison with U/Pb ages. The $^{40}Ar/^{39}Ar$ ages are found to vary systematically with increasing metamorphic grade, between c. 465 Ma for the biotite zone and c. 461 Ma for the sillimanite zone.

There is no evidence for any significant metamorphic heating during the first 15 Myr of the Grampian Orogeny (before c. 473 Ma) or the final 4 Myr (after c. 465 Ma). The Barrovian metamorphism occurred over a period of ~8 Myr within the ~27-Myr Grampian Orogeny. The Barrovian metamorphism records punctuated heating, was temporally and spatially associated with large-scale bimodal magmatism, and developed within crust that was not overthickened. The temporally distinct nature of the Barrovian metamorphic episode within the Grampian Orogeny, and its heating pattern and tectonic context, are not consistent with significant heat contribution from thermal equilibration of overthickened crust. Rather, the Barrovian metamorphism records a transient phase of crustal thermal disequilibrium during the Grampian Orogeny.

Temporal and spatial association with Grampian bimodal magmatism is consistent with production of the Barrovian metamorphic series within the middle crust as the result of advection of heat from the lower crust and/or mantle. The Barrovian metamorphic series – the classic example of 'orogenic regional metamorphism' – did not form in response to crustal thickening and thermal relaxation, but appears to record large-scale contact metamorphism.

1. Introduction

The Barrovian metamorphic series crops out in the SE of the Grampian Terrane, Scotland (Fig. 1), on the southern margin of the Scottish Highlands. The metamorphic sequence is celebrated as the setting for the work of Barrow (1893, 1912), who was the first to use indicative mineral assemblages to map regional metamorphism. The metamorphic progression across the Barrovian metamorphic series was defined by Barrow (1893, 1912) by the sequential first appearance of clastic mica (chlorite), biotite, garnet, staurolite, kyanite and sillimanite in the pelite metamorphic assemblage in the direction of increasing metamorphic grade, to the NW, away from the Highland Boundary Fault (HBF) (Fig. 1). Since Barrow's work, the Barrovian metamorphic series has become ensconced in the geological literature as the classic example of 'intermediate P/T' metamorphism, typically regarded as a consequence of continental collision (e.g. Spear, 1993). Such metamorphism is often referred to as Barrovian-type metamorphism.

The Barrovian metamorphism occurred at c. 470 Ma (Baxter et al., 2002; Oliver et al., 2000) during the Grampian Orogeny of Lambert and McKerrow (1976). The effects of the Grampian Orogeny in Scotland are mostly restricted to the Grampian Terrane, which crops out north of the HBF and south of the Great Glen Fault (Fig. 2). The Grampian Orogeny is also recorded in geological domains that crop out in certain regions of western and Northern Ireland, including the Connemara, Donegal, NW Mayo, Ox Mountains and Tyrone Central Inliers and Slishwood Division (see Fig. 2). Deformation and metamorphism during the Grampian Orogeny affected the sedimentary units of the Neoproterozoic to Ordovician (Strachan and Holdsworth, 2000) Dalradian Supergroup.

U/Pb zircon and Sm/Nd garnet geochronology has been used to restrict the total duration of the main tectonothermal phase of the Ordovician-age Grampian Orogeny of Scotland and Ireland to 12 to 15 million years, between 478 and 463 Ma (Friedrich et al., 1999a; Oliver et al., 2000). U/Pb zircon and Sm/Nd garnet geochronology has also been used to date the timing of metamorphic mineral growth during the Grampian-age Barrovian metamorphism at c. 470 Ma

* Corresponding author at: Department of Civil Engineering, Monash University, Melbourne VIC 3800, Australia.
E-mail addresses: daniel.viete@monash.edu, daniel.viete@gmail.com (D.R. Viete).

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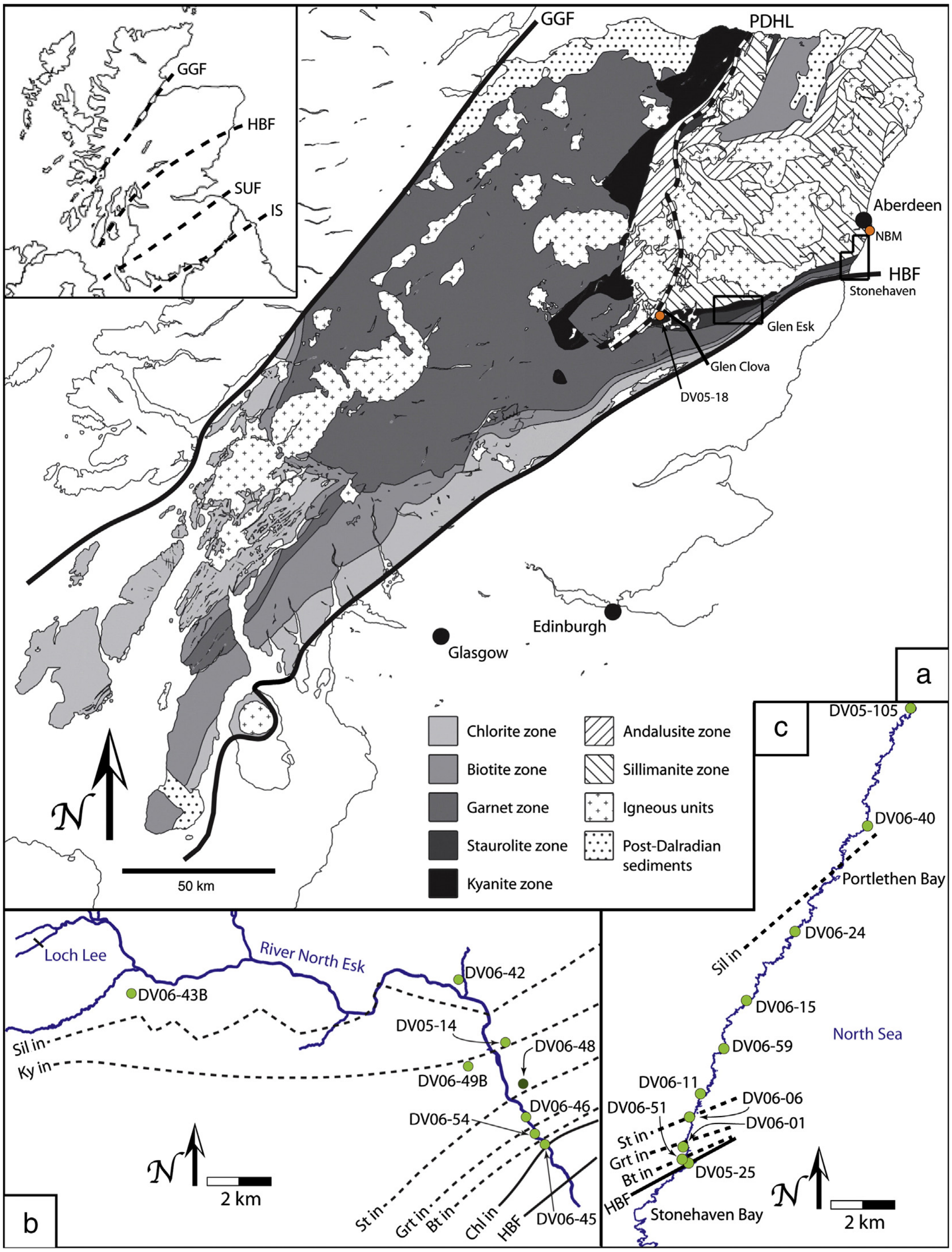

Fig. 1. Map of the Grampian Terrane, Scotland, showing (a) the distribution of metamorphic mineral isograds, magmatic bodies and post-Grampian sediments and the location of U/Pb SHRIMP samples (orange dots), (b) sample localities for $^{40}Ar/^{39}Ar$ samples (green dots) from the Glen Esk transect (enlargement of rectangle of Fig. 1a) and (1c) sample localities for $^{40}Ar/^{39}Ar$ samples (green dots) from the Stonehaven transect (enlargement of polygon of Fig. 1a). Inset at top left of Fig. 1a provides a broader location map. GGF: Great Glen Fault, HBF: Highland Boundary Fault, IS: Iapetus Suture, PDHL: Portsoy–Duchray Hill Lineament, SUF: Southern Uplands Fault. Compiled after Barrow (1912), Elles and Tilley (1930), Hudson (1980), Fettes et al. (1986), Harte (1987) and Viete et al. (2011a). Mineral abbreviations follow the recommendations of Kretz (1983). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article).

(Baxter et al., 2002; Oliver et al., 2000; Vorhies et al., 2013). Recent high-precision 'geospeedometry' and Sm/Nd garnet geochronology works have shown that the Barrovian metamorphism within the Grampian Orogeny occurred over short time scales of a few million years (Baxter et al., 2002; Dewey, 2005; Dewey and Mange, 1999; Oliver et al., 2000; Viete et al., 2011a) or less (Ague and Baxter, 2007) and involved episodic heating (Ague and Baxter, 2007; Viete et al., 2011b). Importantly, the short duration of Barrovian metamorphism has been demonstrated for rocks across the sequence, from the lower-grade (e.g. biotite and garnet) zones to the high-grade core (Ague and Baxter, 2007; Viete et al., 2011a). Such short time scales for the Barrovian regional metamorphism and an association with episodic heating have raised new questions regarding the origin of the heat and the tectonic context for the metamorphism (see Ague and Baxter, 2007; Viete et al., 2011a,b; Vorhies and Ague, 2011). Chief among these is the relative contribution of metamorphic heat derived

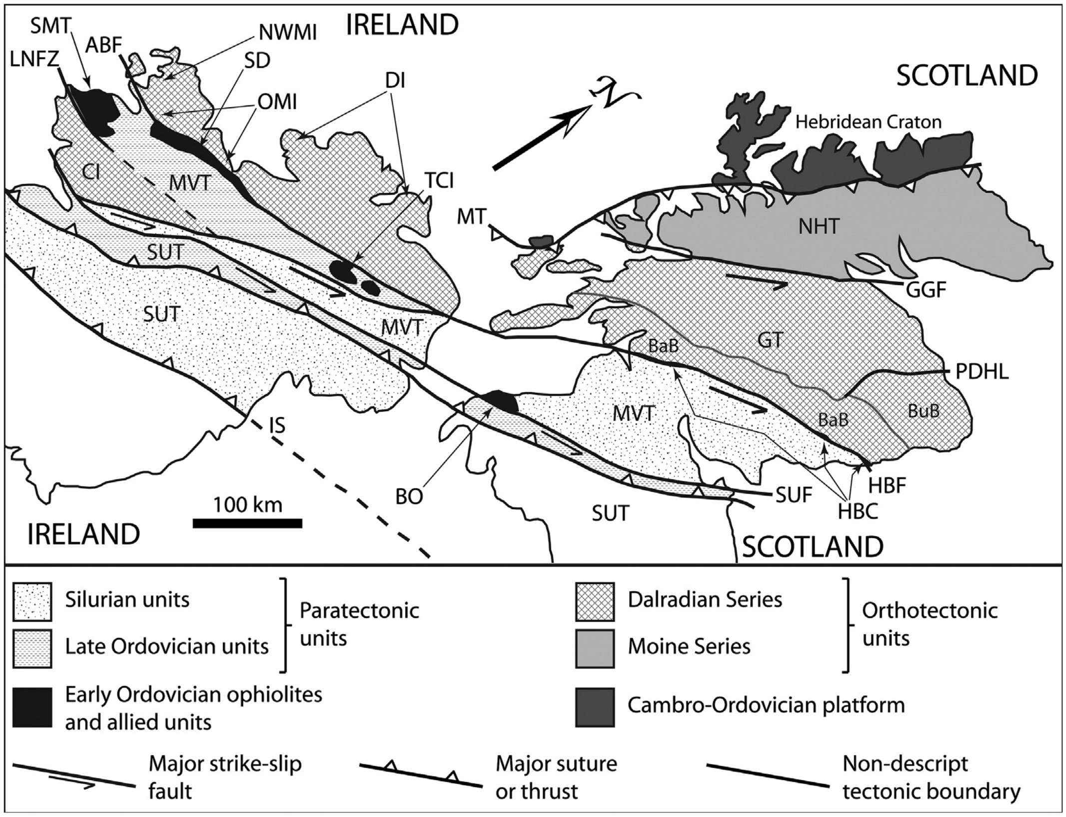

Fig. 2. Simplified tectonic map of the northern British Isles and Ireland showing the distribution of major terranes and the location of features relevant to this discussion of the Grampian Orogeny (modified from Dewey, 2005, p. 15288, Fig. 1a). ABF: Achill Beg Fault, BaB: Barrovian Block, BuB: Buchan Block, BO: Ballantrae Ophiolite, CC: Connemara Inlier, DI: Donegal Inlier, GGF: Great Glen Fault, GT: Grampian Terrane, HBC: Highland Border Complex, HBF: Highland Boundary Fault, IS: Iapetus Suture, LNFZ: Lough Nafooey Fault Zone, MT: Moine Thrust, MVT: Midland Valley Terrane, NHT: Northern Highlands Terrane, NWMI: NW Mayo Inlier, OMI: Ox Mountains Inlier, PDHL: Portsoy–Duchray Hill Lineament, SD: Slishwood Division, SMT: South Mayo Trough, SUF: Southern Uplands Fault, SUT: Southern Uplands Terrane, TCI: Tyrone Central Inlier.

from transient heat sources (e.g. localised magmatism, hot fluids, etc.) v. longer-duration heating related to distributed radioactive decay and conductive equilibration of overthickened crust.

In this study, new SHRIMP U/Pb zircon ages are presented for the timing of peak metamorphism in the highest-grade rocks of the Barrovian metamorphic series. White mica $^{40}Ar/^{39}Ar$ ages from a range of metamorphic zones across the Barrovian sequence are also presented, and constrain the timing of cooling that followed the Barrovian thermal activity. The $^{40}Ar/^{39}Ar$ ages are recalculated using recently-proposed revisions to the ^{40}K decay constants (Renne et al., 2010, 2011) that allow more precise and accurate geological comparison of the $^{40}Ar/^{39}Ar$ ages with U/Pb-based age constraints. The new geochronological constraints are linked to existing U/Pb geochronological constraints for the timing of magmatism and metamorphism during the Grampian Orogeny. Other pertinent information regarding the timing and origin of magmatism during the Grampian Orogeny and its relationship to Grampian regional metamorphism is also presented. Implications of the new geochronological data and previous observations with respect to the timing and context of the Barrovian metamorphism within the Grampian Orogeny, and the nature and origin of the Barrovian heat, are discussed.

2. Geological context

2.1. Other examples of Grampian metamorphism and their relation to the Barrovian metamorphism

The region immediately north of the Barrovian metamorphic series, in the NE of the Grampian Terrane, Scotland (the Buchan Block of Fig. 2), is occupied by the Buchan metamorphic series of Read (1923, 1952). The Buchan metamorphic series is characterised by the sequential first appearance of biotite, cordierite, andalusite and sillimanite in the pelite metamorphic assemblage in the direction of increasing metamorphic grade (Fig. 1). The sequence forms the classic example of regional 'low P/T' metamorphism (e.g. Spear, 1993).

The Barrovian and Buchan metamorphic series developed approximately contemporaneously at c. 470 Ma (Oliver et al., 2000), during the Grampian Orogeny. In places, the classic Barrovian and Buchan sequences share a common sillimanite zone core, which is focussed around the syn- to possibly post-metamorphic gabbroic intrusive rocks of the Grampian Terrane (Ashworth, 1975; Chinner, 1966; Fettes, 1970). In some parts of the sillimanite zones of the Barrovian and Buchan metamorphic series, rocks attained temperatures sufficient to cause partial melting of protoliths and production of migmatites.

Grampian-age metamorphic sequences that preserve pressure–temperature (P–T) conditions intermediate between Barrovian- and Buchan-type values occur within the Connemara Inlier of western Ireland (see Friedrich et al., 1999b; Wellings, 1998; Yardley et al., 1987). Barrovian-type sequences of the same age and nature as the Barrovian metamorphic series of Scotland occur in western and Northern Ireland, within the Donegal, NW Mayo, Ox Mountains and Tyrone Central Inliers and in the Slishwood Division (Chew et al., 2003, 2008; Flowerdew et al., 2005; Yardley et al., 1987).

Grampian blueschists (metabasic rocks with crossite–glaucophane assemblages) have been described in outcrop in South Achill, western Ireland (Gray and Yardley, 1979; Yardley et al., 1987), and the region of Ballantrae, SW Scotland (Bloxam and Allen, 1959; Kawai et al., 2008). Outcrops of Grampian-age high-pressure/low-temperature metamorphic rocks are rare and localised, but significant eclogite/blueschist detritus in Llanvirn- to Caradoc-age sediments of the Southern Uplands Terrane is suggestive of more widespread occurrence during the middle to late Ordovician (Mange et al., 2005; Oliver, 2001). Development of the

Grampian blueschists has been related to a high-pressure (c. 1 GPa: Chew et al., 2003; Sawaki et al., 2010) metamorphic episode that was contemporaneous with regional Barrovian-type metamorphism (Chew et al., 2003).

Barrovian-type metamorphic (biotite, garnet and kyanite zone) assemblages are widespread in the Grampian Terrane, Scotland, extending to the SW and Central Grampian Highlands (see Fig. 1a). Examples of high-pressure Barrovian-type metamorphism in the west of the Grampian Terrane have been related to crustal thickening, with garnet thermobarometry showing that metamorphism involved an increase in pressure for a modest (see Pattison, 2013, Fig. 11, p. 431) or negligible (see Vorhies and Ague, 2011, Fig. 14, p. 1161) increase in temperature. The amphibole thermobarometry of Zenk and Schulz (2004) demonstrated that Barrovian-type 'burial' metamorphism in the Grampian Terrane involved a decrease in temperature-depth ratios and steep Pressure–Temperature trajectories. Metamorphism associated with early-Grampian crustal thickening may have been responsible for both isothermal (or near-isothermal) Barrovian-type burial metamorphism and the blueschist metamorphism of Gray and Yardley (1979) and Yardley et al. (1987). We must emphasise here that this metamorphism was predominantly driven by pressure increase and was not necessarily related to any significant 'thermal event'.

Metamorphism of the high-grade Grampian Barrovian- and Buchan-type metamorphic rocks of the Scottish and Irish Grampians, including the type Barrovian and Buchan sequences, was driven by significant heating during exhumation and decompression (see Chew et al., 2003; Viete et al., 2010; Vorhies and Ague, 2011; Yardley et al., 1987). For example, the Barrovian metamorphic heating did not occur until after peak pressure conditions had already been obtained (see Vorhies and Ague, 2011, Fig. 14, p. 1161). Grampian burial metamorphism preceded Grampian thermal metamorphism (Vorhies and Ague, 2011), meaning that the Barrovian metamorphism was a thermal overprint on pre-existing, higher-pressure metamorphic assemblages that had resulted from essentially isothermal metamorphism.

2.2. Grampian magmatism and ideas on its relationship to the Barrovian metamorphism

Igneous rocks are abundant in the Grampian Terrane, Scotland, particularly in the regions of the Barrovian and Buchan metamorphic series, in the SE and NE, respectively (Fig. 2). On the basis of deformation intensity recorded in the igneous rocks of the Grampian Terrane, Read (1919) was able to recognise 'older' and 'younger' generations of magmatism that affected the Grampian Terrane. The 'older' magmas of Read (1919) were emplaced during deposition of the Dalradian Supergroup (see Tanner et al., 2006) and pre-date the Grampian Orogeny. The 'younger' intrusive bodies of Read (1919) include a Grampian-age bimodal magma (gabbro and granite) sequence and a post-Grampian (S-type) granite sequence (Oliver, 2001; Oliver et al., 2008). In this manuscript magmas are referred to in terms of their emplacement age with respect to Grampian deformation/tectonism (i.e. pre-Grampian, Grampian or post-Grampian).

In parts of the NE of the Grampian Terrane, Scotland, the highest-grade (sillimanite zone) Grampian metamorphic rocks display a strong spatial association with the distribution of the Grampian gabbros (Ashworth, 1975; Chinner, 1966; Fettes, 1970). A similar spatial association has been observed in western Ireland, where the highest-grade Grampian metamorphic rocks of the region crop out in regions surrounding large mafic intrusions (Leake, 1969, 1989; Wellings, 1998; Yardley et al., 1987). In places, sillimanite zone metamorphism and formation of Grampian migmatites has been related to contact metamorphism resulting from the intrusion of the Grampian gabbros in Scotland (Ashworth, 1975, 1976; Chinner, 1966; Fettes, 1970; Harte and Hudson, 1979; Pankhurst, 1970) and associated mafic igneous intrusions in Ireland (Leake, 1989; Wellings, 1998; Yardley et al., 1982, 1987).

Some association between mafic magmatism and high-grade metamorphism (and migmatisation) during the Grampian Orogeny is well established. However, the primary heat sources responsible for the broader Grampian regional metamorphism – including for the classic Barrovian and Buchan metamorphic sequences – are still poorly understood. Fettes (1970) offered two alternatives for interpretation of the connection between Grampian magmatism and the Buchan regional metamorphism: (1) the Grampian gabbros were emplaced into rocks experiencing regional metamorphism and caused a high-grade contact metamorphic overprint on already-formed regional isograds, or (2) the Grampian gabbros were emplaced into rocks that had experienced only limited metamorphism and the heat that they brought with them was the primary driver for the regional metamorphism.

The two alternative interpretations of Fettes (1970) are well summarised by the influential models for Grampian regional metamorphism proposed by Chinner (1966) and Harte and Hudson (1979). Chinner (1966) interpreted contact metamorphism associated with the Grampian gabbros as an overprint on a pre-existing and independent regional metamorphic pattern (and thermal regime). Harte and Hudson (1979) favoured a model in which the Grampian sillimanite and migmatite zones developed within the same heating scenario that produced the regional metamorphic pattern they overprint. The model of Chinner (1966) suggests ongoing regional metamorphism and a later (independent) contact metamorphism related to a distinct heat source. On the other hand, the model of Harte and Hudson (1979) implies that the regional metamorphism was in fact regional contact metamorphism.

The model of Harte and Hudson (1979) follows Barrow's original view of the Barrovian regional metamorphism as contact metamorphism on a grand scale, involving a large-scale network of igneous intrusions (though Barrow, 1893 considered dominantly pre-Grampian granites to have been responsible). The general scarcity of igneous intrusions in the Grampian Highlands and the lack of any clear association between the Barrovian isograds and the location of igneous bodies in outcrop in the lower-metamorphic-grade regions meant that Barrow's proposal for a contact-metamorphic origin of the Barrovian heat was generally unpopular. The Barrovian metamorphism, and associated metamorphism elsewhere within the Grampian Terrane, Scotland, and in Ireland, instead came to be viewed as the result of larger-scale and more enigmatic 'regional metamorphic' heat sources associated with orogenesis and crustal thickening (see Richardson and Powell, 1976; Yardley et al., 1987, and the models for orogenic metamorphism of Oxburgh and Turcotte, 1974; Bickle et al., 1975; England and Thompson, 1984; Jamieson et al., 1998).

2.3. Origins of the Grampian gabbros and tectonic implications

On the basis of geochemistry and mineralogy, the Grampian gabbros of Ireland have been interpreted to originate from arc magmatism (Cooper et al., 2011; Draut and Clift, 2001; Draut et al., 2002, 2009; Thompson et al., 1985; Yardley and Senior, 1982). The geochemistry and mineralogy of the Grampian gabbros of Scotland, on the other hand, do not display the same arc-related geochemistry and mineralogy (Yardley et al., 1982) and their origins have been related to collision and slab breakoff (Oliver, 2002; Oliver et al., 2008), or decompression melting of sub-lithospheric mantle (Viete et al., 2010).

Despite apparent differences in geochemistry and mineralogy between the Grampian gabbros of Ireland and Scotland (see Yardley et al., 1982), the Rare Earth Element (REE) patterns display close similarities (compare results of Viete et al., 2010 from Scotland with those of Draut et al., 2002, 2009; Cooper et al., 2011; Hollis et al., 2012 from Ireland). All Grampian gabbros show little variation in normalised REE abundance with increasing atomic number for the heavy REEs, from Tb to Lu (Fig. 3). Strong fractionation of the heavy REEs, between melt and restitic garnet, during melting in the presence of garnet will produce a significant negative slope across the heavy REEs (see Kay and Gast, 1973). The flat heavy REE patterns for all Grampian gabbros from Ireland and Scotland (Cooper et al., 2011; Draut et al., 2002, 2009; Hollis et al., 2012; Viete et al., 2010) are consistent with their production by mantle melting at depths shallower than the spinel lherzolite–garnet lherzolite transition, which occurs at pressures of between 1.7 and 2.1 GPa, for mantle temperatures between 1100 and 1300 °C, respectively (Klemme and O'Neill, 2000). The flat heavy REE patterns in Grampian gabbros are thus consistent with melting at depths ≤70 km, for mantle temperatures less than 1300 °C and an average overburden density of 3000 kg m^{-3}. For the Scottish gabbros – which are not arc-related and formed by decompression melting of the asthenosphere (Viete et al., 2010) – this requires a lithospheric thickness ≤70 km, which is very thin for a setting experiencing active orogenesis. Some tectonic processes that could produce such thinned lithosphere within an orogenic setting may include lithospheric-scale, syn-orogenic extension (Beltrando et al., 2010; Viete et al., 2010), or catastrophic loss of sub-continental lithosphere by delamination (Nelson, 1992; Schott and Schmeling, 1998) or convective removal (England, 1993; Platt and England, 1993; Platt et al., 2003).

On the basis of a large catalogue of geochemistry of arc basalts, Mantle and Collins (2008) proposed a relationship that predicts depth of the Moho during melting from the maximum Ce/Y ratio obtained for mafic magmas. According to their relationship, mantle melting to produce the Grampian gabbros and basalts of the Connemara Inlier, Ireland, (maximum Ce/Y = 3.19: Draut et al., 2002) Portsoy, Scotland, (maximum Ce/Y = 1.93: Viete et al., 2010) and Tyrone, Ireland,

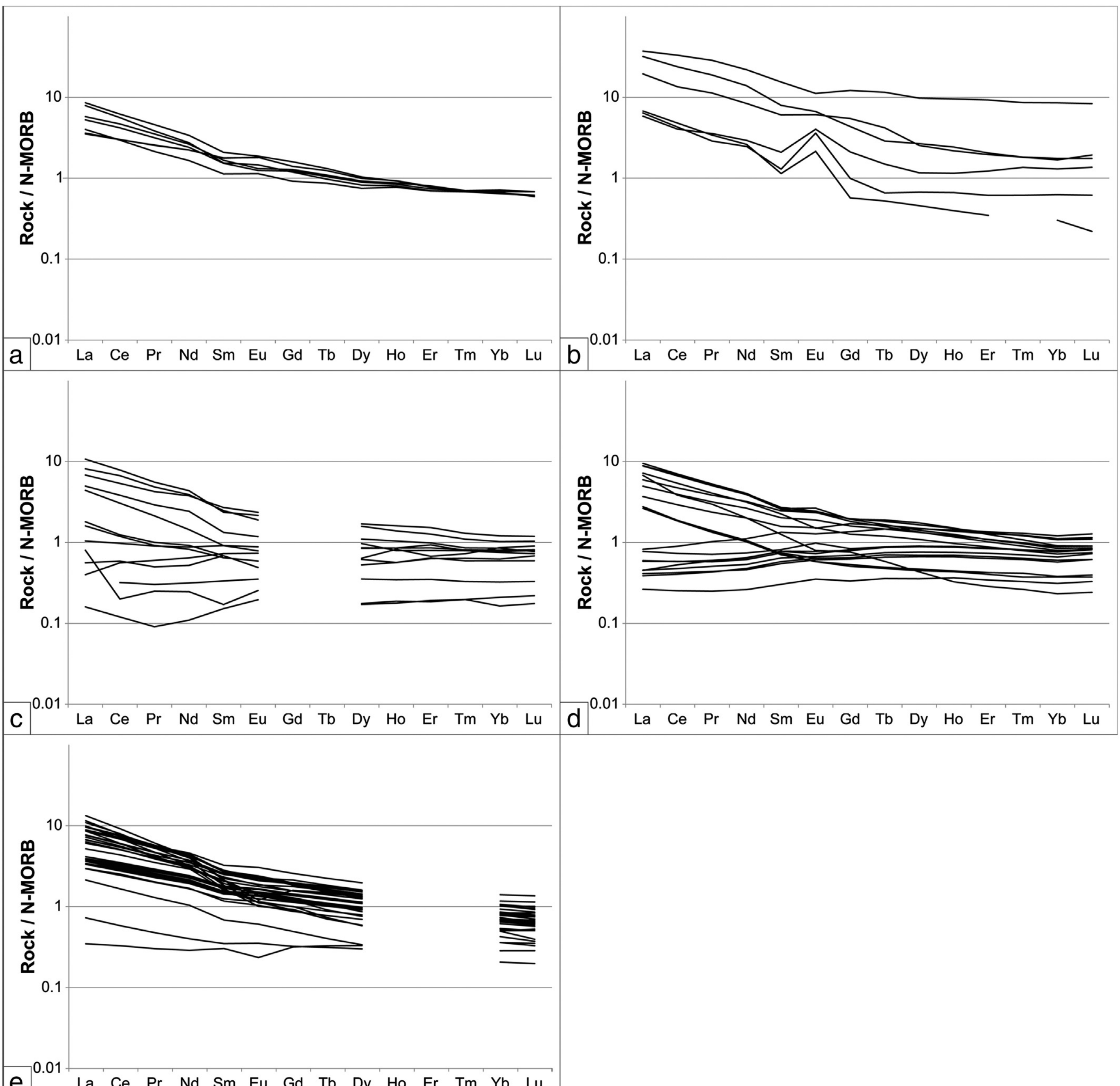

Fig. 3. Rare Earth element plots for Grampian igneous units with SiO_2 <55 wt.%, from: (a) Portsoy, NE Scotland (Viete et al., 2010); (b) Connemara, western Ireland (Draut et al., 2002); (c) Tyrone Central Inlier, Northern Ireland (Draut et al., 2009); (d) Tyrone Central Inlier, Northern Ireland (Cooper et al., 2011), and (e) Tyrone Central Inlier, Northern Ireland (Hollis et al., 2012).

(maximum Ce/Y = 3.51: Draut et al., 2009; Cooper et al., 2011; Hollis et al., 2012) occurred when the crust was between 33 and 44 km thick. It is clear that mantle melting to produce the Grampian gabbros occurred beneath continental crust that was not unusually thick. The oldest Barrovian detritus in basins adjacent to the Grampian Terrane belongs to the middle Llanvirn (c. 465 Ma) Kirkland conglomerate of the Midland Valley Terrane (Dewey and Mange, 1999; Oliver, 2001; Oliver et al., 2000, 2008). The lack of any pre-465 Ma Barrovian detritus in regions that neighboured the Grampian Orogen is not consistent with the presence of any significant orogenic edifice (detritus source) during the Barrovian metamorphic event (i.e. at c. 470 Ma). This observation also suggests that the crust was of normal thickness during the Barrovian metamorphism.

Crust and lithosphere of normal thickness during the Barrovian thermal event is consistent with the Barrovian metamorphism being fundamentally distinct from the burial (pressure-driven) metamorphism observed for the higher-pressure Barrovian-type metamorphic rocks of the SW of the Grampian Terrane, Scotland (see Ague and Baxter, 2007; Baxter et al., 2002; Vorhies and Ague, 2011), and the Grampian blueschists.

2.4. Origins of the Grampian granites

Johnson et al. (2003) demonstrated similarity in the major and trace element geochemistry of the leucosomes of Grampian-age metapelitic migmatites from NE Scotland and that of the local Grampian granites. On the basis of the observed geochemical similarity, Johnson et al. (2003) proposed a common crustal-melting origin for the Grampian granites and the Grampian anatectic migmatites. Although the role of the Grampian gabbro emplacement in driving anatexis in the field area of Johnson et al. (2003) is not entirely clear, production of the Grampian granites by accumulation of anatectic melts within regional domains of elevated metamorphic grade locally associated with the Grampian gabbros has been inferred on the basis of field observations from other regions of NE Scotland (Droop et al., 2003; Goodman, 1991). The mechanisms that would accommodate segregation of crustal melts from regions of elevated metamorphic grade surrounding the Grampian gabbros, and subsequent ascent and emplacement of them to form the Grampian granites, are relatively well understood (see Brown, 1994, 2007 and references therein). Field documentation of mafic enclaves in Grampian tonalites from Ireland (Flowerdew et al., 2005) confirms interaction of mafic and felsic magmas during their melting, emplacement and/or crystallisation history. The results of oxygen isotope analyses performed by Appleby et al. (2010) on zircons from the Nigg Bay Granite of NE Scotland have demonstrated that the magma from which this Grampian granite crystallised carries a mantle component, as would be expected if the Grampian granites formed from anatexis in the region surrounding mid-crustal intrusions of Grampian gabbro.

Experimental and analytical modelling has demonstrated that the heating caused by intrusion of mantle melts into the crust is sufficient to produce significant crustal anatexis and large-volume felsic intrusive magmas (Huppert and Sparks, 1988; Bergantz, 1989). The process of advective heating and melting of the crust is particularly effective in systems where a broad thermal aureole is produced following a sustained period of episodic mafic magma injection (Petford and Gallagher, 2001; Annen and Sparks, 2002). These models predict a progression in the style of magmatism such that magmas become more felsic with time (i.e. early mafic magmatism/late felsic magmatism). This trend in magma character has been observed for the Grampian Orogeny (Yardley et al., 1982; Leake, 1989; Friedrich et al., 1999b; Cooper et al., 2011) and other collisional orogens (e.g. Pitcher, 1978; Ratajevski et al., 2001) and is consistent with felsic magmatism as the result of mid-crustal anatexis, driven by advection of heat from the mantle during the initial phase of mafic magmatism.

3. New data for the timing of the Barrovian metamorphism

In this contribution we present new constraints on the timing of the Barrovian metamorphic event in Scotland. These new dates include: (1) U/Pb SHRIMP zircon dating of metamorphic zircon from Barrovian migmatites to obtain estimates for the timing of peak metamorphism in the highest grades, and (2) calibration of ^{40}Ar/^{39}Ar step heating ages from the Barrovian metamorphic series, published by Viete et al. (2011a), to the U/Pb dating system. Samples for the peak-metamorphic U/Pb SHRIMP work were collected from Glen Clova, in the northern upwaters of the River South Esk [DV05-18, GPS: NO28857343], and from Nigg Bay, immediately south of Aberdeen, on the Scottish east coast [NBM, GPS: NJ97030515]. Locations for the migmatite samples are shown on Fig. 1a. Samples that provided ^{40}Ar/^{39}Ar ages for calibration to the U/Pb system were obtained from two transects through the Barrovian metamorphic series, along the River North Esk at Glen Esk (see Fig. 1b) and on the Scottish east coast, north of Stonehaven (see Fig. 1c).

3.1. New SHRIMP U/Pb ages for the peak of Barrovian metamorphism

To constrain the timing of peak metamorphism during the Barrovian event, a SHRIMP U/Pb approach was used to date the growth of metamorphic zircon within two Grampian-age anatectic pelites (DV05-18 and NBM). In each case, the migmatite samples were obtained from outcrop comprising thin and concordant, K-feldspar-bearing leucosome segregations that delineated thicker, biotite-rich melanosome bands. Due to the textures they preserved, these migmatites were interpreted to have formed by in situ partial melting. McLellan (1983, 1989) made identical interpretations for the mode of formation of the same (Barrovian) sillimanite-grade migmatites.

3.1.1. Method

NBM was dominantly leucosome and was crushed whole. Zircon-rich leucosome for DV05-18 was concentrated by coarse chipping of the sample to remove melanosome material. The leucosome-rich material was crushed and sieved to a >64 mm fraction, which was washed, then milled in tungsten carbide. The milled material was sieved to a 120 to 250 μm fraction, which was subjected to conventional heavy liquid and magnetic separation techniques to concentrate the zircon component. Individual zircon grains from each sample were handpicked under microscope and mounted in epoxy with zircon standards SL13 (U concentration of 238 ppm: Claoué-Long et al., 1995) and Temora 2 (^{206}Pb/^{238}U age of 416.8 ± 1.1 Ma: Black et al., 2003). The epoxy mount was fashioned into a disc then polished using a rotary polisher and diamond paste to expose individual grains at their approximate mid-section. Reflected and transmitted light images were produced for the mount to assist in navigation and identification of physical imperfections within grains. Cathodoluminescence spectroscopy, carried out using the Hitachi S-2250N scanning electron microscope housed at the Research School of Earth Sciences (RSES), the Australian National University (ANU), provided information on growth structure within analysed grains and was used to identify specific targets for analysis.

Isotopic analyses were performed on the Reverse Geometry Sensitive High-Resolution Ion Microprobe (SHRIMP RG) at the RSES, ANU, following methodologies set out in Williams (1998). Following a 40 μm, 120 s raster to clean the analysed surface, a 0.5 nA primary beam was focussed to a 10 μm spot size to provide material for analysis. Measurements for each spot were made from 6 scans of the isotopic masses of interest. One SL13 analysis was made at the start of each session to calibrate for U abundance and one Temora 2 standard was run for every three unknowns to calibrate for the ^{206}Pb/^{238}U ratio. Data reduction was carried out using the purpose-built Lead 6.5.5 and Prawn 6.6.0 applications (Williams et al., 1996; Williams, 1998) for the Mac OS X platform. Data reduction was initially performed using a correction for common Pb based on measured ^{204}Pb, assuming a common Pb (Pb_c) isotopic composition of Broken Hill Pb (methodology as per Williams, 1998). However, this approach was found to produce reverse discordance in $^{206}Pb/^{238}U$ v. $^{207}Pb/^{235}U$ plots due to overestimation of $^{206}Pb_c$ in the sample, caused by low or absent ^{204}Pb. Data reduction was repeated with a ^{207}Pb correction, again assuming a Pb_c isotopic composition of Broken Hill Pb (methodology as per Williams, 1998) that avoided issues arising from low ^{204}Pb counts during acquisition.

3.1.2. Results

Barrovian-age zircon was found to occur as overgrowths on detrital cores that commonly display Grenvillean (c. 1000 Ma) ages (Fig. 4a–e), or as small and elongated primary grains (Fig. 4f). The sectors of the analysed zircons that yielded Grampian ages generally were rich in U and displayed relatively low Th/U ratios (most < 0.01, all < 0.30). These characteristics are consistent with crystallisation of the Grampian-age zircon as the result of metamorphic processes (Williams et al., 1996).

The results of SHRIMP analyses carried out on the anatectic pelite samples DV05-18 and NBM are summarised in Table 1. Cathodoluminescence images and screen captures of ion-probe spots allowed identification of analyses that had sampled inherited cores or regions that were a mixture of core and rim. Only analyses of zircon rims or grains interpreted as of metamorphic origin (on the basis of U content and Th/U values: Williams et al., 1996) were considered in calculating the metamorphic age of each sample.

Thirty-three analyses were collected from sample DV05-18. Ten of these were from inherited cores or a mixture of core and rim, leaving 23 analyses of metamorphic zircon. These 23 analyses combine to give a weighted mean $^{206}Pb/^{238}U$ age of 485 ± 17 Ma (MSWD = 10.5, probability of fit = 0.00). The high MSWD indicates excess scatter in these data. Seven of the 23 metamorphic zircon analyses can be excluded from the mean age calculation on the following basis: (1) analysis 17.1 contains very low U abundance (4 ppm) and as a consequence is very imprecise; (2) analysis 13.2 contains almost 6% common Pb (f206c), and (3) analyses 2.1, 5.1, 8.1, 14.1, 14.2 all contain >2500 ppm U and the ages from these analyses are regarded as suspect due to matrix effects related to their high U content (e.g. Williams and Hergt, 2000; White and Ireland, 2012). Three of the five high-U analyses plot as clearly older outliers relative to the main age population. If these seven analyses with anomalous U content or high common Pb are excluded, the remaining 16 analyses yield a weighted mean $^{206}Pb/^{238}U$ age of 472.2 ± 5.8 Ma (MSWD = 1.4, probability of fit = 0.16). This is regarded as the best estimate of the age of Barrovian migmatite-grade metamorphism in DV05-18.

Thirty-one analyses were collected from sample NBM, of which 13 were from inherited cores or mixed core and rim regions. The remaining 18 analyses yield a weighted mean age of 471.2 ± 5.9 Ma (MSWD = 0.76, probability of fit = 0.74), which is regarded as the best estimate for the timing of migmatite-grade metamorphism in this sample.

3.2. New U/Pb-calibrated $^{40}Ar/^{39}Ar$ ages for the demise of Barrovian metamorphism

Significant uncertainty is introduced when one compares radiometric ages obtained using different isotope systems. This uncertainty arises from the uncertainties in parameters such as decay constants, which are specific to each isotopic method. Such systematic uncertainty can obscure the fine resolution in geochronological data required to discern individual events during a short orogenic episode such as the Grampian Orogeny. The geochronological community has started to address the issue of the disparity between radiometric ages that have used different isotope systems (see Begemann et al., 2001). Work on the topic has generally involved calibration of decay constants by forcing an equivalence of ages obtained using different radiometric systems on a single volcanic sample that is well characterised for geochronology (i.e. dating standards) (e.g. Renne et al., 1998, 2010; Villeneuve et al., 2000; Kwon et al., 2002). The reason for the use of volcanic samples is that they cool from high temperature/crystallise instantaneously (in geological terms), meaning that the various mineral-isotope system pairs should set simultaneously, despite differences in their diffusivities (and closure temperatures).

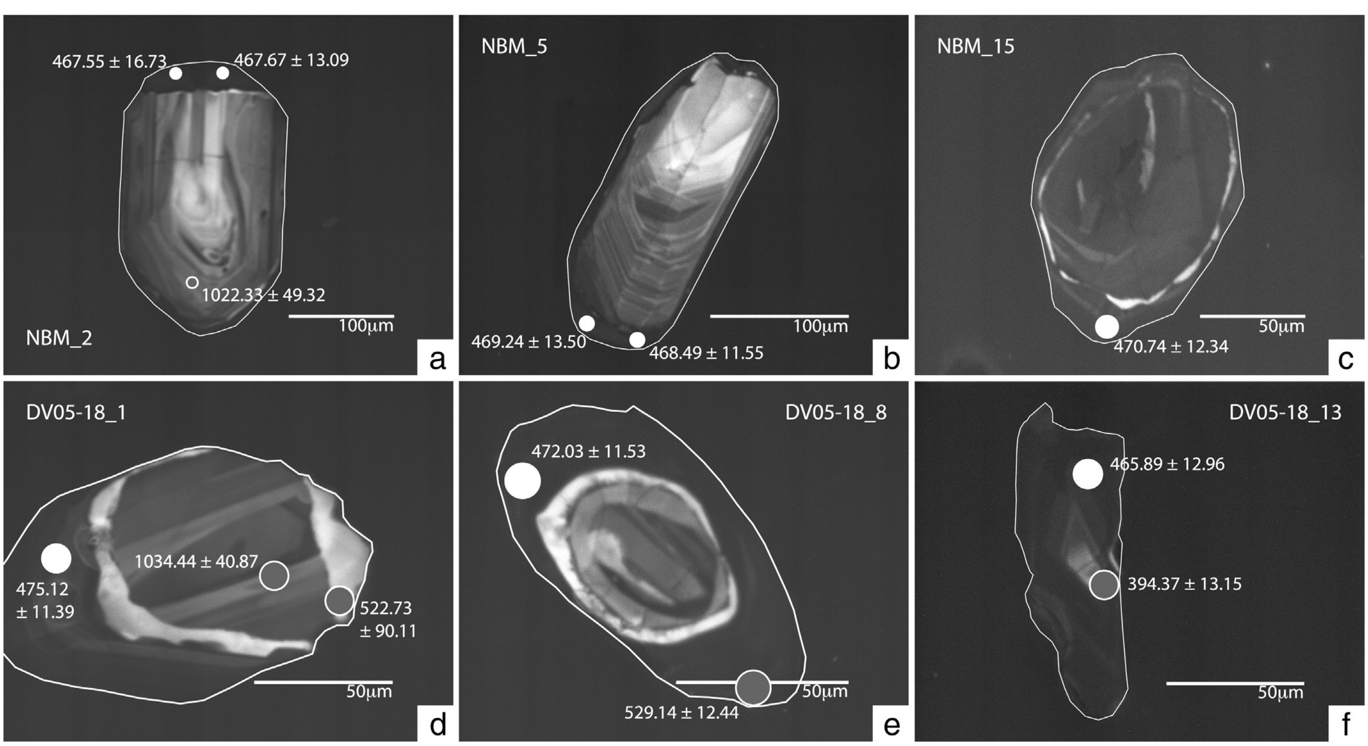

Fig. 4. Cathodoluminescence images of some zircons analysed by SHRIMP. Analysis locations and $^{206}Pb/^{238}U$ spot ages with 1σ uncertainties are shown. Analyses used to calculate metamorphic ages are shown as white spots and those that were not used are shown as grey spots.

Table 1
U/Pb isotope data table for SHRIMP analyses. Uncertainties on individual analysis ages are quoted to 1σ, uncertainties on mean ages are quoted to 2σ. Analyses not used in the final calculation of SHRIMP ages are shaded in grey.

Spot	Domain	U (ppm)	Th (ppm)	Th/U	f206 (%)	$^{238}U/^{206}Pb$	Age (Ma)
DV05-18 [GPS: NO28857343]							
1.1	rim	838	0	0.0003	0.531 ± 0.183	13.074 ± 0.325	475.12 ± 11.39
1.2	mixed	13	0	0.0011	0.839 ± 2.473	11.839 ± 2.110	522.73 ± 90.11
1.3	core	207	119	0.5782	2.291 ± 0.549	5.745 ± 0.245	1034.44 ± 40.87
2.1	rim	2953	2	0.0007	0.104 ± 0.226	12.119 ± 0.366	511.10 ± 14.84
2.2	core	1356	97	0.0716	1.699 ± 0.323	11.228 ± 0.265	549.97 ± 12.44
3.1	rim	422	0	0.0003	0.787 ± 0.640	13.004 ± 0.514	477.58 ± 18.24
4.1	rim	3078	6	0.0021	0.702 ± 0.231	13.301 ± 0.326	467.31 ± 11.06
4.2	rim	1993	1	0.0003	0.444 ± 0.293	12.677 ± 0.349	489.45 ± 13.01
5.1	rim	19786	33	0.0016	−0.039 ± −0.094	10.899 ± 0.256	565.89 ± 12.73
5.2	rim	2917	2	0.0006	0.367 ± 0.239	12.601 ± 0.303	492.28 ± 11.40
6.1	rim	2994	2	0.0007	0.088 ± 0.132	12.906 ± 0.312	481.08 ± 11.23
6.2	core	529	57	0.1085	4.043 ± 0.460	4.564 ± 0.170	1277.06 ± 43.29
7.1	mixed	1898	188	0.0988	3.633 ± 0.189	4.646 ± 0.106	1256.71 ± 26.23
8.1	rim	4390	2	0.0005	0.106 ± 0.122	11.690 ± 0.286	529.14 ± 12.44
8.2	rim	1855	2	0.0009	0.379 ± 0.244	13.163 ± 0.333	472.03 ± 11.53
9.1	rim	1624	1	0.0004	0.358 ± 0.197	13.626 ± 0.352	456.55 ± 11.40
9.2	rim	1019	0	0.0001	0.373 ± 0.223	13.254 ± 0.411	468.91 ± 14.05
10.1	rim	2302	2	0.0007	0.057 ± 0.205	12.784 ± 0.347	485.52 ± 12.70
10.2	core	266	139	0.5230	2.333 ± 0.581	5.993 ± 0.287	994.74 ± 44.24
11.1	met grain	2960	3	0.0010	0.665 ± 0.212	14.040 ± 0.353	443.53 ± 10.80
12.1	rim	1953	2	0.0009	0.452 ± 0.231	13.382 ± 0.342	464.57 ± 11.46
13.1	met grain	1880	0	0.0002	0.417 ± 0.261	13.343 ± 0.384	465.89 ± 12.96
13.2	met grain	884	1	0.0016	5.972 ± 0.763	15.851 ± 0.544	394.37 ± 13.15
14.1	met grain	3740	3	0.0009	0.649 ± 0.349	12.649 ± 1.203	490.48 ± 45.06
14.2	met grain	14314	17	0.0012	−0.016 ± −0.108	11.011 ± 0.165	560.40 ± 8.04
15.1	rim	2131	2	0.0009	0.355 ± 0.293	13.321 ± 0.330	466.64 ± 11.16
15.2	rim	2857	2	0.0008	0.185 ± 0.129	12.818 ± 0.214	484.26 ± 7.78
15.3	core	490	220	0.4491	2.412 ± 0.264	5.792 ± 0.200	1026.67 ± 32.91
16.1	rim	1841	1	0.0006	0.210 ± 0.196	13.518 ± 0.357	460.07 ± 11.73
17.1	rim	4	0	0.0064	8.743 ± 5.550	11.703 ± 3.067	528.57 ± 134.37
18.1	core	98	77	0.7932	2.476 ± 0.925	6.133 ± 0.416	973.72 ± 61.63
19.1	core	244	187	0.7670	4.216 ± 0.451	5.044 ± 0.209	1166.05 ± 44.40
20.1	core	400	145	0.3627	3.006 ± 0.330	5.682 ± 0.185	1044.97 ± 31.47
					Mean age = 472.2 ± 5.8 Ma (MSWD = 1.4, probability of fit = 0.16, n = 16)		
Nigg Bay Migmatite (NBM) [GPS: NJ97030515]							
1.1	core	534	262	0.4916	2.135 ± 0.263	5.650 ± 0.187	1050.44 ± 32.14
1.2	core	2614	525	0.2009	1.813 ± 0.240	11.095 ± 0.248	556.33 ± 11.95
2.1	rim	589	4	0.0065	0.030 ± 0.360	13.294 ± 0.492	467.55 ± 16.73
2.2	rim	552	3	0.0059	0.697 ± 0.358	13.290 ± 0.385	467.67 ± 13.09
2.3	core	141	77	0.5432	2.541 ± 0.459	5.819 ± 0.302	1022.33 ± 49.32
3.1	mixed	79	125	0.1691	2.362 ± 0.251	7.591 ± 0.343	797.78 ± 34.02
4.1	mixed	1052	10	0.0095	0.492 ± 0.229	11.922 ± 0.415	519.23 ± 17.40
5.1	rim	782	4	0.0056	0.291 ± 0.268	13.244 ± 0.395	469.24 ± 13.50
5.2	rim	1108	312	0.2815	26.675 ± 0.981	13.266 ± 0.339	468.49 ± 11.55
6.1	rim	988	12	0.0118	0.691 ± 0.315	13.447 ± 0.442	462.40 ± 14.68
6.2	core	443	394	0.8902	6.319 ± 0.269	3.376 ± 0.086	1672.44 ± 37.57

Table 1 (*continued*)

Nigg Bay Migmatite (NBM) [GPS: NJ97030515]							
7.1	rim	827	5	0.0062	0.015 ± 0.353	12.82 ± 0.3945	484.00 ± 14.35
8.1	rim	630	2	0.0036	0.108 ± 0.423	12.457 ± 0.452	497.77 ± 17.41
9.1	rim	1274	5	0.0041	0.398 ± 0.305	13.109 ± 0.384	473.90 ± 13.41
10.1	mixed	773	173	0.2242	2.647 ± 0.341	7.431 ± 0.281	813.91 ± 29.03
11.1	mixed	690	206	0.2987	4.302 ± 0.251	4.470 ± 0.117	1301.39 ± 30.83
12.1	rim	1077	46	0.0426	0.932 ± 0.266	13.679 ± 0.503	454.83 ± 16.16
12.2	core	343	121	0.3529	15.532 ± 1.734	2.721 ± 0.108	2017.94 ± 69.13
13.1	mixed	284	17	0.0589	14.128 ± 0.762	4.740 ± 0.258	1234.04 ± 61.48
14.1	rim	2161	5	0.0021	−0.320 ± −0.210	13.873 ± 0.375	448.68 ± 11.73
15.1	rim	1019	5	0.0051	0.118 ± 0.394	13.200 ± 0.359	470.74 ± 12.34
16.1	rim	1646	4	0.0024	−0.149 ± −0.189	12.919 ± 0.309	480.61 ± 11.09
17.1	rim	1040	6	0.0054	−0.115 ± −0.299	13.738 ± 0.458	452.96 ± 14.61
18.1	mixed	530	94	0.1766	2.425 ± 0.313	5.548 ± 0.181	1068.23 ± 32.21
19.1	rim	845	5	0.0058	0.382 ± 0.372	12.895 ± 0.403	481.46 ± 14.51
20.1	mixed	775	262	0.3379	2.156 ± 0.351	7.042 ± 0.230	855.97 ± 26.19
21.1	rim	1163	5	0.0039	0.184 ± 0.276	13.380 ± 0.369	464.65 ± 12.38
22.1	rim	2341	14	0.0061	0.230 ± 0.221	13.124 ± 0.280	473.38 ± 9.73
23.1	rim	1429	5	0.0033	−0.038 ± −0.227	12.879 ± 0.289	482.05 ± 10.43
23.2	core	330	163	0.4930	13.844 ± 0.599	2.570 ± 0.066	2118.57 ± 46.48
24.1	rim	1553	5	0.0032	0.333 ± 0.177	13.003 ± 0.311	477.62 ± 11.00
				Mean age = 471.2 ± 5.9 Ma (MSWD = 0.76, probability of fit = 0.74, n = 18)			

Here we have recalculated the recently published $^{40}Ar/^{39}Ar$ ages of Viete et al. (2011a) using the revised (U/Pb system-calibrated) values for the ^{40}K decay constants of Renne et al. (2011). The recalculated $^{40}Ar/^{39}Ar$ ages (here termed 'U/Pb-calibrated ages') are the most appropriate for close geological comparison with U/Pb constraints for metamorphism, including the new SHRIMP ages presented above. Details of the $^{40}Ar/^{39}Ar$ age recalculation method used are provided in the Appendix A supplementary file.

3.2.1. Results of the $^{40}Ar/^{39}Ar$ work of Viete et al. (2011a)

Details on the methodology and results of the muscovite and biotite $^{40}Ar/^{39}Ar$ analyses of Viete et al. (2011a) are given in their manuscript and associated online supplementary file. Viete et al. (2011a) focussed on variation in the nature of Barrovian $^{40}Ar/^{39}Ar$ step-heating spectra with increasing metamorphic grade — i.e. the transition from mixed $^{40}Ar/^{39}Ar$ step-heating spectra (containing detrital components) in the chlorite zone and low-grade parts of the biotite zone to Grampian-age plateau spectra for higher-grade parts of the sequence. The work explored implications of the position of this transition from mixed to plateau $^{40}Ar/^{39}Ar$ step-heating spectra for the duration of Barrovian heating in the biotite zone. The exact nature of the plateau ages was not a primary concern of Viete et al. (2011a), and thus high-resolution figures for the plateau $^{40}Ar/^{39}Ar$ step-heating spectra they obtained were not provided in their manuscript. Fig. 5 provides a higher resolution summary of the 14 $^{40}Ar/^{39}Ar$ step-heating spectra than that given by Viete et al. (2011a). Table 2 also contains details from the work.

The anomalous Silurian $^{40}Ar/^{39}Ar$ step-heating age for DV05-105 (Fig. 5, Table 2) was interpreted by Viete et al. (2011a) to record isotopic resetting in the vicinity of a post-Grampian granite. Close inspection of all Ordovician plateau white mica $^{40}Ar/^{39}Ar$ step-heating ages of Table 2 reveals a systematic increase in Barrovian $^{40}Ar/^{39}Ar$ ages with increasing metamorphic grade. This is demonstrated by a 462.2–460.6–459.4–455.6–459.1–457.6 Ma sequence of white mica $^{40}Ar/^{39}Ar$ ages, with increasing metamorphic grade across the Glen Esk transect, and a 462.5–461.9–460.0–457.3–460.6–459.6 Ma sequence of white mica $^{40}Ar/^{39}Ar$ ages, with increasing metamorphic grade across the Stonehaven transect (Table 2). Systematic variation with metamorphic grade in the $^{40}Ar/^{39}Ar$ ages of Viete et al. (2011a) cannot be associated with any unaccounted for systematic variation in irradiation effects, as the samples were positioned within the can sent to the nuclear reactor (for conversion of ^{39}K to ^{39}Ar) in an arbitrary order (see online supplementary file of Viete et al., 2011a for irradiation details). Furthermore, uncertainties on the isotopic measurement of the irradiation standards are extremely unlikely to produce such consistent, systematic variation in $^{40}Ar/^{39}Ar$ ages for both the Glen Esk and Stonehaven transects. The 2σ uncertainties on isotopic measurement of the unknowns are not able to absorb this variation in $^{40}Ar/^{39}Ar$ ages with increasing metamorphic grade across the Barrovian metamorphic series — e.g. compare the 457.6 ± 1.1 Ma and 459.6 ± 1.0 Ma $^{40}Ar/^{39}Ar$ ages from the highest-grade units (for the Glen Esk and Stonehaven transects, respectively) to the 462.2 ± 1.2 Ma and 462.5 ± 1.2 Ma $^{40}Ar/^{39}Ar$ ages from the lowest-grade units (for the Glen Esk and Stonehaven transects, respectively) (Table 2). Therefore, systematic variation in white mica $^{40}Ar/^{39}Ar$ ages of Viete et al. (2011a) with varying metamorphic grade, from older for the lower-grade zones to younger for the higher-grade zones of the Barrovian metamorphic series, is considered to represent a primary geological feature of the Barrovian metamorphic series.

3.2.2. Results of the U/Pb calibration

Calibration of $^{40}Ar/^{39}Ar$ ages to the U/Pb system (using the method of Renne et al., 2010) was found to increase individual $^{40}Ar/^{39}Ar$ plateau ages by around 0.62%. This equates to a difference of ~3 Myr between Grampian cooling ages determined using the ^{40}K decay constant of Steiger and Jäger (1977) and those determined using the values of Renne et al. (2011) (Table 2). Calibrated $^{40}Ar/^{39}Ar$ ages for the Barrovian metamorphic series are summarised in Table 2. These are the ages most appropriate for comparison with the new peak-metamorphic U/Pb SHRIMP ages for Barrovian metamorphism.

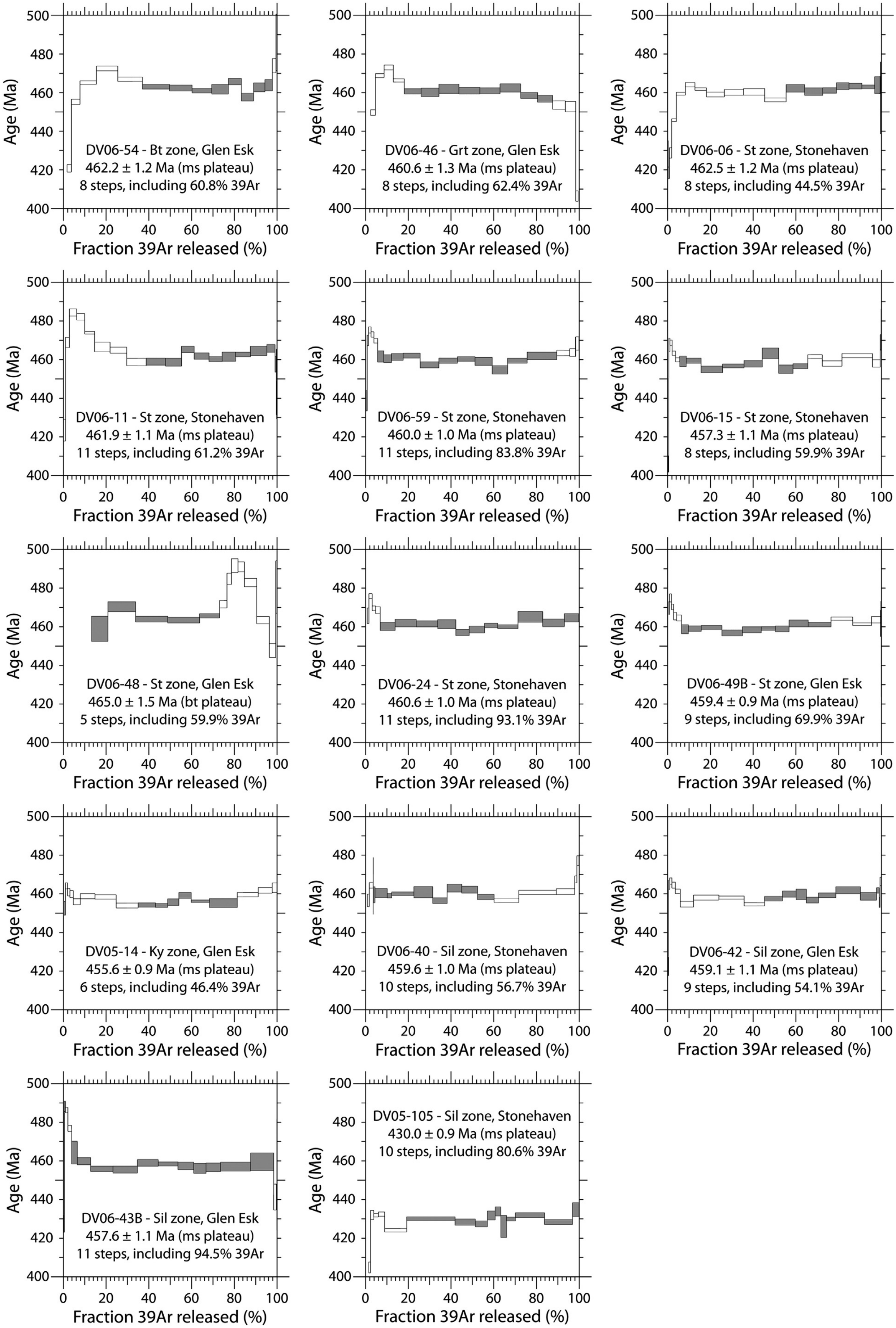
Age (Ma)
Fraction 39Ar released (%)
DV06-54 - Bt zone, Glen Esk
462.2 ± 1.2 Ma (ms plateau)
8 steps, including 60.8% 39Ar
DV06-46 - Grt zone, Glen Esk
460.6 ± 1.3 Ma (ms plateau)
8 steps, including 62.4% 39Ar
DV06-06 - St zone, Stonehaven
462.5 ± 1.2 Ma (ms plateau)
8 steps, including 44.5% 39Ar
DV06-11 - St zone, Stonehaven
461.9 ± 1.1 Ma (ms plateau)
11 steps, including 61.2% 39Ar
DV06-59 - St zone, Stonehaven
460.0 ± 1.0 Ma (ms plateau)
11 steps, including 83.8% 39Ar
DV06-15 - St zone, Stonehaven
457.3 ± 1.1 Ma (ms plateau)
8 steps, including 59.9% 39Ar
DV06-48 - St zone, Glen Esk
465.0 ± 1.5 Ma (bt plateau)
5 steps, including 59.9% 39Ar
DV06-24 - St zone, Stonehaven
460.6 ± 1.0 Ma (ms plateau)
11 steps, including 93.1% 39Ar
DV06-49B - St zone, Glen Esk
459.4 ± 0.9 Ma (ms plateau)
9 steps, including 69.9% 39Ar
DV05-14 - Ky zone, Glen Esk
455.6 ± 0.9 Ma (ms plateau)
6 steps, including 46.4% 39Ar
DV06-40 - Sil zone, Stonehaven
459.6 ± 1.0 Ma (ms plateau)
10 steps, including 56.7% 39Ar
DV06-42 - Sil zone, Glen Esk
459.1 ± 1.1 Ma (ms plateau)
9 steps, including 54.1% 39Ar
DV06-43B - Sil zone, Glen Esk
457.6 ± 1.1 Ma (ms plateau)
11 steps, including 94.5% 39Ar
DV05-105 - Sil zone, Stonehaven
430.0 ± 0.9 Ma (ms plateau)
10 steps, including 80.6% 39Ar

Table 2
$^{40}Ar/^{39}Ar$ ages for units of the Barrovian metamorphic series (from Viete et al., 2011a). Fusion ages and plateau ages are calculated using the ^{40}K decay constants recommended by Steiger and Jäger (1977). U/Pb-calibrated ages are calculated from the plateau ages, but using the revised K decay constants of Renne et al. (2011) (see Section 3.2 and the Appendix A supplementary file for details). Mineral abbreviations follow the recommendations of Kretz (1983). All ages are muscovite ages except that for DV06-48, which is a biotite age.

Sample	Mineral zone	GPS locality	Grain size (μm)	Fusion age (Ma)	±[a]2σ (Ma)	Plateau age (Ma)	±X/Y/Z[b]2σ (Ma)	U/Pb-calibrated age (Ma)	±[c]2σ (Ma)
DV06-45[#]	Chl zone	[NO58627331]	120–180	787.3	7.4	n/a			
DV06-54[#]	Bt zone	[NO58307364]	120–180	461.4	3.5	462.2	1.2/6.8/11.0	465.1	7.4
DV06-46[#]	Grt zone	[NO58037416]	120–180	457.2	3.9	460.6	1.3/7.7/11.6	463.5	8.2
DV06-48[#]	St zone	[NO57947521]	250–420	451.8	7.0	465.0	1.5/6.0/10.6	467.9	6.6
DV06-49B[#]	St zone	[NO56207575]	250–420	460.4	2.8	459.4	0.9/5.5/10.2	462.3	6.2
DV05-14[#]	Ky zone	[NO57387650]	250–420	456.8	3.1	455.6	0.9/9.3/12.6	458.4	9.6
DV06-42[#]	Sil zone	[NO55907847]	250–420	458.2	3.0	459.1	1.1/6.5/10.8	461.9	7.0
DV06-43B[#]	Sil zone	[NO45647802]	250–420	458.2	5.7	457.6	1.1/7.9/11.6	460.4	8.4
DV05-25[#]	Chl zone	[NO89248767]	90–120	568.9	3.8	n/a			
DV06-51[†]	Bt zone	[NO89068778]	120–180	709.3	3.6	n/a			
DV06-01[†]	Bt zone	[NO89088814]	120–180	528.4	3.3	n/a			
DV06-06[†]	St zone	[NO89278901]	120–180	460.1	3.3	462.5	1.2/10.0/13.2	465.3	10.4
DV06-11[†]	St zone	[NO89598968]	120–180	463.9	4.0	461.9	1.1/8.1/11.8	464.8	8.6
DV06-59[†]	St zone	[NO90279098]	250–420	460.3	3.7	460.0	1.0/6.1/10.6	462.8	6.6
DV06-15[†]	St zone	[NO90939237]	250–420	458.7	3.2	457.3	1.1/6.4/10.6	460.2	7.0
DV06-24[†]	St zone	[NO92349436]	250–420	461.2	4.1	460.6	1.0/5.7/10.4	463.5	6.4
DV06-40[†]	Sil zone	[NO94439741]	250–420	459.5	3.4	459.6	1.0/8.7/12.2	462.4	9.2
DV05-105[†]	Sil zone	[NJ95690081]	250–420	427.6	3.5	430.0	0.9/8.5/12.0	n/a	

[a] Uncertainties on integrated total fusion ages include only uncertainty in isotopic measurement of the unknown.
[b] Uncertainties on plateau ages are presented in the form ± X/Y/Z where X includes only the uncertainty in isotopic measurement on the unknown, Y also includes uncertainty in isotopic measurement of the irradiation standard (commonly referred to as the J-factor), and Z also includes uncertainties in the age of the irradiation standard and ^{40}K decay constants, propagated via the method of Karner and Renne (1998).
[c] Uncertainties on U/Pb-calibrated ages include: (1) uncertainties in isotopic measurements; (2) uncertainties in intercalibration of irradiation standard 77–600 hornblende with Fish Canyon sanidine (FCs) via GA1550 biotite, using the intercalibration values of Spell and McDougall (2003), and (3) uncertainties in $^{40}Ar^*/^{40}K$ for FCs and ^{40}K decay constants from Renne et al. (2011), with uncertainties propagated via Equation 7 of Renne et al. (2010).
[#] Sample from the Glen Esk transect.
[†] Sample from the Stonehaven transect.

4. Discussion

4.1. Timing and duration of the Grampian Orogeny, magmatism and metamorphism

Previous work carried out on the timing of metamorphism and magmatic activity relevant to the Barrovian metamorphic event and equivalent Grampian-age metamorphic activity in Scotland and Ireland is summarised in the Appendix B supplementary file. The discussion of timing and duration of tectonothermal activity, below, considers the new ages published in this manuscript, in addition to the existing literature summarised in the Appendix B supplementary file.

As discussed in Section 3.2, ages obtained using a given radioactive decay system may harbour systematic uncertainties that offset those ages relative to age estimates made using a different radioactive decay system. To ensure that we are 'comparing apples with apples', inferences on the timing and duration of the Barrovian metamorphism are made here only on U/Pb ages or on $^{40}Ar/^{39}Ar$ ages that have been calibrated to the U/Pb reference frame. Fig. 6a provides a summary of all published U/Pb ages for the Grampian gabbros and granites, and post-Grampian granites of Scotland. U/Pb or U/Pb-calibrated $^{40}Ar/^{39}Ar$ ages for metamorphic crystallisation or cooling during regional metamorphism in Scotland are also shown in Fig. 6a. Fig. 6b provides the equivalent data compilation for the Grampian igneous and metamorphic rocks of Ireland.

4.1.1. Timing and duration of the Grampian Orogeny

U/Pb zircon secondary ion mass spectrometry (SIMS) performed on zircons extracted from plagiogranite boulders derived from the pre-Grampian Lough Nafooey volcanic arc has yielded ages for magmatism of 487.8 ± 2.3 Ma and 489.9 ± 3.1 Ma (Chew et al., 2007). According to Chew et al. (2007), these ages demonstrate subduction in the region at or before 490 Ma and constrain the timing of Grampian arc–continent collision to after 490 Ma.

The c. 485 Ma white mica $^{40}Ar/^{39}Ar$ age – c. 488 Ma when calibrated to the U/Pb system – for early folding in the lowest-grade rocks of the Barrovian metamorphic series (Viete et al., 2011a) is taken to represent the early stages of Grampian crustal thickening, following arc–continent collision after 490 Ma. Dempster (1985) obtained similar (when uncalibrated) K/Ar whole-rock and muscovite ages of 489 ± 10 Ma and 484 ± 10 Ma, respectively, for the lowest-grade rocks of the Barrovian metamorphic series, which occur adjacent to the Highland Border Ophiolite of the Highland Border Complex. We take Grampian collision (and the Grampian Orogeny) to have commenced by c. 488 Ma (see Fig. 6).

Calculation of the oldest ages for emplacement of intrusive bodies that demonstrably post-date the Grampian Orogeny provides a means of dating the demise of tectonothermal activity responsible for the Barrovian metamorphism and synchronous metamorphism in other parts of Scotland and Ireland. The undeformed Oughterard Granite of Connemara, western Ireland, with a U/Pb xenotime age of 462.5 ± 1.2 Ma (Friedrich et al., 1999a), is the oldest post-tectonic (i.e. post-Grampian) igneous intrusion from Scotland or Ireland. Its age shows that the Grampian Orogeny was completed before 461 Ma. This estimate for the age of demise of Grampian tectonothermal activity is consistent with latest cooling (to temperatures at which significant radiogenic Ar is retained in white mica) of the highest-grade Barrovian metamorphic rocks by c. 461 Ma, as demonstrated in the U/Pb-calibrated $^{40}Ar/^{39}Ar$ plateau ages presented here. We take the Grampian Orogeny to have finished by c. 461 Ma (see Fig. 6).

Fig. 5. Details of the original $^{40}Ar/^{39}Ar$ work of Viete et al. (2011a). Shading demonstrates the steps used in calculation of the plateau age given for each sample. Uncertainty on the plateau age given for each sample considers only the 2σ isotopic uncertainty in the age of the unknown. Expanded uncertainties for each sample are given in Table 2. Mineral abbreviations follow the recommendations of Kretz (1983).

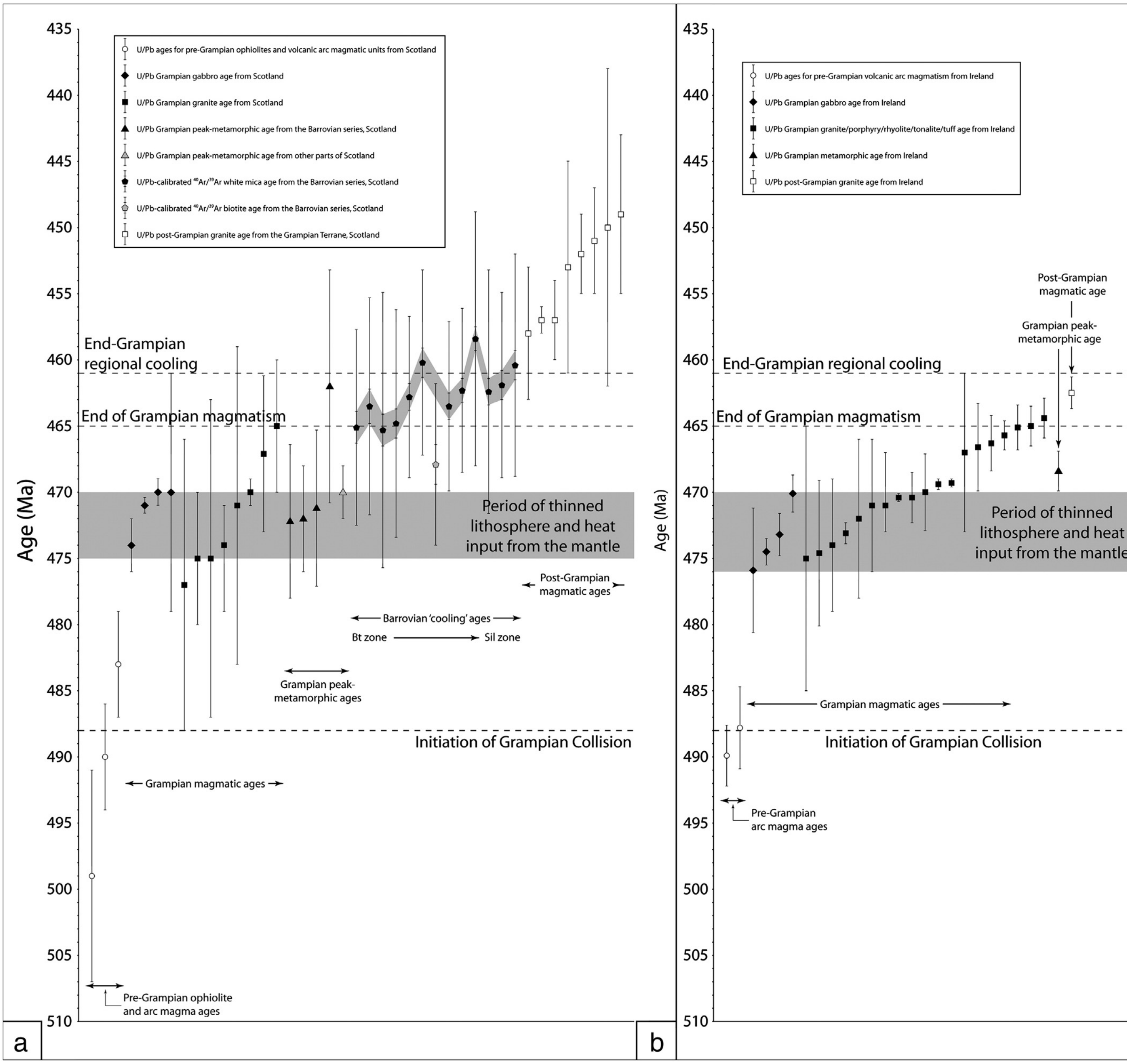

Fig. 6. Summary diagrams for U/Pb and U/Pb-calibrated geochronology from (a) the Grampian Terrane, Scotland, and (b) Grampian units from Ireland. Legend provides an explanation of the symbols used. All ages are shown with 2σ uncertainty bars. Grey shading behind the U/Pb-calibrated $^{40}Ar/^{39}Ar$ ages demonstrates variation in age with metamorphic grade, with range showing only uncertainties on isotopic measurement of the unknown. References for the geochronology are in Section 4.1 of the manuscript or in Table B.1 of the Appendix B supplementary file. Mineral abbreviations follow the recommendations of Kretz (1983).

In summary, the Grampian Orogeny appears to have commenced by c. 488 Ma and finished at c. 461 Ma, suggesting a duration from start to finish on the order of 27 Myr.

4.1.2. Timing and duration of Grampian magmatism

Ages of 474 ± 2 Ma for a phase of the Portsoy Gabbro of Scotland (Oliver et al., 2008), and 474.5 ± 1.0 Ma for the Currywongaun Gabbro of western Ireland (Friedrich et al., 1999a) and 475.9 ± 4.7 for the Scalp Layered Gabbro of Northern Ireland (our reinterpretation of the data of Cooper et al., 2011: see the Appendix B supplementary file for details) are consistent with Grampian mafic magmatism starting at c. 475 Ma. The youngest Grampian gabbro ages – 470 ± 1 Ma for a phase of the Portsoy Gabbro of Scotland (Carty et al., 2012), 470 ± 9 Ma for the Insch Gabbro of Scotland (Dempster et al., 2002) and 470.1 ± 1.4 Ma for the Cashel-Laugh Wheelaun Gabbro (Friedrich et al., 1999a) of western Ireland – are consistent with a c. 470 Ma age for the latest mafic magmatism during the Grampian Orogeny. The latest Grampian granitoid ages from the Tyrone Central Inlier of Ireland (Cooper et al., 2011) and Nigg Bay Granite of Scotland (Appleby et al., 2010) constrain latest felsic magmatism during the Grampian Orogeny at c. 466 to 465 Ma. Inspection of U/Pb magmatic ages from the Grampian regions of Scotland and Ireland are consistent with near identical first appearance of the Grampian granites to that for the Grampian gabbros, but continuation of felsic magmatism for about 5 Myr after cessation of mafic magmatism (see Fig. 6).

U/Pb mafic magma emplacement ages from Scotland and Ireland constrain mantle melting to a period of about 5 to 6 Myr, ending at c. 470 Ma. Felsic magmas (probably related to melting of the crust) persisted for a period of 4 to 5 Myr after cessation of mantle melting and mafic magmatism. Grampian magmatic activity occurred over a total duration of about 10 Myr, within the ~27-Myr Grampian Orogeny (see Fig. 6).

4.1.3. Timing and duration of the Barrovian metamorphism

Metamorphic U/Pb ages for peak metamorphism in the highest-grade parts of the Barrovian metamorphic series, Scotland are in agreement with peak metamorphism within the range 473 to 470 Ma (Fig. 6). These ages include: (1) SIMS U/Pb spot analyses, short depth profiles and long depth profiles performed on Barrovian zircons using the Cameca ims 1270 by Vorhies et al. (2013), which produced a 472 $\pm$ 4 Ma (n =19) estimate for the timing of peak Barrovian metamorphism across the Barrovian metamorphic series, and (2) the new U/Pb SHRIMP ages of 472.2 $\pm$ 5.8 and 471.2 $\pm$ 5.9 Ma presented here. Published Sm/Nd garnet ages and their uncertainties, from Oliver et al. (2000) and Baxter et al. (2002), are consistent with garnet growth during Barrovian metamorphism between about 473 and 465 Ma. It should be noted, however, that intercalibration values and updated estimates for the ^{147}Sm decay constant (for use in rigorous calibration of Sm/Nd ages to the U/Pb system) do not yet exist, and thus Sm/Nd estimates for the timing of metamorphic mineral growth were not included in the geochronological summary of Fig. 6.

U/Pb-calibrated white mica ^{40}Ar/^{39}Ar ages from the Barrovian metamorphic series vary systematically across grade, from c. 465 Ma for the lowest metamorphic grades to c. 461 Ma for the highest (Table 2; Fig. 6). Cooling of the Barrovian metamorphic series appears to have been diachronous across metamorphic grade: white-mica ^{40}Ar/^{39}Ar ages form an array that suggests cooling occurred earlier in rocks of lower metamorphic grade (Fig. 6). Interestingly, cooling in the lowest-grade zones of the Barrovian metamorphic series (i.e. the chlorite, biotite and garnet zones) appears to have occurred at 465 Ma, whilst Barrovian metamorphism in the higher-grade zones (e.g. sillimanite zone) was ongoing. This observation of thermal activity in the high-grade zones outlasting thermal activity in the low-grade zones is reflected in the manner by which the timing of peak metamorphism relative to structural fabric development varies across the Barrovian metamorphic sequence (see Robertson, 1994). That the Barrovian metamorphism was occurring in the deeper, higher-grade zones after the more shallow, lower-grade zones had cooled may have been related to metamorphic heating during rapid exhumation or to the presence of a cold block above the Barrovian metamorphic series during heating (Chinner, 1978; Harte and Hudson, 1979), or the combined thermal influence of both these scenarios.

The Barrovian metamorphism appears to represent a discrete event within the Grampian Orogeny, spanning ~8 Myr (from c. 473 Ma to 465 Ma) within the ~27-Myr orogenic episode (see also Baxter et al., 2002). Despite some c. 488 Ma mica growth within axial planar fabrics associated with the earliest Grampian folding (see Viete et al., 2011a) there is no geochronological evidence from Scotland or Ireland for significant metamorphic heating during the first 15 Myr of the Grampian Orogeny (between c. 488 Ma and 473 Ma). Metamorphism appears to have finished at c. 465 Ma, with 4 Myr remaining during the Grampian Orogeny.

4.2. A discrete Grampian heating event

Previous work concerning the duration of the thermal event recorded in the Barrovian metamorphic series (and related Grampian metamorphic sequences) gives estimates of a few million years (Dewey and Mange, 1999; Oliver et al., 2000; Baxter et al., 2002; Dewey, 2005; Viete et al., 2011a) or less (Ague and Baxter, 2007). This has been shown to be the case for all metamorphic grades preserved in the Barrovian sequence, from the biotite to the sillimanite zone (Ague and Baxter, 2007; Viete et al., 2011a). Moreover, peak Barrovian metamorphism appears to have occurred contemporaneously across all zones of the Barrovian metamorphic series (Baxter et al., 2002; Vorhies et al., 2013). In addition, recent work has shown that the Barrovian metamorphic heating resulted from punctuated accumulation of heat within a localised region of the middle crust, following numerous, short time scale heating events that were closely spaced in time (Ague and Baxter, 2007; Viete et al., 2011b). The Barrovian regional metamorphism was brief and involved episodic (incremental) heating.

The geochronological data (old and new) synthesised above has shown no evidence for metamorphic heating during the first 15 Myr, or last 4 Myr, of the Grampian Orogeny. Thus, the thermal event that produced the Barrovian metamorphic series was, fundamentally, a discrete and transient feature of the Grampian Orogeny. Elsewhere within Scotland and Ireland, Grampian, high-pressure Barrovian-type and blueschist (burial) metamorphism may have been driven by crustal thickening, but carries no evidence to suggest significant thermal input was involved (see Section 2.1).

4.3. The role of thermal relaxation in the Barrovian metamorphism

Recognition of the Barrovian metamorphism as a short lived and discrete thermal event within the Grampian Orogeny significantly weakens arguments for the metamorphism having been driven by 'orogenic' heat originating from processes outlined in the 'thermal relaxation' family of models (see Oxburgh and Turcotte, 1974; Bickle et al., 1975; Richardson and Powell, 1976; England and Thompson, 1984). These thermal relaxation models consider metamorphic heating to result from thermal equilibration of overthickened crust, involving distributed internal heating by radioactive decay, in addition to conduction of heat from the base of the crust. They require long time scales (10s of Myr) to produce significant heat for metamorphism (e.g. Thompson and England, 1984) and are unable to account for the discrete timing of the Barrovian heating episode within the Grampian Orogeny. Furthermore, recognition that the Barrovian metamorphism was first initiated during bimodal magmatism and a period of normal crustal and lithospheric thickness – a 33–44 km thickness of crust is significantly thinner than that expected for a collisional orogen – is not consistent with a first-order requirement of thermal relaxation models: that the crust was overthickened during metamorphic heating.

Episodicity in heating for the Barrovian metamorphism (see Ague and Baxter, 2007; Viete et al., 2011b) is not consistent with metamorphism by a steady accumulation of heat (i.e. by constant-rate heating), as predicted from models of the thermal relaxation family, or models involving localised radioactive heat generation (e.g. Jamieson et al., 1998; Engi et al., 2001). Instead, it requires episodic heat contribution, either by incremental (episodic) advection of heat into a fixed zone of heating or by punctuated heat production relating to some actuating episodic process, or both these mechanisms.

Nonetheless, some recent models for the Barrovian metamorphism have combined short-term heating with a broad regional heating event that resulted from thermal relaxation. Baxter et al. (2002), Ague and Baxter (2007) and Vorhies and Ague (2011) each proposed that the Barrovian metamorphism resulted from overprinting of a significant and widespread metamorphic thermal regime relating to overthickening and thermal relaxation by a brief (<1 Myr), episodic heating event. Moreover, Vorhies and Ague (2011) proposed that some Barrovian-type metamorphic rocks in the SW of the Grampian Terrane did not 'see' the short time scale, episodic thermal overprint and that, in those regions, the Barrovian metamorphism was driven exclusively by crustal thickening and thermal relaxation. The models of Baxter et al. (2002), Ague and Baxter (2007) and Vorhies and Ague (2011) follow the original suggestion of Chinner (1966): that the sillimanite zone of the Barrovian metamorphic series, in the NE of the Grampian Terrane (Fig. 6), defines a contact-metamorphic overprint on a pre-existing and entirely independent metamorphic pattern that developed in response to more enigmatic heat sources.

Recognition of the Barrovian metamorphic series as a distinct episode within the broader Grampian Orogeny, the episodic nature of the metamorphism, and its association with crust of normal thickness precludes significant (first-order) heat contribution from thermal relaxation. The burial metamorphism that produced the Grampian high-pressure Barrovian-type and blueschist metamorphism (see Section 2.1) did not involve any significant heating by thermal equilibration of overthickened crust (cf. thermal relaxation models of Baxter et al., 2002; Vorhies and Ague, 2011). In light of new evidence against the role of thermal relaxation in the Barrovian metamorphism, the proposal of Harte and Hudson (1979) – (1) that the Barrovian sillimanite zone formed within the same thermal regime responsible for the entire Barrovian metamorphic pattern, and (2) that the Barrovian metamorphic series formed by regional contact metamorphism – provides a more reasonable alternative for thermal development of the Barrovian metamorphic series.

4.4. The Barrovian regional metamorphism: a classic example of contact metamorphism

The Barrovian metamorphic series developed in response to a discrete episode of thermal activity within the Grampian Orogeny. It records conditions of thermal disequilibrium (i.e. a localised 'hot' feature) within the crust rather than a return toward crustal-scale thermal equilibrium. The thermal disequilibrium recorded by the Barrovian metamorphism must have been caused by rapid advection of heat into, and/or rapid production of heat within, a focussed region of the middle crust. Episodicity in the Barrovian heating, as recognised by Ague and Baxter (2007) and Viete et al. (2011b), must have been related to incremental heating by recurring episodes of heat advection and/or production.

On the basis of field observations, emplacement of the Grampian gabbros into the middle crust occurred contemporaneously with, and provided heat for, the Barrovian and Buchan metamorphism in Scotland (Chinner, 1966; Fettes, 1970; Pankhurst, 1970; Ashworth, 1975; Harte and Hudson, 1979) and the analogous Grampian-age Barrovian-type sequences in Ireland (Leake, 1989; Yardley et al., 1982; Wellings, 1998; Friedrich et al., 1999b). The c. 475 to c. 470 Ma range in U/Pb ages for the Grampian gabbros of Scotland and Ireland overlaps with the earliest evidence of Barrovian metamorphism, at c. 473 Ma. The metamorphic episode continued for at least ~5 Myr after Grampian mafic magmatism had finished, persisting until Grampian felsic magmatism was complete, at c. 465 Ma. The close temporal and spatial association between bimodal magmatism and a discrete episode of regional metamorphism during the Grampian Orogeny suggests that the Grampian regional metamorphic sequences were produced by advection of heat into the middle crust, from the lower crust and/or mantle. The Grampian regional metamorphism looks to have resulted from large-scale contact metamorphism.

Barrow (1893, 1912) considered the Barrovian metamorphic series to have formed as the result of heating associated with a large-scale network of magmatic bodies – the dominantly pre-Grampian granites of Glen Clova and Glen Esk – and quoted his contemporary Dr Charles Barrois in stating his view that "regional metamorphism and contact metamorphism are much the same thing" (Barrow, 1893, p. 353). The evidence now available for the discrete nature of the Barrovian metamorphic episode within the Grampian Orogeny and its tectonic and magmatic associations suggest that, in the case of his sequence, Barrow (1893) may well have been correct in his assertion that metamorphism was driven by magma emplacement (though wrong in the exact magmas responsible).

Advective heating can play a significant role in the development of high temperature/low-pressure (Buchan-type) regional metamorphism in orogenic settings (see Wickham and Oxburgh, 1985; Sandiford and Powell, 1986). In particular, contact metamorphism as the result of large-scale magmatism, driven by thinning of the lithosphere (see McKenzie and Bickle, 1988), has been associated with development of Buchan-type regional contact metamorphic sequences (Barton and Hanson, 1989; Collins, 2002; De Yoreo et al., 1991). This work on the classic example of Barrovian-type metamorphism has shown that Barrovian-type regional metamorphic sequences may develop from regional-scale contact metamorphism, in an analogous fashion to Buchan-type regional metamorphic sequences. This represents a significant departure from the popular view of Barrovian-type orogenic metamorphism simply as a consequence of thermal equilibration of the crust following tectonic overthickening.

5. Conclusions

Peak metamorphism in the highest-grade rocks of the Barrovian metamorphic series, Scotland, has been dated by SHRIMP U/Pb ages of 472.2 ± 5.8 Ma and 471.2 ± 5.9 Ma for new zircon grown during Barrovian migmatisation. New U/Pb-calibrated $^{40}Ar/^{39}Ar$ ages from the Barrovian metamorphic series vary systematically with increasing metamorphic grade, from c. 465 Ma in the biotite zone to c. 461 Ma in the sillimanite zone, suggesting that cooling was diachronous across the sequence. Analysis of the new ages, within the context of existing geochronological, geochemical and field data, has revealed the following:

1. The Barrovian metamorphism appears to have occurred during a discrete phase of tectonothermal activity spanning some 8 Myr within the ~27-Myr Grampian Orogeny;
2. There is no evidence for significant Barrovian metamorphic heating prior to c. 473 Ma, or prior to first evidence of large-scale, Grampian-age bimodal magmatism, with which it shares an intimate spatial association in its highest grades;
3. Mafic magmas emplaced during the bimodal magmatism with which the Barrovian metamorphism was associated were produced by melting below a crust of thickness 33–44 km and a lithospheric section of thickness $\leq$70 km, precluding development of the Barrovian metamorphism by overthickening of the crust during orogenesis;
4. The Barrovian metamorphism developed by localised, episodic heating.

The above findings are consistent with no major contribution toward the Barrovian metamorphism from heat sources relating to thermal relaxation of overthickened crust during the Grampian Orogeny. Instead, the Barrovian metamorphic series appears to record the short-lived and transient thermal anomaly that developed in the vicinity of a narrow zone of episodic heat advection and/or production in the middle crust during the Grampian Orogeny. It appears likely that significant heat toward this localised and transient thermal anomaly was introduced by advection from the mantle and lower crust during an extended (~10 Myr) period of Grampian magmatism. Our findings are consistent with the suggestion of Barrow (1893) and later Harte and Hudson (1979): that the Barrovian metamorphism – the classic example of orogenic regional metamorphism – formed as the result of contact metamorphism.

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Supplementary data. (Appendix A and B)

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