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Caledonian hot zone magmatism in the "Newer Granites": insight from the Cluanie and Clunes plutons, Northern Scottish Highlands

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Scottish "Newer" Granites record the evolution of the Caledonides resulting from Iapetus subduction and slab breakoff during the Silurian-Devonian Scandian Orogeny, but relationships between geodynamics, petrogenesis and emplacement are incomplete. Laser ablation U-Pb results from magmatic zircons at the Cluanie Pluton (Northern Highlands) identify clusters of concordant Silurian data points. A cluster with a weighted mean $^{206}Pb/^{238}U$ age of 431.6 ± 1.3 Ma (2σ confidence interval, n = 6) records emplacement whilst older points (clustered at 441.8 ± 2.3 Ma, n = 9) record deep crustal hot zone magmatism prior to ascent. The Cluanie Pluton, and its neighbour the ~428 Ma Clunes tonalite, have adakite-like high Na, Sr/Y, La/Yb and low Mg, Ni and Cr characteristics, and lack mafic facies common in other "Newer Granites". These geochemical signatures indicate the tapping of batches of homogenised, evolved magma from the deeper crust. The emplacement age of the Cluanie Pluton confirms volumetrically modest subduction-related magmatism occurred beneath the Northern Highlands before slab breakoff, probably as a result of crustal

thickening during the ~450 Ma Grampian 2 event. Extensive new in-situ geochemical-geochronological studies for this terrane may further substantiate the deep crustal hot zone model and the association between Caledonian magmatism and potentially metallogenesis. The term “Newer Granites” is outdated as it ignores the demonstrated relationships between magmatism, Scandian orogenesis and slab breakoff. Hence, “Caledonian intrusions” would be a more appropriate generic term to cover those bodies related to either Iapetus subduction or to slab breakoff.

Short title: Northern Highlands Newer Granites
Keywords: Adakite, Caledonian, Geochemistry, Geochronology, Scotland

Plutonism during the Caledonian Orogeny and its aftermath in Scotland and the wider British Isles includes the ~426-390 Ma “Newer Granites” (*sensu* Read 1961), commonly proposed to have resulted from Iapetus slab breakoff (e.g., Atherton & Ghani 2002 and subsequent authors). However, partial melting of lower crust and upper mantle during the orogenic cycle can be ascribed to a range of other geodynamic processes such as subduction, crustal thickening, slab rollback, lithospheric delamination, and sub-lithospheric convection (England & Thompson 1984; von Blanckenburg & Davies 1995; Keskin 2003; Kaislaniemi *et al.* 2014). Some of the Newer Granites of the Scottish Caledonides remain to be convincingly assigned an emplacement age or subjected to detailed geochemical characterisation. As such, their geodynamic associations, petrogenesis, and even metallogenic potential are unclear. Internationally, various granitoid petrogenetic concepts have grown in prominence in the past three decades. These include the construction of plutons in temporally distinct phases (Miller *et al.* 2007), the association of magmatic ‘flare-ups’ with geodynamic events (Ardila *et al.* 2019), and the role of deep crustal, near-solidus magma processing over protracted periods, such as the deep crustal hot zone of Annen *et al.* (2005), melting-assimilation-hybridisation – MASH – processes of Hildreth & Moorbath (1988); and transcrustal magmatic systems of Cashman *et al.* (2017). However, these concepts are only just beginning to be more widely applied to Caledonian magmatism in the British Isles (e.g., Archibald *et al.* 2021, 2022; Fritschle *et al.* 2018; Miles & Woodcock 2018; Miles *et al.* 2014; Woodcock *et al.* 2019) and have yet to be consistently applied to the "Newer Granite" plutons, especially in the Grampian and Northern Highlands of Scotland (e.g., Bruand *et al.* 2014; Clemens *et al.* 2009; Oliver *et al.* 2008).

This study presents new data from the Cluanie and Clunes plutons of the Northern Highlands, firstly to fill existing knowledge gaps about "Newer Granite" timing and petrogenesis in this part of the Caledonides. Using zircon U-Pb laser ablation inductively coupled mass spectrometry (LA-ICP-MS) and whole rock elemental and Sr-Nd-Hf isotopic geochemistry, we present evidence for a) the age of Cluanie's emplacement and its association with Iapetus subduction, b) operation of a deep crustal hot zone during the Caledonian Orogeny, and c) the petrogenesis of the two plutons. We therefore propose that higher resolution of the magmatic record of Iapetus subduction and its transition to collision and slab breakoff more widely across the British Isles will provide clarity over the geological relevance and consequences for critical metal enrichment of the Caledonian intrusions (c.f. Richards 2015).

Regional geology

The Cluanie and Clunes plutons are located north of the Great Glen in the Northern Highlands (Fig. 1a-d). Both bodies were emplaced within psammites and semi-pelites of the Loch Ness Supergroup, of the Northern Highland Terrane (Figure 1b; stratigraphy after Krabbendam et al., 2021). The Northern Highland Terrane is bound by the Moine Thrust to the west and is dominated by Neoproterozoic 'Moine' metasedimentary succession and the largely concordant Late Proterozoic West Highland Granite Gneiss bodies, all of which sit on a Meso-Paleoproterozoic gneissose basement of Laurentian and proposed Baltican affinity (Strachan *et al.* 2020). The Moine succession underlies large tracts of northern Scotland and comprises the recently assigned Wester Ross and Loch Ness Supergroups (Krabbendam *et al.* 2021; Strachan *et al.* 2002; 2010 and references therein). All record evidence of poly-metamorphism, typically up to amphibolite facies. The Wester Ross Supergroup records Renlandian events (960 to 920 Ma; Bird *et al.* 2018) and the Loch Ness records Knoydartian events (820 Ma to 725 Ma; Rogers *et al.* 1998; Vance *et al.* 1998; Tanner and Evans, 2003; Cutts *et al.* 2009a, 2010, 2015). Both supergroups record Caledonian (Grampian and Scandian) metamorphism (Bird *et al.* 2013; Johnson *et al.* 2017).

The Palaeozoic Caledonian Orogeny in Scotland resulted from closure of the Iapetus Ocean primarily between Laurentia, Baltica, Avalonia, as well as several arc terranes (e.g., van Staal *et al.* 2021). Ordovician arc-continent and proposed microcontinent-continent collisions first resulted in the Grampian Orogeny(ies) (~488-450 Ma; Bird *et al.* 2013; Johnson *et al.* 2017; Dunk *et al.* 2020; Walker *et al.* 2020) (Fig. 2). Oblique continent-continent collision between Baltica and Laurentia is recorded north of the Great Glen Fault Zone in the Northern Highlands as the Scandian Orogeny (~437-415 Ma; Strachan *et al.* 2020) (Fig. 2). The Scandian Orogeny partly overlaps with Avalonia-arc-Laurentia collision which mostly affected southern Scotland and England (see Soper *et al.* 1992 and Dewey & Strachan 2003, for discussion). Widespread magmatism occurred across Scotland from ~426-390 Ma (Oliver *et al.* 2008), with intrusive bodies frequently referred to as the "Newer Granite" Suite (Read 1961).

The "Newer Granites"

These bodies post-date the Grampian orogeny(ies), overlapping with Iapetus subduction, Scandian and Acadian deformation. Many such bodies are widely called the "Newer Granites" despite their broad spectrum of compositions, ages, and potential geodynamic triggers for melting and emplacement, as challenged in the discussion below. Various "Newer Granites" pre-date a critical geodynamic event at ~428 Ma in Scotland, marked by uplift in the Grampian Highlands, and shortly followed by deposition of the Lower Old Red Sandstone and the majority of Caledonian granitoid emplacement and concurrent volcanic activity (Conliffe *et al.* 2010, Table 1, Fig. 2). It is proposed that event was the breakoff of the Iapetus slab at ~428 Ma, prior to termination of the Baltica-Laurentia collision (e.g., Atherton & Ghani 2002; Neilson *et al.* 2009; Conliffe *et al.* 2010; Strachan *et al.* 2020). This style of continental collision more widely termed the "Turkic-type orogen" (Şengör & Okuroğullari 1991) is of potential relevance to Scotland, because the switch from subduction and continental arc magmatism to slab breakoff and rapid uplift is widely associated with critical metal mineralisation (e.g., Richards 2009, 2015).

In this paper's primary focus, the Northern Highlands of Scotland, there are a few magmatic events recorded between the end of magmatism associated with the Grampian Orogeny and the emplacement of the bulk of the "Newer Granites" (Fig. 2, Table 1). The Glen Dessary syenite, ascribed to continental arc magmatism on the Laurentian margin, was emplaced at ~448 Ma (Fowler 1992; Goodenough *et al.* 2011). Strachan *et al.* (2020) dated granitoid sheets associated with the Naver Thrust in Sutherland to ~432 Ma. The next known magmatic events on the mainland include the Assynt Alkaline Suite, also ascribed to supra-subduction processes (~431-429 Ma; Goodenough *et al.* 2011; Thompson & Fowler 1986; Thirlwall & Burnard 1990; Table 1, Fig. 1a). Explanations for such limited magmatic output, compared to the voluminous post breakoff episode, have included periods of highly oblique or flat slab subduction (Oliver *et al.* 2008; Dewey *et al.* 2015), or further collisional events (Grampian 2) suppressing magmatic activity (Bird *et al.* 2013). There are also recent data from the Brae, Graven, Muckle Roe and Ronas Hill bodies of Shetland and the Orkney Granite Complex, which indicate apparent supra-subduction granitoid magmatism from ~460-428 Ma, but the relationship of these bodies to the mainland's limited emplacement record is uncertain (Lancaster *et al.* 2017; Lundmark *et al.* 2018; Fig. 2).

Thereafter, the bulk of the “Newer Granites” in the Northern Highlands apparently crystallised from ~426-418 Ma (Oliver *et al.* 2008) (Fig. 1a, 2; Table 1). Microdiorite and appinite minor intrusions and stocks are widespread (Smith 1979). The plutons themselves often contain felsic, intermediate, and mafic, even ultramafic facies and decameter-scale mafic to intermediate magmatic enclaves. Many such bodies share a high Ba-Sr affinity, with a petrogenetic relationship proposed between different facies of high Ba-Sr granitoids, indicating a mantle-derived origin for this suite (e.g., Fowler *et al.* 2001; 2008). The notable exceptions to this pattern are the more homogeneous and felsic Cluanie pluton (Neill & Stephens, 2009) and the Clunes tonalite (Stewart *et al.* 2001). Whole rock elemental, radiogenic and stable isotope geochemistry does largely support the “Newer Granites” being derived from melts of subduction-modified mantle, plus varying proportions of fractional crystallisation and crustal contamination (Fowler *et al.* 2008, Neilson *et al.* 2009). However, an almost exclusive role for crustal melting has previously been proposed for some felsic plutons (Halliday & Stephens 1984; Harmon *et al.* 1984; Neill & Stephens 2009).

The Cluanie and Clunes Plutons

The Cluanie Pluton

The Cluanie Pluton (Leedal, 1952) is a 20 km^2 un-deformed magmatic body between Glen Moriston and Glen Shiel (Fig. 1b, c). The Cluanie Pluton intrudes psammites and semi-pelites of the Loch Eil Group of the Loch Ness Supergroup. The pluton lies at the intersection of mapped strike-slip faults striking NW-SE and NE-SW near the southern termination of the Strathglass Fault, parallel to the Great Glen Fault (Peacock *et al.* 1992, Fig. 1b). The intersection of faults has been proposed as a low-strain zone permitting emplacement of the Cluanie magmas, though that model may require as yet unidentified right-lateral motion on the supposedly left-lateral Strathglass fault (Neill & Stephens 2009), which are part of a major orogen parallel left-lateral fault system at 425-410 Ma (Jacques & Reavy 1994).

The Cluanie Pluton is comprised of oscillatory-zoned plagioclase (~60 %; An_{15-30}), alkali feldspar (~15 %, usually megacrystic); quartz (~15 %), hornblende (5-10 %), and biotite (0-5 %) (Peacock *et al.* 1992). Accessories include titanite (~1%), apatite, zircon, allanite, and ilmenite. Alkali feldspar megacrysts frequently reach 1-3 cm across, with the groundmass typically grain size typically 3-4 mm. According to the quartz-alkali feldspar-plagioclase classification of Streckeisen (1976), the Cluanie Pluton can be described as a granodiorite. However, on an alkali ternary plot (see Neill & Stephens, 2009) the pluton plots in the tonalite-trondhjemite-granodiorite field. Thus, the pluton has been described as a porphyritic granodiorite with trondhjemitic affinity (Neill & Stephens 2009).

The intrusion is penecontemporaneous with a suite of porphyritic minor intrusions (Smith 1979), some of which are partially mingled with the pluton (Neill & Stephens 2009). These "porphyrites" are plagioclase-phyric micro-granodiorites consisting of roughly equal proportions of quartz, alkali feldspar and plagioclase (>90 %) plus 1-2 mm-scale biotite and hornblende, with accessory titanite. The pluton is sharply cross-cut by micro-diorite dykes of ~1 mm grain size, <0.5 m across, containing plagioclase, hornblende, quartz, and alkali feldspar (Smith 1979). The porphyrites cluster around the Cluanie pluton whereas the micro-diorites are regionally extensive and not obviously related to the pluton (Smith 1979). Although Cluanie is petrographically similar to many Northern Highlands "Newer Granites", and geochemically belongs to the near-ubiquitous high Ba-Sr class (Tarney & Jones 1994; Neill & Stephens 2009), it has $\varepsilon Nd_i > 0$ (Fowler *et al.* 2008), a high Na_2O/K_2O character (Neill & Stephens 2009), no mafic plutonic lithologies, no association with appinitic or microdioritic minor intrusions (Peacock *et al.* 1992; Neill & Stephens 2009), and no mafic enclaves, except for rare cm-scale amphibole- and titanite-bearing clots and schlieren interpreted as restite (Neill & Stephens 2009). Fowler *et al.* (2008) placed the Cluanie pluton within their mantle-derived models of high Ba-Sr "Newer Granite" petrogenesis. However, based on the lack of a clear association with mafic, mantle-derived parental magmas, Neill & Stephens (2009) argued for the Cluanie pluton to have a geologically young amphibolitic melt source.

A U-Pb zircon isochron intercept of ~417 Ma (no error given; Pidgeon & Aftalion 1978) and a whole rock Rb-Sr age of 425 ± 4 Ma (Brook 1985) are the only available geochronological constraints. Revision of the ^{87}Rb decay constant means the published Rb-Sr age is recalculated to ~433.5 Ma (Nebel *et al.* 2011). This age pre-dates all the Northern Highlands “Newer Granites” (Table 1), calling into question its association with the “Newer Granites” and importantly slab breakoff (~428 Ma) (Figure 2, Table 1). Lastly the emplacement depth of Cluanie has been estimated at ~13-18 km based on Al-in-hornblende geobarometry (Neill & Stephens 2009; see Supplementary Item for additional refinement).

The Clunes Pluton

The ~6 km^2 Clunes Pluton crops out just north of the Great Glen Fault Zone (GGFZ, Fig. 1b, d), with emplacement facilitated by a shear zone utilizing a mechanical boundary between the Glenfinnan and Loch Eil Groups of the Loch Ness Supergroup (Stewart *et al.* 2001). The body is largely tonalitic, with equigranular plagioclase, quartz, hornblende, and biotite, and rare patches of more granitic, granodioritic or dioritic compositions on its margins (Fig. 1d). The Clunes pluton is cut by as yet undated felsic sheets thought to be part of the Glen Garry Vein Complex (Fettes & MacDonald 1978). No geochemical analyses have been published. Zircon chemical abrasion isotope dilution thermal ionisation mass spectrometry (CA-ID-TIMS) dating of a sample close to the western margin of the pluton gave an apparent emplacement age of 427.8 ± 1.9 Ma (2σ, n = 4, weighted mean of ^{207}Pb/^{206}Pb ages) (Stewart *et al.* 2001). Two discordant grains had similar ^{207}Pb/^{206}Pb ages and upper intercept ages similar to these four grains. The data indicate a maximum emplacement age of ~428 Ma. This age, and the pluton’s left-lateral swing of magmatic fabric at its NE margin is regionally important, as it pins sinistral movement on the GGFZ to have been active by ~428 Ma (Stewart *et al.* 2001). Confirmation of this interpretation comes from ~427-430 Ma U-Pb zircon and Re-Os molybdenite dates from the Loch Shin and Grudie plutons (Holdsworth *et al*., 2015; Table 1). These plutons immediately southwest of the NW-SE Loch Shin-Strath Fleet fault system north of the GGFZ are interpreted to have intruded along these faults in a stress regime consistent with sinistral motion on the GGFZ.

Analytical Methods

Several kilogrammes of trondhjemite were collected from the shore of Loch Cluanie at NH 1444 0995. Zircons were separated by traditional crushing and heavy liquid separation at the University of Glasgow and mounted on resin stubs. Each grain was checked for suitable zones for laser analysis and photographed using cathodoluminescence (CL) on a Carl Zeiss Sigma scanning electron microscope at the Imaging, Spectroscopy and Analysis Centre, University of Glasgow. Selected grains were lasered at the University of Glasgow using an Australian Scientific Instruments RESOlution laser operating at 4.5 J and 10 Hz. Spots of 30 μm diameter were ablated for 30 seconds each. Ablated material was transported in Ar and analysed on a Thermo iCAP-RQ single collector mass spectrometer. Data were reduced in Iolite v.3 (Paton *et al.* 2011). Results were standardised to NIST-610 and checked against Plešovice zircon, producing a mean ^{208}Pb/^{236}U age of 336.9 ± 0.4 Ma (2σ, n = 57), uncorrected versus a published value of 337.13 ± 0.37 Ma (Sláma *et al.* 2008). Final data presentation was completed using IsoPlotR (Vermeesch 2018). >250 spots were analysed across >50 grains, with >50 spots displaying 95% concordance or better. Any spots whose 2σ analytical error margins failed to overlap the concordia on a Wetherill diagram were further removed, leaving <40 spots for further investigation.

Whole rock samples from the Cluanie pluton, porphyrites and microdiorites, were analysed for major and trace elements at Cardiff University as per McDonald & Viljoen

(2006). Samples were crushed and powdered using a steel jaw crusher and agate ball mill. Dry powders from loss-on-ignition determination were fused on a propane burner in platinum crucibles with $LiBO_2$ then dissolved in nitric acid. Inductively coupled plasma optical emission spectrometry (ICP-OES) analysis for major elements and Sc was carried out on a JY-Horiba Ultima 2 and trace elements were analysed on a Thermo Elemental X7 ICP-MS. Reference materials JB-1A, BIR-1 and NIM-G were analysed throughout. First relative standard deviations for most major elements during runs of these materials were typically <2.7 % (P_2O_5 = 5.8 %), < 3 % for most trace elements (excepting 5 % for Ni, 4 % for Cu and 8 % for Rb), and <5 % for the REE. Neodymium and hafnium isotope compositions were analysed at the NERC Isotope Geosciences Laboratory, Nottingham. Samples were dissolved using a standard HF-HNO_3 procedure. Hafnium was separated using a single LN-SPEC column procedure following Münker *et al.* (2001). The Hf isotope composition of the samples was analysed using a Thermo Scientific Neptune Plus MC (mass collector)-ICP-MS. Correction for Lu and Yb interference on mass 176 was carried out using reverse-mass-bias correction using empirically predetermined ^{176}Yb/^{173}Yb and ^{176}Lu/^{175}Lu. The analysed samples contained no detectable Lu, and very low Yb, so these corrections are negligible. Analysis of the JMC475 standard gave ^{176}Hf/^{177}Hf = 0.282151 ± 0.000003 (1σ, n = 35) comparable to a preferred value of 0.282160 (Nowell & Parrish 2001). Analyses of BCR-2 gave 0.282873 ± 0.000001 (1σ, n = 3), relative to JMC475 = 0.282160. The LREEs (light REEs) were concentrated using cation exchange columns (Eichrom AG50x8), and Sm and Nd were then separated using LN-SPEC columns. Neodymium was loaded on double-rhenium filament assemblies and analysed in multi-dynamic mode on a Thermo Scientific Triton thermal ionisation mass spectrometer. ^{143}Nd/^{144}Nd is reported normalised to a preferred value of 0.511860 for the La Jolla standard. Measured ^{143}Nd/^{144}Nd ratios for the La Jolla standard were ^{143}Nd/^{144}Nd = 0.511853 ± 0.000008 (1σ, n = 3). An Sr fraction and LREE (light REE) fraction were separated using cation exchange columns (Eichrom AG50x8), and Sm and Nd were then separated using LN-SPEC columns. Sr fractions were loaded onto outgassed single Re filaments using a TaO activator solution and analysed in a Thermo Scientific Triton mass spectrometer in multi-dynamic mode. Data are normalised to ^{86}Sr/^{88}Sr = 0.1194. Analyses of the NBS987 standard gave a value of 0.710253 ± 0.000005 (1σ, n = 9). Sample data are normalised using a preferred value of 0.710250 for this standard. Whole rock samples from a transect across the Clunes tonalite were collected in 2016 (Fig. 2) and analysed for major and trace elements according to the methodology written up in full in Milne (2020). The complete geochemical-geochronological dataset is in the Supplementary Item.

Results

Zircon U-Pb results from Cluanie

Grain images, spot locations, and full results can be found in the Supplementary Item. Many subhedral zircons are stubby, with acute apices and either: i) zoned magmatic cores, with sharp or slightly resorbed boundaries and an outer, oscillatory zoned mantle (e.g., Stub 1 Grains 1-2) or ii) opaque or more complex cores, again with oscillatory mantles (e.g., Stub 1 Grain 21, Stub 3 Grain 3). The only obvious relationship between textures and ages is that those with opaque or complex cores typically returned ^{206}Pb/^{238}U ages from those cores in the region of 540 - 1300 Ma.

On Figures 2a and 2b, 14 spots from apparently magmatic zircon cores form a prominent ~1590-1700 Ma cluster, with a further five clustered around 1450-1475 Ma. These spots may represent inheritance from detrital zircons found in the Loch Ness Supergroup, given that the Glenfinnan Group has equivalent detrital age peaks (Kirkland *et al.* 2008). The older cluster might represent late Laxfordian events in the Hebridean or sub-Northern

Highlands basement, where Laxfordian-aged zircon peaks have now been observed (Strachan *et al.* 2020), though whole rock isotopic results indicate Lewisianoid assimilation is strictly limited. There are several spots on complex zircon cores with $^{206}Pb/^{238}U$ ages ranging from ~540-1300 Ma, the oldest probably inherited from the Moinian rocks (Kirkland *et al.* 2008). One spot at 985 ± 18 Ma may correspond to Renlandian events recorded on the northern Scottish mainland and Shetland (Bird *et al.* 2018; Walker *et al.* 2020) suggesting a deeper interaction with the Wester Ross Supergroup or its basement. Three spots provided ages around ~865 Ma, similar to the ~870 Ma age of protoliths of the West Highland Granite Gneiss which intrudes the Moinian rocks (Friend *et al.* 1997). Single spots at 772 ± 20 and 731 ± 10 Ma could be Knoydartian, based on existing geochronological constraints (Mako, 2019). Results of 625 ± 15, 588 ± 22 and 540 ± 4 Ma overlap with Iapetus rifting, with one indistinguishable from the nearby Carn Chuinneag intrusion (594 ± 11 Ma; U-Pb zircon ion probe; Oliver *et al.* 2008).

The 15 remaining spots are from cores or mantles containing magmatic zoning, with concordia ages from ~430-450 Ma, forming two apparent clusters (Fig. 2c). The weighted mean $^{206}Pb/^{238}U$ age of all 15 spots is 437.9 ± 3.2 Ma (2σ confidence interval with probability cut-off of 0.05). The older cluster has a weighted mean age of 441.8 ± 2.3 Ma (n = 9), the younger cluster 431.6 ± 1.3 Ma (n = 6) (Fig. 2d). Confidence intervals of the two clusters do not overlap, and the younger cluster is indistinguishable from the re-calculated Rb-Sr age of 433.5 ± 4 Ma of Brook (1985). U concentrations in the analysed spots drop from ~1610 to 1050 ppm and Th from ~350 to 200 ppm from the older to the younger cluster (Supplementary Item). These elements behaved compatibly during Cluanie magmatism, implying the younger magmatic zones grew from a more evolved melt. We therefore designate the $^{206}Pb/^{238}U$ weighted mean of 431.6 ± 1.3 Ma (2σ, n = 6) to represent emplacement, coinciding with the early part of the Scandian Orogeny and overlapping with the ages of granitoids associated with the Naver Thrust, and the Assynt Alkaline Suite (Goodenough *et al.* 2011; Strachan *et al.* 2020). The older cluster covers a wider range of $^{206}Pb/^{238}U$ ages, from ~437-450 Ma, versus ~430-435 Ma for the younger, implying the older weighted mean age to be representative of protracted zircon growth over ~13 Ma.

Whole rock geochemistry from Cluanie and Clunes

Major and trace element data are plotted against SiO_2 (Fig. 3). The granodiorites at Cluanie ranges from quartz monzonite to granite on a total alkali-silica plot (Middlemost 1994) (Fig. 3a), whereas the slightly less evolved felsic porphyrites fall in the granodiorite and quartz monzonite fields. The microdiorite dykes range from monzonite to granodiorite. The granodiorites have a uniform composition of 68-72 wt.% SiO_2 and ~1 wt.% MgO. The felsic porphyrites have 66-68 wt.% SiO_2 and 1.2-1.3 wt.% MgO, and the microdiorites have 61-67 wt.% SiO_2 and 2.0-3.8 wt.% MgO. P_2O_5, TiO_2, and many trace elements including Zr, Th, U and the REE display compatible behaviour (Fig. 3b-g). This observation is consistent with fractionation of amphibole, biotite, and various reported accessory minerals such as apatite, zircon, titanite and allanite (Leedal 1952). A chondrite-normalised plot (Fig. 4a) shows the trondhjemites have light REE (LREE)-enriched compositions, with La/Yb_{CN} = 7-19, and slight U-shaped patterns consistent with involvement of MREE-compatible minerals such as zircon, apatite, or amphibole. Moderate-low Ho/Yb_{CN} ratios (<1) in the main granodiorite facies do not clearly indicate a role for heavy REE (HREE)-loving garnet, but such ratios can be tempered by fractionation of the MREE-compatible phases. Primitive mantle-normalised distributions (Fig. 4b) demonstrate the high Ba-Sr nature of these rocks and elevated Rb relative to Th and the LREE. All samples have negative Nb-Ta and Ti anomalies but positive Zr-Hf anomalies. The less-evolved granodiorites and microdiorites have higher overall REE abundances, and these both have less-U-shaped patterns on Figure

4a, with Ho/Yb$_{CN}$ of 1.0-1.2. The microdiorites also contain relatively lower Ba and Sr (Fig. 4b).

As there are few samples for the adjacent Clunes tonalite, meaningful trends on Harker plots cannot be discerned. Samples have SiO_2 concentrations from 60-65 wt.%, with 2-3 wt.% MgO (Figs. 3a-g), ranging from monzonite to quartz monzonite and granodiorite. Clunes' chondrite-normalised REE patterns show La/Yb$_{CN}$ = 14-22, and Ho/Yb$_{CN}$ ~1.2, giving smoothly decreasing HREE abundances rather than the U-shape patterns of the Cluanie granodiorites (Fig. 4c). The major and trace element concentrations of Clunes are very similar to the Cluanie microdiorites (Fig. 4d).

Cluanie and Clunes bear geochemical comparison with modern adakites, a prominent suite of similarly sodic, HREE-depleted igneous rocks, with high La/Yb and Sr/Y and low MgO (Martin *et al.* 2005; Fig. 5a-c). Neill & Stephens (2009) previously noted the affinity of the Cluanie pluton with tonalite-trondhjemite-granodiorite (TTG) suites, which dominate the felsic record of Archaean magmatism (e.g., Johnson *et al.* 2019). Petrographically, however, the Cluanie pluton's ubiquitous alkali feldspars are not associated with TTGs (Martin *et al.* 2005). The origin of adakites is debated, from melting of garnet amphibolite or eclogite in subducting slabs (Defant *et al.* 1992; Drummond *et al.* 1996) or lower crust, to fractionation of garnet or amphibole from mantle-derived precursors (Macpherson *et al.* 2006). Only the lattermost model corresponds to the mantle-derived origin proposed for the Cluanie Pluton by Fowler *et al.* (2008), prompting Neill & Stephens (2009) to explore alternative hypotheses for the apparently homogeneous facies and geochemistry present at Cluanie. On a chondrite-normalised La/Yb vs Yb plot (Fig. 5a), all samples have low La/Yb ratios but do mostly lie within the adakite field, overlapping the island arc field. The two least-evolved microdiorites plot exclusively in the island arc field. On a Sr/Y vs Y plot (Fig. 5b), all bar one sample from Clunes and the two Cluanie microdiorites plot in the adakite field. On Figures 5a-c, the

Cluanie and Clunes plutons are notably more homogeneous and sodic than the other, younger, Northern Highlands "Newer Granites", with more favourable major and trace element similarities to some modern adakites.

Previous Nd-Sr radiogenic isotope analyses (Halliday 1984; Fowler *et al.* 2008) were taken from a single quarry site at Cluanie where zircon inheritance was picked up by Pidgeon & Aftalion (1978). One of our two samples is also from this quarry. Our εNd_i values are +4.0 and +4.2, the highest yet observed in the Northern Highlands granitoids, with $^{87}Sr/^{86}Sr_i$ of 0.7044-0.7048 (Fig. 5d). εHf_i values are +7.2 and +7.5 (Supplementary Item). These data indicate a dominant mantle- or recent mantle-derived component within the pluton. Cluanie has the most depleted mantle-like isotopic signature of all published Northern Highlands Caledonian granitoids, with the exception of the older, more mafic, Glen Dessary body.

Discussion

Magma series and magmatic evolution at Cluanie and Clunes

Before considering the petrogenesis of the plutons, the effect of assimilation of Moine meta-sediments must be considered. Zircon inheritance is evident at Cluanie, though the whole rock radiogenic isotope data show moderate $^{87/86}Sr$ vs high $^{143/144}Nd$, precluding involvement of low Rb/Sr Lewisian (<1; Chamberlain *et al.* 1986) basement, but consistent with Wester Ross or Loch Ness Supergroup involvement (Rb/Sr 1-7; Bird 2011). Fowler *et al.* (2008) modelled a subduction-modified Scottish Caledonian parental mantle source for the Newer Granites, with $\varepsilon Nd_{i(425)}$ of ca +4.5 for Cluanie. Their assimilation-fractional crystallisation (AFC) model, using $\varepsilon Nd_{i(425)}$ ca +2.6 for Cluanie, estimated AFC at ~15%. Our samples, with $\varepsilon Nd_{i(432)} = +4.1$, would require only ca 5% AFC with Fowler *et al.*'s model. INC4 was collected from the same quarry as Fowler *et al.*'s samples, implying localised isotopic heterogeneities in the pluton, highly likely as Glenfinnan Group xenoliths, ghost xenoliths and roof pendants are found close to the quarry site. The sample for U-Pb dating was taken from within a few hundred metres of the pluton's western margin. Overall, though, crustal assimilation may only have had a modest effect on major and trace element concentrations across the wider pluton where the majority of whole rock samples are from.

Given the sharp cross-cutting relationship between the regionally-common microdiorites at Cluanie we assume that the Cluanie granodiorites and microdiorites are genetically unrelated. The latter are geochemically most akin to the Clunes tonalite as described above. Given they post-date the Cluanie pluton, a genetic relationship is possible between the Clunes tonalite and the regional microdiorite suite. Evidence for mingling of felsic porphyrite with the granodiorite at Cluanie was reported by Neill & Stephens (2009), so the felsic porphyrites could be considered as a parental magma which evolved by AFC processes to form the granodiorites, albeit the porphyrites are far evolved from any potential mantle-derived parent. Common major and trace element trends between the felsic porphyrites and the plutonic facies (Figs 3-4) are consistent with this genetic relationship.

The limited geochemical variation of the Clunes pluton is not amenable to detailed modelling of magmatic evolution. However, samples from Cluanie were plotted on La vs. Rb and La vs. Yb plots alongside Rayleigh fractional crystallisation (FC) vectors for the known mineral phases (Figs 6a and 6b), starting with the lowest-SiO_2 porphyrite. Partition coefficients are listed in the Supplementary Item. Most samples define a trend towards low La concentrations at fixed or decreasing Rb. Potential fractionating phases in which La and Yb are strongly compatible (e.g., titanite, apatite, zircon, and allanite) and in which Rb is strongly incompatible generate near-vertical trends on Figure 6a. Several samples do trend to the left of the diagram, explained by FC of a Rb-bearing phase such as biotite, common in marginal facies of the pluton but less common elsewhere (Fig. 1b). Figure 6b better distinguishes fractionation of different REE-compatible accessory phases, with samples falling between vectors consistent with FC of apatite, titanite, biotite, allanite, and zircon. REE concentrations were also modelled using Rayleigh FC with mineral proportions iteratively modified (Fig. 6c; Supplementary Item). The lowest-SiO_2 porphyrite was taken as the parental magma, and ~10% FC reasonably reproduced the most evolved granodiorite, including the generation of a pronounced spoon-shaped REE pattern with a slight positive Eu anomaly. The modest proportion of FC modelled is consistent with the limited major element variation of the suite from porphyrites to the plutonic samples from ~66-72 wt.% SiO_2 and ~3 to 2 wt.% MgO.

Source(s) of partial melt and origin of adakitic geochemical signatures

The main outcome from the radiogenic isotope results and the lack of pre-Laxfordian inherited zircons is that the source of Cluanie magmatism was not an ancient crustal reservoir. The deep crust beneath the Northern Highlands is likely to consist of low-Rb/Sr Archaean-Paleoproterozoic Lewisian gneisses, an unsuitable melt source for this pluton (Fowler *et al.* 2008). Therefore, bearing in mind the adakite-like composition of both plutons, they may have originated principally from: a) the down-going Iapetus slab; b) partial melting of an unrecognised young crustal source such as a mafic underplate; c) a variation of the model of FC plus minor crustal assimilation from a subduction-modified mantle source (Fowler *et al.* 2008), considering the new evidence for a protracted history of zircon crystallisation described in the results above.

Option a) slab melting: In the geodynamic model of Dewey *et al.* (2015), flat-slab subduction occurred during the Ordovician beneath Scotland, accounting for both the contractile deformation of Bird *et al.* (2013) and the perceived lack of magmatism between the Grampian and Scandian Orogenies. Flat-slab scenarios are certainly associated with slab melting, with adakitic slab melts retaining low MgO and transition metal characteristics owing to limited interaction with a very thin mantle wedge above the shallow slab (Hastie *et* *al.* 2015). However, a serious problem is the occurrence of the Shetland granitoids, the Assynt Alkaline Suite, and the Glen Dessary syenite. All these bodies are proposed to be the end product of evolution from mafic, mantle-derived parental magmas, indicating mantle melting was occurring prior to 430 Ma beneath Scotland (Fowler *et al.* 1992; Goodenough *et al.* 2011; Lancaster *et al.* 2017; Fig. 2).

Option b) lower crustal melting: whole rock isotopic data indicate the source has to be geologically young, so an Iapetus rift- or subduction-related magmatic underplate may be considered (e.g., Atherton & Petford 1993, Thybo & Artemieva 2013). Neill & Stephens (2009) favoured this model for the Cluanie pluton, based on its uniformly adakite-like composition, lack of mafic facies, and presence of possible restite ‘clots’. Experimental results demonstrate crustal melting can produce magmas of ≥60 wt.% SiO_2 (e.g., Rapp et al., 1991; Wolf & Wyllie, 1994; Rapp & Watson, 1995), encompassing all analysed rocks. Lower crust-derived adakites can contain alkali feldspar (e.g., Guo *et al.* 2007). However, there are problems with this model, too. Firstly, there are few dated igneous rocks to substantiate regionally extensive underplating beneath the Northern Highlands prior to the emplacement of the Cluanie pluton (Oliver *et al.* 2008). Secondly, we cannot be certain that the mafic clots and schlieren of Neill & Stephens (2009) necessarily are source-derived restite, as opposed to a product of incomplete crustal assimilation, or reaction between mafic cumulate autoliths and the host magma. Finally, the occurrence of largely felsic bodies within ultimately mantle-derived magmatic arcs and post-collision settings is globally ubiquitous, so a lack of mafic facies at Cluanie and Clunes should not *a priori* preclude mantle melting as their ultimate source.

Option c) the deep crustal hot zone hypothesis: The older cluster of magmatic zircons clearly indicates that magmatism was active beneath Cluanie for around 20 myr prior to emplacement, and before the accepted onset of “Newer Granite” magmatism. The zircon history is consistent with the development of a deep crustal hot zone where magma addition, storage, and differentiation could occur over such timescales. The Grampian-2 event of Bird et al. (2013) and Walker et al. (2020) at ~450 Ma, and the onset of the Scandian event at ~437 Ma (Strachan *et al.* 2020), effectively bracket the older cluster of zircon dates from Cluanie, supporting a period of subduction-related magmatism which generated the parental magmas to Cluanie and probably Clunes. Modelling and experimental work demonstrates that a mantle-derived magma which had evolved to andesitic composition with >8 wt.% H_2O would fractionate hornblende ± garnet at depths of ~30 km (Alonso-Perez *et al.* 2008), generating adakite-like chemistry (e.g., Richards *et al.* 2012). The limited geochemical ranges of the two plutons and the extended zircon crystallisation history at Cluanie may reflect the ascent and emplacement of well-homogenised, long-stored batches of magma. Significant mantle-derived magma flux and disturbance of the hot zone would have occurred after slab breakoff at ~428 Ma, resulting in the considerably more varied facies and geochemical ranges of younger members of the “Newer Granites”. The adakite-like geochemistry of Cluanie and Clunes does however contrast with the more potassic and REE-enriched geochemistry of the penecontemporaneous Assynt Alkaline Suite towards the hinterland (Thompson & Fowler 1986). Such differences might reflect the latter having experienced differentiation within thinner crust on the margins of the orogenic belt, lower degrees of mantle melting further from the Iapetus slab, and a higher proportion of crustal assimilation from high grade Hebridean rocks, compared to the more fertile lithologies of the Wester Ross and Loch Ness Supergroups.

The timing of Caledonian geodynamic events

Stewart *et al.* (2011) used their geochronology and structural analysis of the Clunes tonalite to show that the Great Glen Fault was undergoing left-lateral motion ~428 Ma ago. However, the Cluanie Pluton is slightly older than the Clunes tonalite. Neill & Stephens

(2009) proposed that Cluanie was emplaced in a pull-apart at the junction of fault sets associated with dextral motion on the NE-SW-striking Strathglass Fault and other NW-SE-striking faults (Fig. 1b), in turn relating the Strathglass Fault to movement on the Great Glen Fault Zone. Given an emplacement age of ~432 Ma, the Cluanie pluton therefore sets not only a new minimum age for strike-slip faulting in the Northern Highlands, but also importantly sets a maximum age for a switch from initial dextral to subsequent sinistral motion of the GGFZ and associated faults shortly after emplacement between ~432 and ~430 Ma (Holdsworth *et al.* 2015).

Above, we have followed the popular interpretation that slab breakoff occurred at ~428 Ma beneath Scotland, so Cluanie, the Assynt Alkaline Suite, and the granitoids associated with the Naver Thrust were thus emplaced during the last stages of Iapetus subduction prior to slab breakoff. It is commonly accepted that "Newer Granite" magmatism in the Northern Highlands began prior to that in the Grampian Highlands (Table 1). Therefore, is a diachronous Baltica-Laurentia collision and breakoff a feasible alternative scenario to explain this age progression, as discussed by Archibald *et al.* (2022)? Post-breakoff "Newer Granite" magmatism would thus occur first beneath Shetland (ca 440 Ma), progressing along the Laurentian margin beneath the Northern Highlands including Orkney (ca mid-430's Ma), then the Grampian Highlands (ca 428 Ma), the Midland Valley and finally the Southern Uplands (ca 415 Ma), where termination of accretionary prism sedimentation occurred at ~425 Ma (e.g., Archibald *et al.* 2022).

However, in the Northern Highlands, "peak" Scandian metamorphism is dated to ~425 Ma from U-Pb zircon dating of East Sutherland migmatites (Kinny *et al.* 1999; Friend *et al.* 2003). Slab breakoff is thought unlikely to occur prior to peak metamorphic conditions during orogenesis (e.g., Henk *et al.* 2000; Platt *et al.* 2003). Yet, if "peak" metamorphism records magmatic advection, not maximum lithospheric thickness, the temporal order between breakoff and peak metamorphism may not hold true. A clearer indication of breakoff timing is that the greatest volume of Newer Granite magmatism occurs after ~426 Ma in both the Northern and Grampian Highlands (Oliver *et al.* 2008), so the modest volumes identified before this time strongly implies a distinct geodynamic regime pre-426 Ma in both the Grampian and Northern Highlands. Therefore, we conclude that a diachronous slab breakoff is not likely with respect to the Northern and Grampian Highlands, and our data supports that Northern Highlands magmatism prior to ~426 Ma reflects supra-subduction activity. Our data do however constrain the timing of a change in geodynamic environment, between ~432 and ~430 Ma, which may be reflected in a kinematic switch in the sense of strike-slip faulting in the Northern Highlands associated with the initiation of the GGFZ.

The overall paucity of continental arc plutons from ~450-430 Ma is somewhat negated by recent and new dates from the Cluanie pluton, the Naver granitoids and the Orkney and Shetland plutons. Nevertheless, modest magmatic output prior to the main phase of "Newer Granite" magmatism still requires explanation. It seems likely that oblique subduction beneath the Laurentian margin (Oliver *et al.* 2008) was combined with a compressive upper plate regime, limiting magmatic emplacement to low-strain intersections of pre-existing lineaments and strike-slip faults. New evidence in this work for long-term storage of magma from ~450-430 Ma also supports this hypothesis. Additionally, Slagstadt & Kirkland (2018) argued that lower-plate high pressure metamorphism in Scandinavia, pre-dating the Scandian Orogeny, occurred because Baltican promontories collided with Laurentia in advance of terminal collision. The argument for a contemporary compressive upper plate regime follows from that model. In Scotland, which solely represents the upper plate, the Grampian-2 event at ~450 Ma is now widely recognised, though argued to result from microcontinent accretion as opposed to collision with the leading edge of Baltica (Bird *et al.* 2013; Walker *et al.* 2020). All in, limited volumes of subduction-related magmatism from ~450-430 Ma in Scotland can be ascribed to substantively thickened upper plate crust being present prior to the Scandian episode.

Future geochronological research on the Caledonian intrusions

The existing geochronological framework for Caledonian magmatism has been constructed from multiple approaches (Table 1) with multiple reporting conventions such as the use of weighted mean $^{207}Pb/^{206}Pb$ vs $^{206}Pb/^{238}U$ ages (e.g., Rogers & Dunning 1991 vs Oliver *et al.* 2008). Caution is therefore required in terms of how published ages are interpreted, particularly whether they represent final emplacement or hot zone growth, and whether we assign plutons to subduction or post breakoff settings. The above new data are among very few LA-ICP-MS U-Pb zircon results for Caledonian plutons (e.g., Lundmark *et al.* 2018; Archibald *et al.* 2022). Otherwise, zircon chemical- or air-abrasion isotope dilution U-Pb thermal ionisation mass spectrometry (CA/AA-ID-TIMS) has otherwise been commonplace in the Northern Highlands, often without cathodoluminescence images for textural control (e.g., Rogers & Dunning 1991). An ion probe study by Oliver *et al.* (2008) dominates the Grampian Highlands record, and there are also titanite and baddeleyite U-Pb, sulphide Re-Os and whole rock Rb-Sr dates in both terranes (Table 1; Brook 1985; Conliffe *et al.* 2010; Holdsworth *et al.* 2015; Rogers & Dunning 1991). Recent ion probe studies in Southern Scotland and Ireland have, like Cluanie, shown distinct zircon populations, covering a wider (and younger) range of dates compared to regular CA-ID-TIMS methods (e.g., Archibald *et al.* 2022; Fritschle *et al.* 2018; Miles *et al.* 2014; Miles & Woodcock 2018; Woodcock *et al.* 2019). Oliver *et al.* (2008) included age ranges of up to 30 myr in $^{206}Pb/^{238}U$ weighted mean ages assigned to magmatic emplacement (e.g., Foyers, Ross of Mull, Boat of Garten, Laggon, Findhorn and Skene). These results could also be interpreted as evidence for recycling of antecrystic zircon populations from a deep crustal hot zone.

Such complexities are not clearly shown in the ID-TIMS studies of Northern Highlands (e.g., Rogers & Dunning 1991). U-Pb ID-TIMS typically uses few hand-picked grains, so the sampling of well-formed crystals *may* bias ages towards crystals which grew in the deep crustal hot zone. Thus, it may be prudent to consider some published ID-TIMS ages as maxima for emplacement, unless structural, textural, or associated in-situ geochronological studies provide supporting evidence. Our texturally-constrained LA-ICP-MS approach with larger numbers of analysed crystals than the ID-TIMS studies to date, gives the added potential of capturing the duration of deep crustal hot zone activities as well as the timing of emplacement. As shown in Table 1, ages for Rogart, the LREE prospect at Loch Loyal, Ratagain, Strath Halladale, Rogart and Strontian may be enhanced with such additional analysis. Lawrence *et al.* (2022) also queried the age of the Ratagain pluton based on its magnetic fabrics, kinematics of emplacement, and bulk geochemistry, in turn implying the incorporation of antecrystic zircons during past dating (Rogers & Dunning 1991). Therefore, a small but growing body of evidence points towards the opportunity for refinements to the geochronological framework of the Northern Highlands "Newer Granites". The interpretation of ID-TIMS data may be significantly enhanced through a combination of preliminary LA-ICP-MS or ion probe work, cathodoluminescence imaging of selected half-grains, and improved abrasion techniques to resolve multiple events (e.g., Gaynor *et al.* 2022).

"Newer Granites" vs subduction-related and post-subduction Caledonian intrusions

Read's (1961) definition of the "Newer Granites" was split between "forceful" and "last" (or "permitted") intrusions which were all emplaced sometime after the metamorphism and folding of the Moine and Dalradian series. The two types were distinguished on the basis of the apparent migmatisation at the margins of the former, and their possible association with appinitic magmas; and the shallow crustal, fault-controlled emplacement of the latter. Read's examples of the former in the Scottish Highlands included Rogart, Strontian, Rannoch

Moor, Ballachulish and Ross of Mull, whereas the latter included Glen Coe, Nevis, Cruachan and Lochnagar. The Assynt Alkaline Suite was treated as un-categorised because it could not be structurally associated with events further south and east in the Northern Highlands (Read 1961). Several aspects of our modern understanding, including the new data in this paper, render the term "Newer Granites" problematic. Firstly, "forceful" emplacement has been overtaken as a concept by the association of these plutons with orogen-parallel strike-slip systems and associated shear zones (e.g., Hutton 1988; Hutton & McErlean 1991; Neill & Stephens 2009; Stewart *et al.* 2001). Secondly, the presumption that the "Newer Granites" post-date folding and metamorphism in the Moine and Dalradian series is incorrect given their temporal overlap with the Scandian Orogeny between ~437 and 415 Ma (Strachan *et al.* 2020) and the imprint of Scandian fabrics on some "Newer Granites" (e.g., Glen Scaddle at 426 ± 3 Ma, Table 1). "Forceful" bodies such as those listed by Read (1961) range in age from 433.5 ± 1.8 Ma (Ballachulish, Conliffe *et al.* 2010, molybdenite Re-Os) to approximately 418 Ma (Ross of Mull and Strontian 2, Table 1). Aside from the temporal overlap with Scandian events, the oldest of these intrusions relate to Iapetus subduction (e.g., Ballachulish, Assynt Alkaline Suite, Cluanie, Clunes) whereas a majority are attributable to post breakoff magmatism, including some of the "forceful" bodies and all those deemed "passive" (Read 1961).

Hence, not all post-Grampian Caledonian intrusions are "Newer", self-evidently not all are "Granites", and the term is not characteristic of a specific petrogenetic pathway or a geodynamic setting. The more neutral term "Caledonian intrusions" is suggested as a replacement. There may also be value in specifically associating Caledonian intrusions with subduction processes (prior to 426 Ma in the Northern and Grampian Highlands) or with mantle melting following Iapetus breakoff ("post-subduction" magmatism of Richards 2009). One reason for this important distinction is that post breakoff Caledonian intrusions remain potentially of economic interest, including in the Cairngorms for Li, W and Nb-Ta (British Geological Survey 2020), Kilmelford in the Grampian Highlands for porphyry Cu (Ellis *et al.* 1977) and Dumfries and Galloway for Cu sulphides and Ag (Brown *et al.* 1979; Leake *et al.* 1981). Slab breakoff is globally associated with rapid uplift and addition of new volatile-rich mantle-derived magmas and sulphides, which contribute to favourable conditions for mineralisation (Richards 2009, 2015; Vos *et al.* 2007). Hence careful geochronological study of Caledonian intrusions suspected to pre- or post-date slab breakoff may provide context for future exploration.

Conclusions

LA-ICP-MS U-Pb zircon dating demonstrates the Cluanie Pluton in the Northern Highlands of Scotland was emplaced at ca 432 Ma. Pre-emplacement zircon growth from ca 435-450 Ma took place in a deep crustal hot zone. The adakite-like geochemistry of both Cluanie and the ca 428 Ma Clunes tonalite are distinct in the Northern Highlands and reflect tapping of well-homogenised magma reservoirs prior to more extensive mantle-derived magma addition and stirring of the hot zone following Iapetus slab breakoff. The comparatively few Northern Highlands intrusions emplaced before the bulk of Newer Granite magmatism reflect the latter stages of Iapetus subduction beneath an already-thickened crust after the Grampian-2 event. The differing geochemical signatures of these ‘early’ plutons such as the sodic, REE-depleted Cluanie and Clunes bodies, vs the potassic, REE-enriched, Assynt Alkaline Suite, may reflect different mantle melting conditions, varying crustal storage conditions and the nature of crustal contaminants. We contend that the zircon growth history recorded within the Cluanie pluton may be present in many other Northern Highlands plutons and could be detected through refined geochronological studies. The term “Newer Granites”, widely used to describe plutons such as Cluanie and Clunes, is also discussed. It is suggested that the term be replaced with the more neutral “Caledonian intrusions” and that individual bodies are better categorised by their association either with Iapetus subduction or slab breakoff, particularly from a metallogenetic perspective.

Declaration of Competing Interests

Authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Contributions

E.M. - sample preparation, analysis, initial writing, and interpretation. I.N. – original concept, fieldwork, sample preparation, writing, figures, and interpretation. I.L.M. – sample preparation, analysis, and initial writing. I.M. – analysis. A.F.B. – interpretation, editing and revision. E.D.D. – interpretation and editing. V.O. – analysis. N.O. – analysis. E.C.W. – analysis.

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Supplementary Item

A single supplementary item contains three Excel worksheets:

Worksheet 1 – laser ablation data

Worksheet 2 – all major and trace element data and radiogenic isotope results

Worksheet 3 – comparison of existing and new geobarometric data

Table and Figure Captions

Table 1. Geochronology of Northern Highlands “Newer” granites. Z = zircon; MB = molybdenite; M = monazite; B = baddeleyite; T = titanite; ID-TIMS = isotope dilution thermal ionisation mass spectrometry; LA-ICP-MS = laser ablation inductively-coupled plasma mass spectrometry; SHRIMP = sensitive high resolution ion microprobe. Helmsdale and Fearn remain undated, as do many minor intrusions. Discussion of the timing of geodynamic events is in the text.

Figure 1. a) Map of Scotland after Lancaster *et al.* (2017) and Lundmark *et al.* (2019). b) Satellite image of key Northern Highlands faults and plutons near Cluanie. Open access high resolution satellite data from 2022 via Bing Maps (https://bing.com/maps/aerial) and Geology Digimap (Geological Map Data BGS © UKRI 2022). c) Map of the Cluanie Pluton after Neill & Stephens (2009). d) Map of the Clunes tonalite after Stewart et al. (2001).

Figure 2. Caledonian intrusions and their relationship to major geodynamic events in the Northern and Grampian Highlands, after Lancaster *et al.* (2017). Data are in Table 1 for the Northern Highlands and Oliver *et al.* (2008) for the Grampian Highlands, with the exception of Ballachulish and Kilmelford from Conliffe *et al.* (2010).

Figure 3. U-Pb data for the Cluanie Pluton. For full data see Supplementary Item. a) Zircon inheritance record for the Cluanie Pluton with a Wetherill concordia plot and

examples of typical zircon textures. b) Kernel density plot of the same data highlighting key events in the Scottish geological record. c) Cathodoluminescence (CL) images of zircon grains yielding spots which contributed to calculation of the emplacement age and duration of deep crustal storage. d) Wetherill concordia plot showing all concordant Caledonian analyses and highlighting two apparent clusters of data. e) Concordant Caledonian analyses ordered by age, showing weighted mean ages and confidence intervals for the interpreted emplacement age and deep crustal hot zone growth.

Figure 4. Major and trace element variation diagrams for the Cluanie Pluton and Clunes tonalite.

Figure 5. Chondrite- (McDonough & Sun 1995) and primitive mantle- (Sun & McDonough 1989) normalised plots for the Cluanie Pluton and Clunes tonalite.

Figure 6. a-c) Adakite geochemical classification diagrams based on Martin (1999), with data from Fowler et al. (2008) for the Northern Highlands; d) Radiogenic isotope data for the Northern Highlands plutons from Fowler et al. (2008) including new results for the Cluanie Pluton.

Figure 7. Trace element modelling of fractional crystallisation in the Cluanie Pluton from a starting composition of felsic porphyrite minor intrusion IN/27-6/2. All modelling parameters are in the Supplementary Item. (a) La vs. Rb vector plot showing the strong influence of accessory minerals on the composition of the Cluanie Pluton samples. (b) La vs. Yb vector plot. (c) Modelling of Rayleigh fractional crystallisation of the REE. The REE model was constructed using the following mineral proportions: 0.5 plagioclase, 0.35 amphibole, 0.2 biotite, 0.12 apatite, 0.1 titanite, 0.005 zircon, 0.0035 allanite, 0.001 ilmenite (fractionating) and –0.18175 quartz, -0.09775 orthoclase (accumulating).

Figure 1

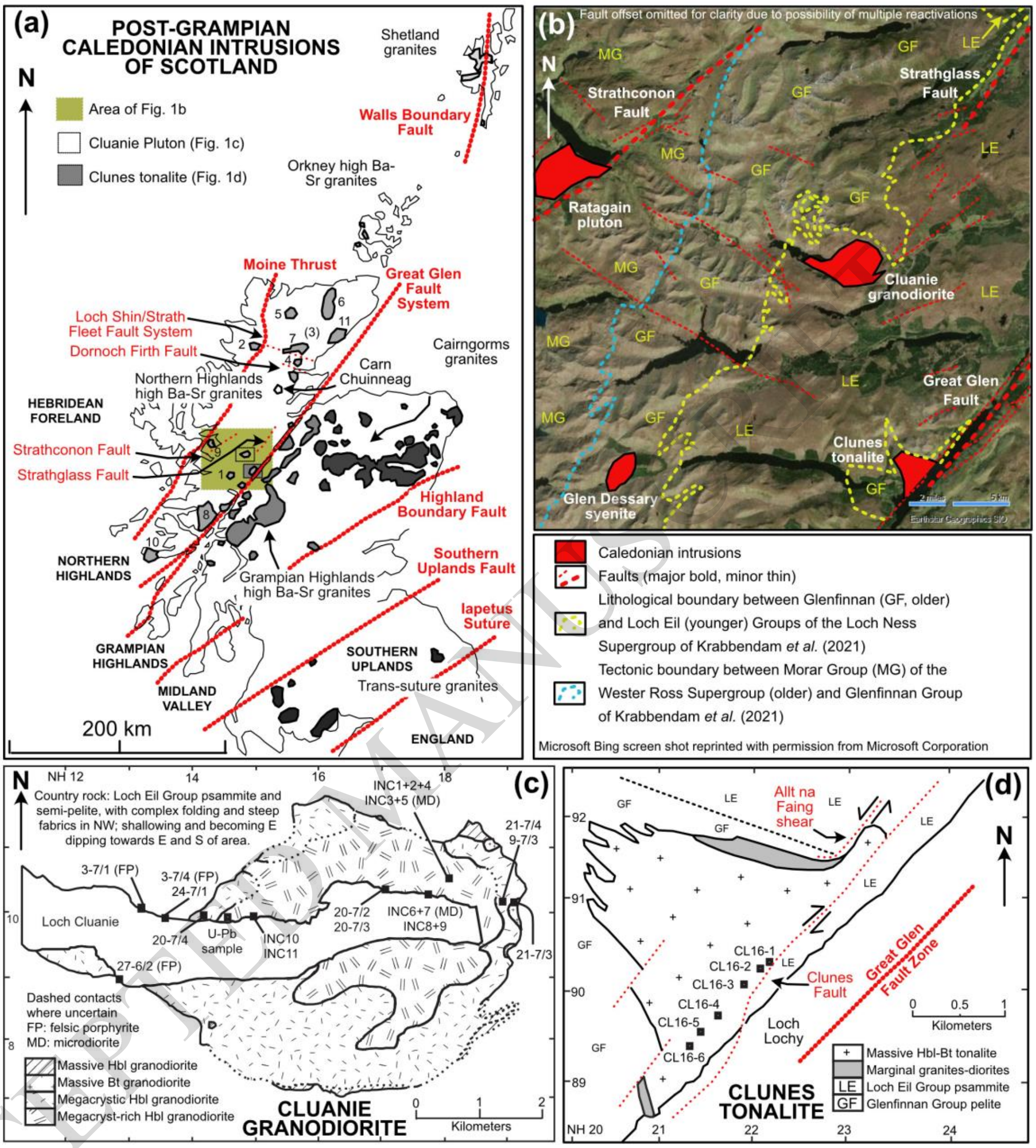
(a)
POST-GRAMPIAN CALEDONIAN INTRUSIONS OF SCOTLAND
N
Area of Fig. 1b
Cluanie Pluton (Fig. 1c)
Clunes tonalite (Fig. 1d)
Shetland granites
Walls Boundary Fault
Orkney high Ba-Sr granites
Moine Thrust
Great Glen Fault System
Loch Shin/Strath Fleet Fault System
Dornoch Firth Fault
Cairngorms granites
Carn Chuinneag
Northern Highlands high Ba-Sr granites
HEBRIDEAN FORELAND
Strathconon Fault
Strathglass Fault
Highland Boundary Fault
NORTHERN HIGHLANDS
Grampian Highlands high Ba-Sr granites
Southern Uplands Fault
Iapetus Suture
GRAMPIAN HIGHLANDS
SOUTHERN UPLANDS
MIDLAND VALLEY
Trans-suture granites
200 km
ENGLAND
1
2
(3)
4
5
6
7
8
9
10
11
(b)
Fault offset omitted for clarity due to possibility of multiple reactivations
N
MG
GF
LE
Strathconon Fault
Strathglass Fault
Ratagain pluton
Cluanie granodiorite
Great Glen Fault
Clunes tonalite
Glen Dessary syenite
2 miles
5 km
Earthstar Geographics SIO
Caledonian intrusions
Faults (major bold, minor thin)
Lithological boundary between Glenfinnan (GF, older) and Loch Eil (younger) Groups of the Loch Ness Supergroup of Krabbendam et al. (2021)
Tectonic boundary between Morar Group (MG) of the Wester Ross Supergroup (older) and Glenfinnan Group of Krabbendam et al. (2021)
Microsoft Bing screen shot reprinted with permission from Microsoft Corporation
(c)
N
NH 12
14
16
18
10
8
Country rock: Loch Eil Group psammite and semi-pelite, with complex folding and steep fabrics in NW; shallowing and becoming E dipping towards E and S of area.
INC1+2+4
INC3+5 (MD)
21-7/4
9-7/3
3-7/1 (FP)
3-7/4 (FP)
24-7/1
Loch Cluanie
20-7/4
U-Pb sample
INC10
INC11
20-7/2
20-7/3
INC6+7 (MD)
INC8+9
21-7/3
27-6/2 (FP)
Dashed contacts where uncertain
FP: felsic porphyrite
MD: microdiorite
Massive Hbl granodiorite
Massive Bt granodiorite
Megacrystic Hbl granodiorite
Megacryst-rich Hbl granodiorite
CLUANIE GRANODIORITE
0
1
2
Kilometers
(d)
92
91
90
89
GF
LE
Allt na Faing shear
CL16-1
CL16-2
CL16-3
CL16-4
CL16-5
CL16-6
Clunes Fault
Loch Lochy
Great Glen Fault Zone
N
0
0.5
1
Kilometers
Massive Hbl-Bt tonalite
Marginal granites-diorites
Loch Eil Group psammite
Glenfinnan Group pelite
CLUNES TONALITE
NH 20
21
22
23
24

Figure 2

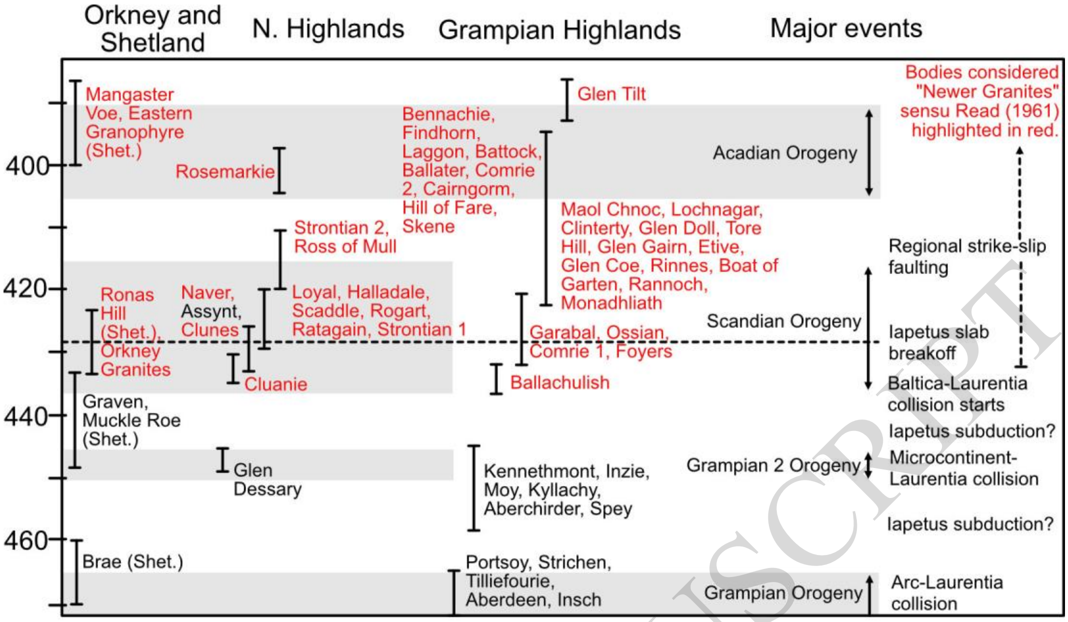
Orkney and Shetland
N. Highlands
Grampian Highlands
Major events
400
420
440
460
Mangaster Voe, Eastern Granophyre (Shet.)
Rosemarkie
Bennachie, Findhorn, Laggon, Battock, Ballater, Comrie 2, Cairngorm, Hill of Fare, Skene
Glen Tilt
Acadian Orogeny
Bodies considered "Newer Granites" sensu Read (1961) highlighted in red.
Strontian 2, Ross of Mull
Maol Chnoc, Lochnagar, Clinterty, Glen Doll, Tore Hill, Glen Gairn, Etive, Glen Coe, Rinnes, Boat of Garten, Rannoch, Monadhliath
Regional strike-slip faulting
Ronas Hill (Shet.), Orkney Granites
Naver, Assynt, Clunes
Loyal, Halladale, Scaddle, Rogart, Ratagain, Strontian 1
Garabal, Ossian, Comrie 1, Foyers
Scandian Orogeny
Iapetus slab breakoff
Cluanie
Ballachulish
Baltica-Laurentia collision starts
Graven, Muckle Roe (Shet.)
Iapetus subduction?
Glen Dessary
Kennethmont, Inzie, Moy, Kyllachy, Aberchirder, Spey
Grampian 2 Orogeny
Microcontinent-Laurentia collision
Iapetus subduction?
Brae (Shet.)
Portsoy, Strichen, Tilliefourie, Aberdeen, Insch
Grampian Orogeny
Arc-Laurentia collision

Figure 3

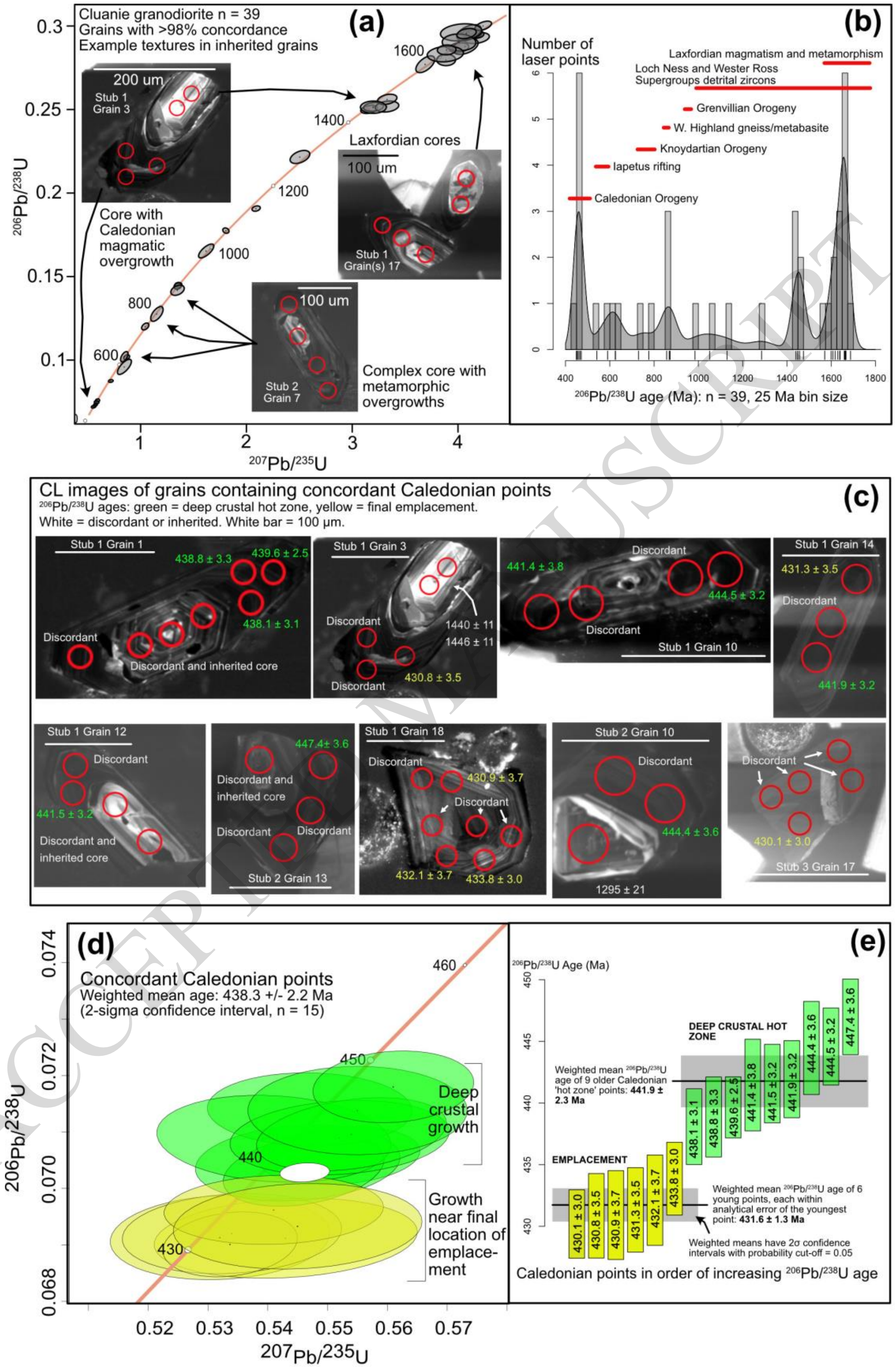
(a)
Cluanie granodiorite n = 39
Grains with >98% concordance
Example textures in inherited grains
200 um
Stub 1 Grain 3
Core with Caledonian magmatic overgrowth
Laxfordian cores
100 um
Stub 1 Grain(s) 17
100 um
Stub 2 Grain 7
Complex core with metamorphic overgrowths
600
800
1000
1200
1400
1600
0.1
0.15
0.2
0.25
0.3
1
2
3
4
206Pb/238U
207Pb/235U
(b)
Number of laser points
Laxfordian magmatism and metamorphism
Loch Ness and Wester Ross Supergroups detrital zircons
Grenvillian Orogeny
W. Highland gneiss/metabasite
Knoydartian Orogeny
Iapetus rifting
Caledonian Orogeny
0
1
2
3
4
5
6
400
600
800
1000
1200
1400
1600
1800
206Pb/238U age (Ma): n = 39, 25 Ma bin size
(c)
CL images of grains containing concordant Caledonian points
206Pb/238U ages: green = deep crustal hot zone, yellow = final emplacement.
White = discordant or inherited. White bar = 100 μm.
Stub 1 Grain 1
438.8 ± 3.3
439.6 ± 2.5
438.1 ± 3.1
Discordant
Discordant and inherited core
Stub 1 Grain 3
Discordant
1440 ± 11
1446 ± 11
Discordant
430.8 ± 3.5
441.4 ± 3.8
Discordant
444.5 ± 3.2
Discordant
Stub 1 Grain 10
Stub 1 Grain 14
431.3 ± 3.5
Discordant
441.9 ± 3.2
Stub 1 Grain 12
Discordant
441.5 ± 3.2
Discordant and inherited core
447.4± 3.6
Discordant and inherited core
Discordant
Discordant
Stub 2 Grain 13
Stub 1 Grain 18
Discordant
430.9 ± 3.7
Discordant
432.1 ± 3.7
433.8 ± 3.0
Stub 2 Grain 10
Discordant
444.4 ± 3.6
1295 ± 21
Discordant
430.1 ± 3.0
Stub 3 Grain 17
(d)
Concordant Caledonian points
Weighted mean age: 438.3 +/- 2.2 Ma
(2-sigma confidence interval, n = 15)
460
450
440
430
Deep crustal growth
Growth near final location of emplacement
0.068
0.070
0.072
0.074
0.52
0.53
0.54
0.55
0.56
0.57
206Pb/238U
207Pb/235U
(e)
206Pb/238U Age (Ma)
430
435
440
445
450
DEEP CRUSTAL HOT ZONE
Weighted mean 206Pb/238U age of 9 older Caledonian 'hot zone' points: 441.9 ± 2.3 Ma
EMPLACEMENT
430.1 ± 3.0
430.8 ± 3.5
430.9 ± 3.7
431.3 ± 3.5
432.1 ± 3.7
433.8 ± 3.0
438.1 ± 3.1
438.8 ± 3.3
439.6 ± 2.5
441.4 ± 3.8
441.5 ± 3.2
441.9 ± 3.2
444.4 ± 3.6
444.5 ± 3.2
447.4 ± 3.6
Weighted mean 206Pb/238U age of 6 young points, each within analytical error of the youngest point: 431.6 ± 1.3 Ma
Weighted means have 2σ confidence intervals with probability cut-off = 0.05
Caledonian points in order of increasing 206Pb/238U age

Figure 4

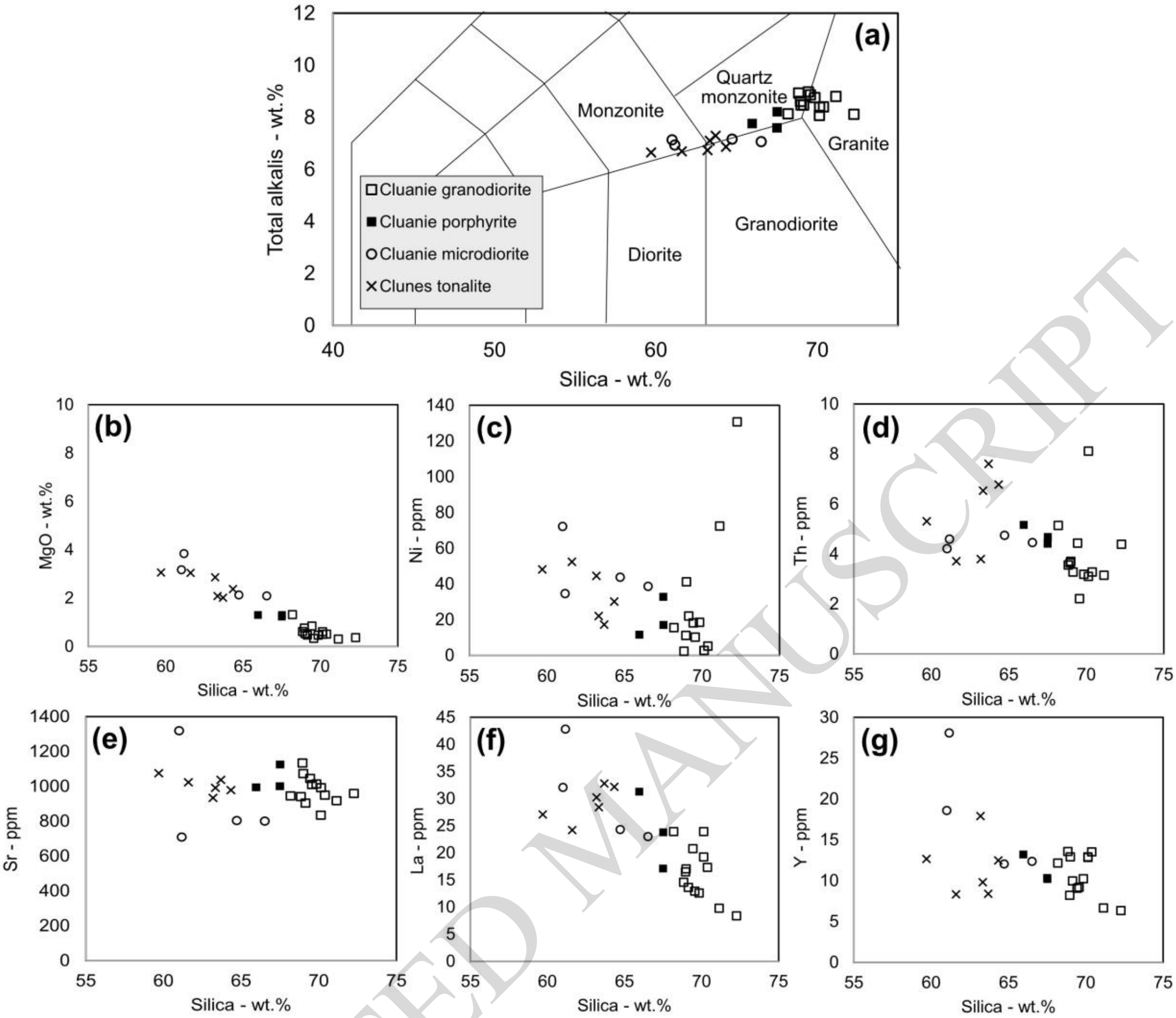
(a)
Total alkalis - wt.%
Silica - wt.%
Monzonite
Quartz monzonite
Granite
Diorite
Granodiorite
□ Cluanie granodiorite
■ Cluanie porphyrite
○ Cluanie microdiorite
× Clunes tonalite
(b)
MgO - wt.%
(c)
Ni - ppm
(d)
Th - ppm
(e)
Sr - ppm
(f)
La - ppm
(g)
Y - ppm
Silica - wt.%

Figure 5

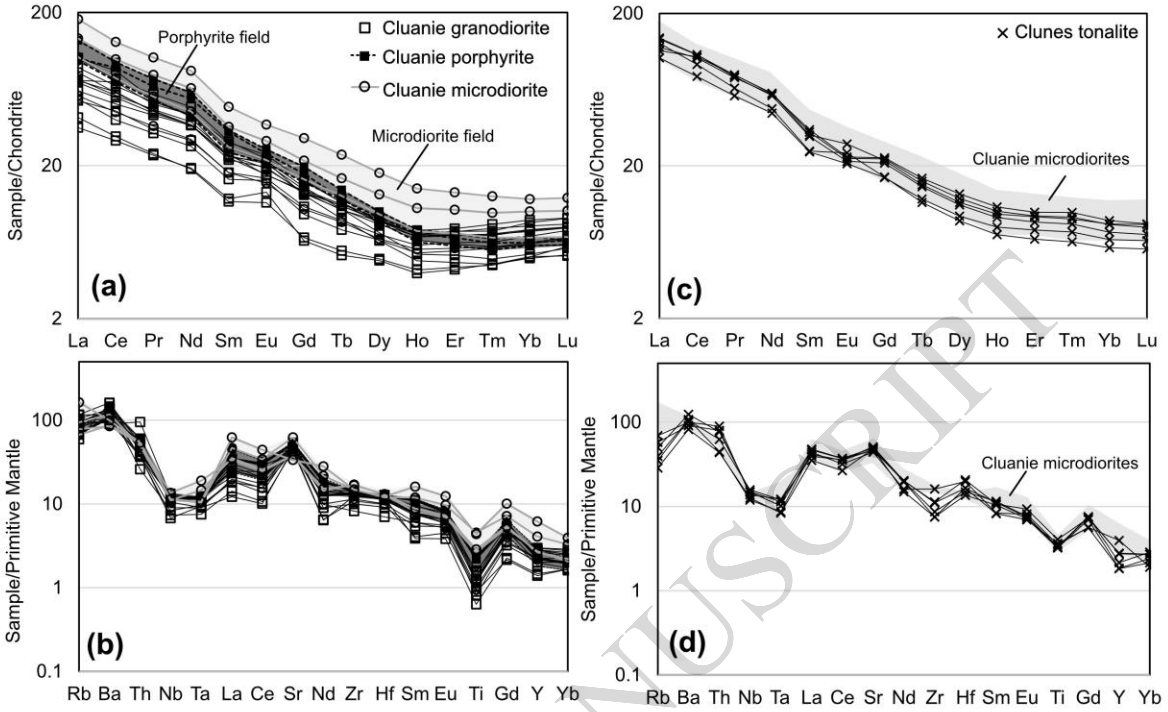
200
20
2
Sample/Chondrite
Porphyrite field
Microdiorite field
Cluanie granodiorite
Cluanie porphyrite
Cluanie microdiorite
(a)
La Ce Pr Nd Sm Eu Gd Tb Dy Ho Er Tm Yb Lu
100
10
1
0.1
Sample/Primitive Mantle
(b)
Rb Ba Th Nb Ta La Ce Sr Nd Zr Hf Sm Eu Ti Gd Y Yb
× Clunes tonalite
Cluanie microdiorites
(c)
La Ce Pr Nd Sm Eu Gd Tb Dy Ho Er Tm Yb Lu
Cluanie microdiorites
(d)
Rb Ba Th Nb Ta La Ce Sr Nd Zr Hf Sm Eu Ti Gd Y Yb

Figure 6

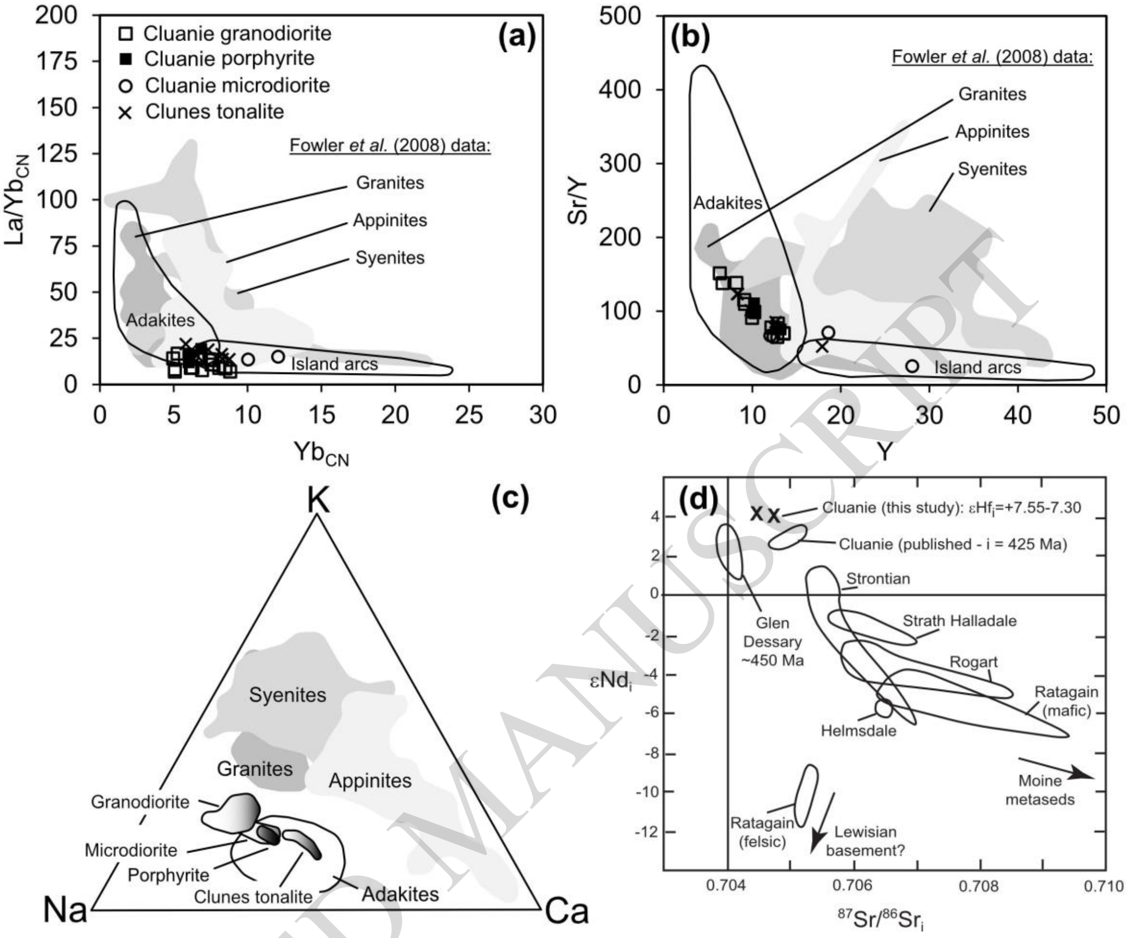
(a)
Cluanie granodiorite
Cluanie porphyrite
Cluanie microdiorite
Clunes tonalite
Fowler et al. (2008) data:
Granites
Appinites
Syenites
Adakites
Island arcs
La/Yb$_{CN}$
Yb$_{CN}$
200
175
150
125
100
75
50
25
0
0
5
10
15
20
25
30
(b)
Fowler et al. (2008) data:
Granites
Appinites
Syenites
Adakites
Island arcs
Sr/Y
Y
500
400
300
200
100
0
0
10
20
30
40
50
(c)
K
Na
Ca
Syenites
Granites
Appinites
Granodiorite
Microdiorite
Porphyrite
Clunes tonalite
Adakites
(d)
Cluanie (this study): εHf$_i$=+7.55-7.30
Cluanie (published - i = 425 Ma)
Strontian
Glen Dessary ~450 Ma
Strath Halladale
Rogart
Ratagain (mafic)
Helmsdale
Moine metaseds
Ratagain (felsic)
Lewisian basement?
εNd$_i$
^{87}Sr/^{86}Sr$_i$
4
2
0
-2
-4
-6
-8
-10
-12
0.704
0.706
0.708
0.710

Figure 7

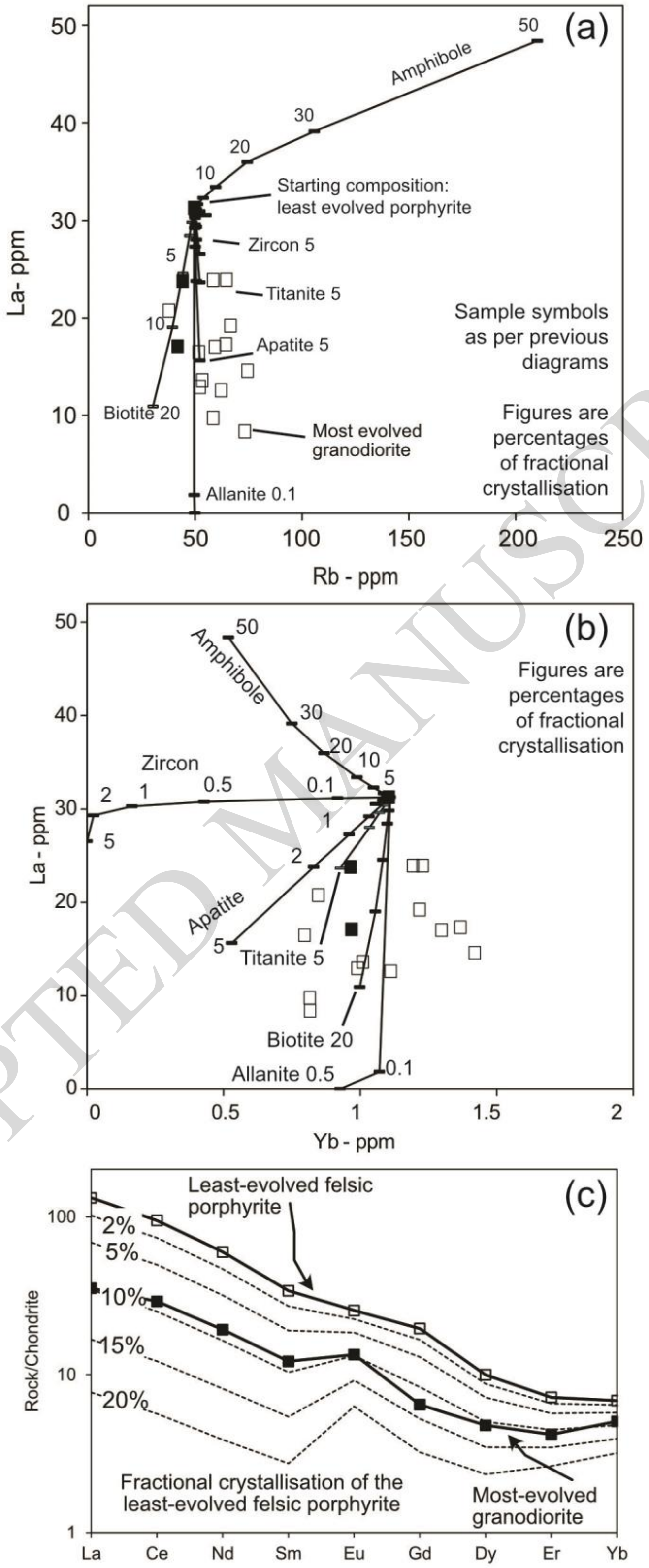
(a)
La- ppm
Rb - ppm
0
50
100
150
200
250
10
20
30
40
50
Amphibole
50
30
20
10
Starting composition:
least evolved porphyrite
Zircon 5
Titanite 5
Apatite 5
5
10
Biotite 20
Most evolved
granodiorite
Allanite 0.1
Sample symbols
as per previous
diagrams
Figures are
percentages
of fractional
crystallisation
(b)
La - ppm
Yb - ppm
0
0.5
1
1.5
2
Amphibole
50
30
20
10
5
Zircon
2
1
0.5
0.1
5
1
2
Apatite
5
Titanite 5
Biotite 20
Allanite 0.5
0.1
Figures are
percentages
of fractional
crystallisation
(c)
Rock/Chondrite
1
10
100
La
Ce
Nd
Sm
Eu
Gd
Dy
Er
Yb
Least-evolved felsic
porphyrite
2%
5%
10%
15%
20%
Fractional crystallisation of the
least-evolved felsic porphyrite
Most-evolved
granodiorite

Table 1. Geochronology of Northern Highlands "Newer" granites. Z = zircon; MB = molybdenite; M = monazite; B = baddeleyite; T = titanite; ID-TIMS = isotope dilution thermal ionisation mass spectrometry; LA-ICP-MS = laser ablation inductively-coupled plasma mass spectrometry; SHRIMP = sensitive high resolution ion microprobe. Discussion of the timing of geodynamic events is in the text.

Granitoid	**Types**	**Emplacement timing (Ma)**	**Methodology**	**Reference**
End of the Grampian 2 orogenic event, continuation of Iapetus subduction				
Glen Dessary	Syenite; pluton	447.9±2.9	U-Pb Z ID-TIMS	Goodenough *et al.* (2011)
Graven, Shetland	Granodiorite and other granitoids; sheets	439.8±3.1	U-Pb Z LA-ICP-MS	Lancaster *et al.* (2017)
Northmaven, Shetland	Granite, granophyre, and other more mafic rocks; sheets	438.0±7.6 to 389.3±2.6	U-Pb Z LA-ICP-MS	Lancaster *et al.* (2017)
Onset of the Scandian orogenic event and dextral strike-slip faulting				
Naver Suite incl. Vagastie, Creag nan Suibheag, Creag Mhor	Granite to monzo-diorite; sheets	432.4±0.5 to 425.7±0.2	U-Pb Z ID-TIMS	Strachan *et al.* (2020)
Orkney granite complex	Granite, pegmatite, aplite; sheets	431.9±0.5 to 428.5±0.3	U-Pb Z ID-TIMS	Lundmark *et al.* (2019)
Cluanie	**Trondhjemite; pluton**	**431.9±1.7**	**U-Pb Z LA-ICP-MS**	***This study***
Assynt Alkaline Suite	Syenite and other alkaline rocks; small plutons, sheets	431.1±1.2 to 429.2±0.5	U-Pb Z ID-TIMS	Goodenough *et al.* (2011)
Slab breakoff and onset of left-lateral strike-slip faulting				
Grudie Bridge and Loch Shin	Monzogranite; stock and minor intrusions	429.9±5.2 to 427.9±2.8	Re-Os MB TIMS	Holdsworth *et al.* (2015)
Clunes	Tonalite; sheet	427.8±1.9	U-Pb Z ID-TIMS	Stewart *et al.* (2001)
Loch Loyal	Syenite and associated rocks; pluton	426±9	U-Pb Z ID-TIMS	Halliday *et al.* (1987)
Upsurge in postsubduction magmatism attributable to slab breakoff				
Strath Halladale	Ultramafic to granite; pluton	426±2	U-Pb M ID-TIMS	Kocks *et al.* (2006)
Glen Scaddle	Mafic to granite; pluton	426±3	U-Pb Z ID-TIMS	Strachan & Evans (2008)
Rogart	Ultramafic to granite; pluton	425±1.5	U-Pb Z ID-TIMS	Kocks *et al.* (2014)
Ratagain	Ultramafic to granite; pluton	425±3	U-Pb Z+B ID-TIMS	Rogers & Dunning (1991)
Strontian	Appinite to granite; pluton	425±3 423±3 418±1	U-Pb Z+T ID-TIMS	Rogers & Dunning (1991) Paterson *et al.* (1993)
Ross of Mull	Appinite to granite; pluton	418±5	U-Pb Z SHRIMP	Oliver *et al.* (2008)
Termination of the Scandian Orogenic event				
Rosemarkie	Leucogranite veins	400.8±2.6	U-Pb Z ID-TIMS	Mendum & Noble (2010)