A non-peer reviewed preprint submitted to EarthArXiv prior to submission to the Journal of the Geological Society of London in May 2022. Comments are welcomed via e-mail.

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 to the middle crust. 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 the Newer Granites. These distinct geochemical signatures indicate the tapping of batches of homogenised, evolved magma from the deeper crust, plus crustal assimilation recorded isotopically and by inherited zircons. The emplacement age of the Cluanie pluton confirms the presence of volumetrically modest subduction-related magmatic activity beneath the Northern Highlands predating slab breakoff. Crustal thickening caused by the ca. 450 Ma Grampian 2 event is proposed to have limited the extent of continental arc magmatism from ca. 450-428 Ma. Extensive new in-situ geochemical-geochronological studies for this terrane may further substantiate the deep crustal hot zone model and further substantiate the association between Caledonian magmatism and metallogenesis.

Short title: Northern Highlands Newer Granites

Keywords: Adakite, Caledonian, Geochemistry, Geochronology, Scotland

Globally, collision zone magmatism is strongly associated with metallogenesis, particularly economically important resources such as Cu, Li, Mo, Ag, Au, and rare earth elements (REEs) which are critical to our energy transition (e.g., Richards 2015). Within the British Isles, there is growing political awareness around critical metal supply (European Commission 2020), including renewed interest in Palaeozoic metal deposits associated with plutonism during the Caledonian Orogeny and its aftermath in Scotland (Rice *et al.* 2012; Walters *et al.* 2013; Spence-Jones *et al.* 2018; British Geological Survey, 2020 and associated reports). As the metallogenic potential of plutonic systems is strongly linked to magma chemistry and its geodynamic stetting, it is important to closely constrain the tectonic associations and petrogenesis of such plutons to aid exploration.

Many of the ~426-390 Ma ‘Newer Granites’ (*sensu* Read 1961) of the Scottish Caledonides are proposed to have resulted from Iapetus slab breakoff (e.g., Atherton & Ghani 2002; Neilson *et al.* 2009). However, partial melting of orogenic lower crust and/or 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 (pre- or post-breakoff), petrogenesis, and metallogenic potential are unclear. Additionally, 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 hot zones (Annen *et al.* 2005) or MASH (melting-assimilation-storage-homogenisation) processes (Hildreth & Moorbath 1988) are all key scientific advances in granite petrogenesis which have had modest application to 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 to fill some existing knowledge gaps about Newer Granite timing and petrogenesis. 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 propose that greater clarity on the nature and timing of the magmatic record as subduction gave way to collision and slab breakoff may aid with critical metal knowledge and exploration in Scotland (c.f. Richards 2015).

Regional geology

The Cluanie and Clunes plutons are located north of the Great Glen in the Northern Highlands (Figure 1a). 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 between Laurentia, Baltica and Avalonia. 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). 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). The Scandian Orogeny partly overlaps with Avalonia-Laurentia collision and concurrent Acadian events, mostly affecting 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 termed the 'Newer Granite' Suite (Read 1961). These bodies post-date the Grampian orogeny(ies), overlapping with Iapetus subduction, the Scandian event and Acadian deformation. Importantly, many such bodies are widely called '*Newer Granites*' despite their broad spectrum of compositions, ages, and potential geodynamic triggers for melting and emplacement.

A critical geodynamic event in Scotland is recorded with a phase of uplift in the Grampian Highlands at ~428 Ma, shortly followed by deposition of the Lower Old Red Sandstone and the majority of granitoid emplacement and concurrent volcanic activity (Conliffe *et al.* 2010). It is proposed that uplift and magmatism were a consequence of Iapetus slab breakoff 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 collisional style is akin to the Turkic-type orogen of Şengör & Okuroğullari (1991) recognised in Turkey, the Caucasus and Iran, where Tethyan slabs broke off in the last 10-20 Ma, yet collision, magmatism, and intra-montane sedimentation continues to the present day.

In Scotland there are few magmatic events recorded between the end of magmatism associated with the Grampian Orogeny and the emplacement of the Newer Granites (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 Newer Granite episode, have included periods of highly oblique or flat slab subduction (Oliver *et al.* 2008; Dewey *et al.* 2015), or further collisional events suppressing magmatic activity (Bird *et al.* 2013).

Thereafter, the bulk of the Newer Granite Suite in the Northern Highlands apparently crystallised from ~426-418 Ma (Oliver *et al.* 2008) (Fig. 1a; 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. A petrogenetic relationship is commonly proposed between the different facies indicating a mantle-derived origin for the whole 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 and Rb-Sr 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). 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).

The Cluanie and Clunes Plutons

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 (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).

The Cluanie pluton is comprised of porphyritic I-type trondhjemitic granodiorite with alkali feldspar megacrysts (Neill & Stephens 2009). A typical assemblage is of oscillatory-zoned plagioclase (~60 %; An_{15-30}), alkali feldspar (~15 %); quartz (~15 %), hornblende (5-10 %), and biotite (0-5 %) (Peacock *et al.* 1992). Accessories include titanite (~1%), apatite, zircon, allanite, and ilmenite. The pluton 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 mm-cm scale biotite and hornblende, with accessory titanite. The pluton is sharply cut by micro-diorite dykes, <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 belongs to the high Ba-Sr granitoid class like the Northern Highlands Newer Granites (Tarney & Jones 1994; Neill & Stephens 2009), it has $\varepsilon Nd_i > 0$ (Fowler *et al.* 2008), a high Na_2O/K_2O trondhjemitic 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, with the exception of rare cm-scale amphibole- and titanite-bearing clots and schlieren interpreted as restite (Neill & Stephens 2009). Neill and Stephens (2009) argued for the Cluanie pluton to have a geologically young amphibolitic melt source, but Fowler *et al.* (2008) placed the Cluanie pluton within their mantle-derived models of Newer Granite petrogenesis. 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. Recent revision of the ^{87}Rb decay constant re-calculates the published Rb-Sr age to ~433.5 Ma (Nebel *et al.* 2011). This age pre-dates all the Northern Highlands Newer granites, calling into question its association with the Newer Granites and importantly slab breakoff (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 ~6 km^2 Clunes tonalite outcrops just north of the Great Glen Fault Zone (GGFZ, Fig. 1b, d), with emplacement facilitated by a shear zone, related to the GGFZ, 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 plagioclase, quartz, hornblende, and biotite in varying abundances and rare patches of more granitic, granodioritic or dioritic compositions on its margins (Fig. 1d). The Clunes pluton is cut by felsic sheets thought to be part of the (undated) 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 Late Caledonian 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 at ~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). 262 spots were analysed across 59 grains, with 59 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 39 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 5 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). Unpublished work of I.L. Millar (*pers. comm.* 2013) has a similar range of ages with a weighted mean of 435.8 ± 1.8 Ma, 2σ analytical uncertainty, n =16). Our 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 slightly 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 less geologically meaningful and 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 trondhjemitic facies 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 trondhjemites 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 middle MREE-compatible minerals such as zircon, apatite, or amphibole. Moderate-low Ho/Yb_{CN} ratios (<1) in the main trondhjemite 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 K_2O and Rb relative to Th and the LREE. All samples have negative Nb-Ta and Ti anomalies but positive Zr-Hf anomalies. The less-evolved trondhjemites 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). 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 trondhjemites (Fig. 4c). The major and trace element concentrations of Clunes are very similar to the Cluanie microdiorites (Fig. 4d).

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). TTGs typically have >54 wt.% SiO_2, >15 wt.% Al_2O_3, 3-7 wt.% Na_2O, a sodic character (K_2O/Na_2O < 0.6) and low Y (<20 ppm) and HREE (Yb < 1.8 ppm) (Condie 2005; Martin *et al.* 2005; Moyen & Martin 2012). Cluanie and Clunes also 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). The origin of adakites and that of the ancient TTG suites 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. TTGs are petrographically slightly different to our samples as the former typically lack alkali feldspar megacrysts. Nevertheless, the Cluanie trondhjemites, felsic porphyrites and the Clunes tonalite to meet the geochemical parameters for TTGs above, as do all bar one of the microdiorite samples with slightly higher HREE and Y. The Cluanie pluton's very low MgO, V, Ni and Cr concentrations (Supplementary Item) make it similar to average Eoarchaean through Middle to Late Archaean TTG suites, often proposed to result from crustal melting without significant mantle input (Hastie *et al.* 2015). Clunes, with slightly higher transition metal concentrations, is most similar to Middle to Late Archaean TTG suites, which may have had more significant interaction with mantle components (e.g., Smithies 2000; Hastie *et al.* 2015). 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 Archaean TTG suites and 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 basement, but consistent with Wester Ross or Loch Ness Supergroup involvement. 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 m 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 trondhjemites 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 trondhjemite was reported by Neill & Stephens (2009), so the felsic porphyrites could be considered as a parental magma to the trondhjemites, albeit themselves 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 broadly 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. The majority of samples fall on the trend for apatite, but there may be limited roles for 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 trondhjemite, 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 TTG- or 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) FC plus minor crustal assimilation from a subduction-modified mantle source, as preferred by Fowler *et al.* (2008), or d) a more complicated petrogenetic history.

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.

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 trondhjemitic composition, lack of mafic facies, and presence of possible restite 'clots'. Additionally, the broad similarity between both plutons and TTG suites may support a model of partial melting of mafic lower crust (e.g., Condie 2005 for TTGs). 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. 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 dislodged hydrous cumulates 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) FC processes vs option d) 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, long before the accepted onset of Newer Granite magmatism. Magmatic addition during the period ~450 to ~430 Ma rules out option c), as FC of a singular batch of magma cannot simply explain Cluanie's zircon growth history. The zircon history is more 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 in the deep crust. Modelling and experimental work demonstrates that an andesitic magma of >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 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 more varied facies and geochemical ranges of other Newer Granites. The adakite-like geochemistry of Cluanie and Clunes does however contrast with the more potassic and REE-enriched geochemistry of the contemporaneous 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 lower 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 pluton to show that the Great Glen Fault was undergoing left-lateral motion ~428 Ma ago. However, Cluanie is slightly older than the Clunes tonalite. Neill & Stephens (2009) suggested 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 a new minimum age for strike-slip faulting in the Northern Highlands and importantly a switch from early dextral to subsequent sinistral motion of the GGFZ and associated faults shortly after emplacement between ~432 and ~430 Ma (Holdsworth *et al.* 2015). Further research into this link between early dextral motion of the GGFZ and magmatism in areas adjacent to the Great Glen will be detailed in a subsequent communication.

Above, we have followed the popular interpretation that slab break-off occurred at ~428 Ma beneath Scotland, and that Cluanie, the Assynt Alkaline Suite, and the granitoids associated with the Naver Thrust were emplaced during the last stages of Iapetus subduction. 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? 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 the Avalonia-Laurentia collision at ~425 Ma becomes influential. However, "peak" Scandian metamorphism is dated to ~425 Ma from U-Pb zircon dating of East Sutherland migmatites (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. The greatest volume of Newer Granite magmatism clearly occurs after ~426 Ma in the Northern Highlands (Oliver *et al.* 2008), so the modest volumes identified before this time does strongly a distinct geodynamic regime in both the Grampian and Northern Highlands which switched at ~428 Ma. Therefore, we conclude that a diachronous slab breakoff is not likely and our data supports the prevailing hypothesis that Northern Highlands magmatism prior to ~428 Ma reflects supra-subduction activity. Our data does 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 crust being present prior to the Scandian episode.

Implications for future geochronological and metallogenic research in Scotland

The above data are among very few LA-ICP-MS U-Pb zircon results for Caledonian plutons. The existing geochronological framework for Caledonian magmatism has been constructed from multiple approaches. Caution is therefore required in terms of interpretation of published ages, including whether we assign plutons to pre- and post- breakoff settings, and how we therefore assess their metallogenic potential (Vos *et al.* 2007). Zircon chemical- or air-abrasion isotope dilution U-Pb thermal ionisation mass spectrometry (CA/AA-ID-TIMS) has been commonplace in the Northern Highlands, often without published 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 various terranes (Table 1; Brook 1985; Conliffe *et al.* 2010; Holdsworth *et al.* 2015; Rogers & Dunning 1991). Ion probe studies in Acadian granitoids in England and Southern Scotland have already shown distinct zircon populations in several plutons, covering a wider (and younger) range of dates compared to regular CA-ID-TIMS methods (e.g., Miles *et al.* 2014; Miles & Woodcock 2018; Woodcock *et al.* 2019). Oliver *et al.* (2008) included age ranges of up to 30 Ma 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 be interpreted as evidence for recycling of antecrystic zircon populations from a deep crustal hot zone, as included in the geodynamic interpretation of Oliver *et al.* (2008).

Such complexities are not clearly shown in the ID-TIMS studies of Northern Highlands (e.g., Rogers and 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 the formerly-mined Strontian pluton may be enhanced with such additional analysis. Lawrence *et al.* (2022; in review) 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 future high-precision ID-TIMS studies may be significantly enhanced through a combination of preliminary LA-ICP-MS or ion probe work, more routine cathodoluminescence imaging of selected half-grains, and improved abrasion techniques to resolve multiple events (e.g., Gaynor et al., 2022).

The Northern and Grampian Highlands have only a limited history of metal resource exploitation, notably Au-Ag at Cononish and Pb-Zn at Strontian. As global resource recovery targets ever more marginal locations, and as environmentally-sustainable biological extraction or remediation systems come on stream, the Newer Granites may yet provide local sources of various critical metals such as the REE (e.g., Walters *et al.* 2013). Cluanie and Clunes plutons were not enriched in economically important metalliferous fluids, save for a single dm-scale baryte vein at Cluanie (Peacock 1992). This observation is at odds with the global association of adakite-like magmas with porphyry mineralisation (e.g., Kepezhinskas *et al.* 2022). In light of the new geochronology for Cluanie, it seems the barren nature of these plutons is probably related to its mid-crustal emplacement prior to slab breakoff. More substantive mantle-derived magmatic activity, stirring of the deep crustal hot zone and enhanced hydrothermal activity in Scotland after breakoff is likely to have promoted some mineralisation in younger bodies (Vos *et al.* 2007).

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, 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. With an interest in critical metal exploration, the association of Caledonian granites with geodynamic events such as slab breakoff need clarification.

Acknowledgements

E.M. was funded by an Angus Mitchell MSc by Research scholarship at the University of Glasgow (2017), the Alwyn Williams fund of the same (2018), and by the Inverness Field Club (2019). I.N. was funded by a NERC PhD studentship [NE/F00219X/1] (2007-2011). George Strachan introduced I.N. to the pluton in 2003. Preparation and analysis steps were assisted by Ley Woolley of Cardiff University and Peter Chung, Robert MacDonald, Maria Olivia Torcea and Amy Riches, all lately of the University of Glasgow.

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 and editing. E.D.D. – interpretation and editing. V.O. – analysis. N.O. – analysis. E.C.W. – analysis.

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Supplementary Items

An Excel Workbook containing three 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 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.

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)
Approximate timing of 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)
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)

Figure 1. a) Scottish map after Lancaster *et al.* (2017) and Lundmark *et al.* (2019). b) Satellite image of key Northern Highlands faults and plutons near Cluanie. Data from: https://bing.com/maps and Edina Digimap https://digimap.edina.ac.uk/roam/map/geology. c) Map of the Cluanie pluton after Neill & Stephens (2009). d) Map of the Clunes tonalite after Stewart et al. (2001).

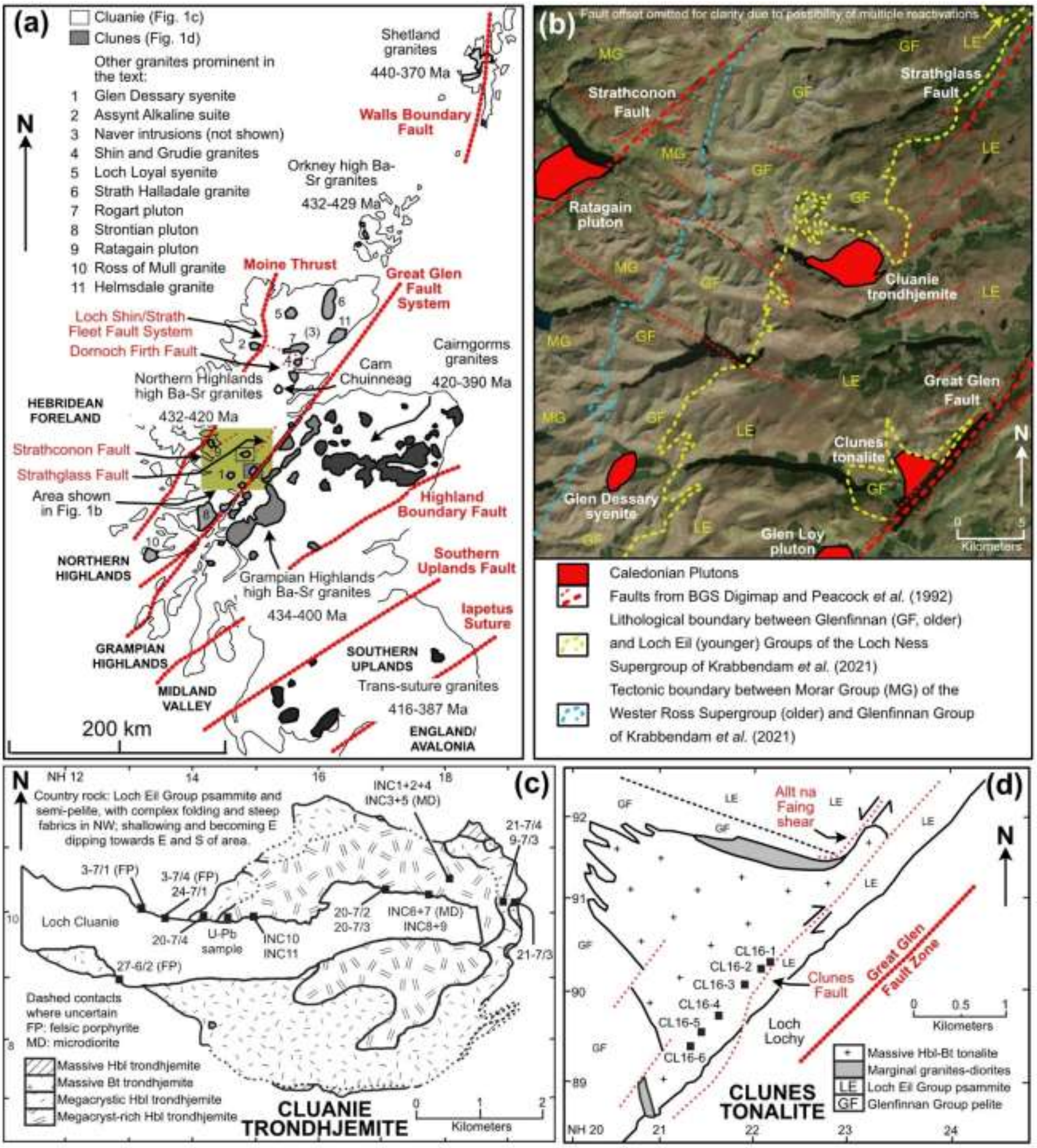

Figure 2. U-Pb data for the Cluanie pluton. For full data see Supplementary Item. a) Zircon inheritance record for the Cluanie pluton with a Wetherill concordia plot and examples of typical zircon textures. b) Kernel density plot of the same data highlighting key events in the Scottish geological record. c) Wetherill concordia plot showing all concordant Late Caledonian analyses and example of a typical zircon texture. d) Concordant Late Caledonian/post-Grampian 2 analyses ordered by age, showing weighted mean ages and confidence intervals for proposed deep crustal hot zone and emplacement-related clusters.

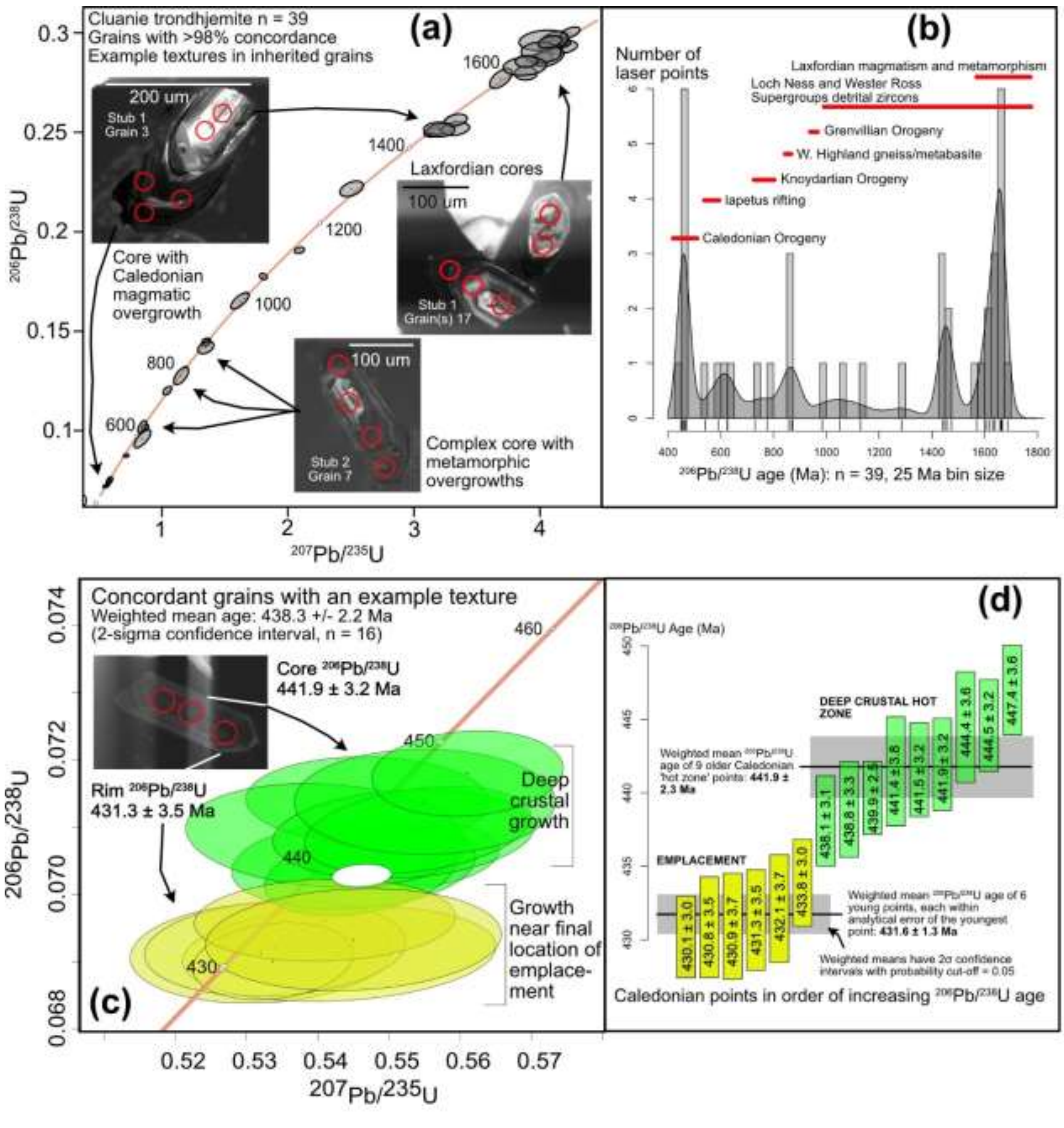

Figure 3. Major and trace element variation diagrams for the Cluanie pluton and Clunes tonalite.

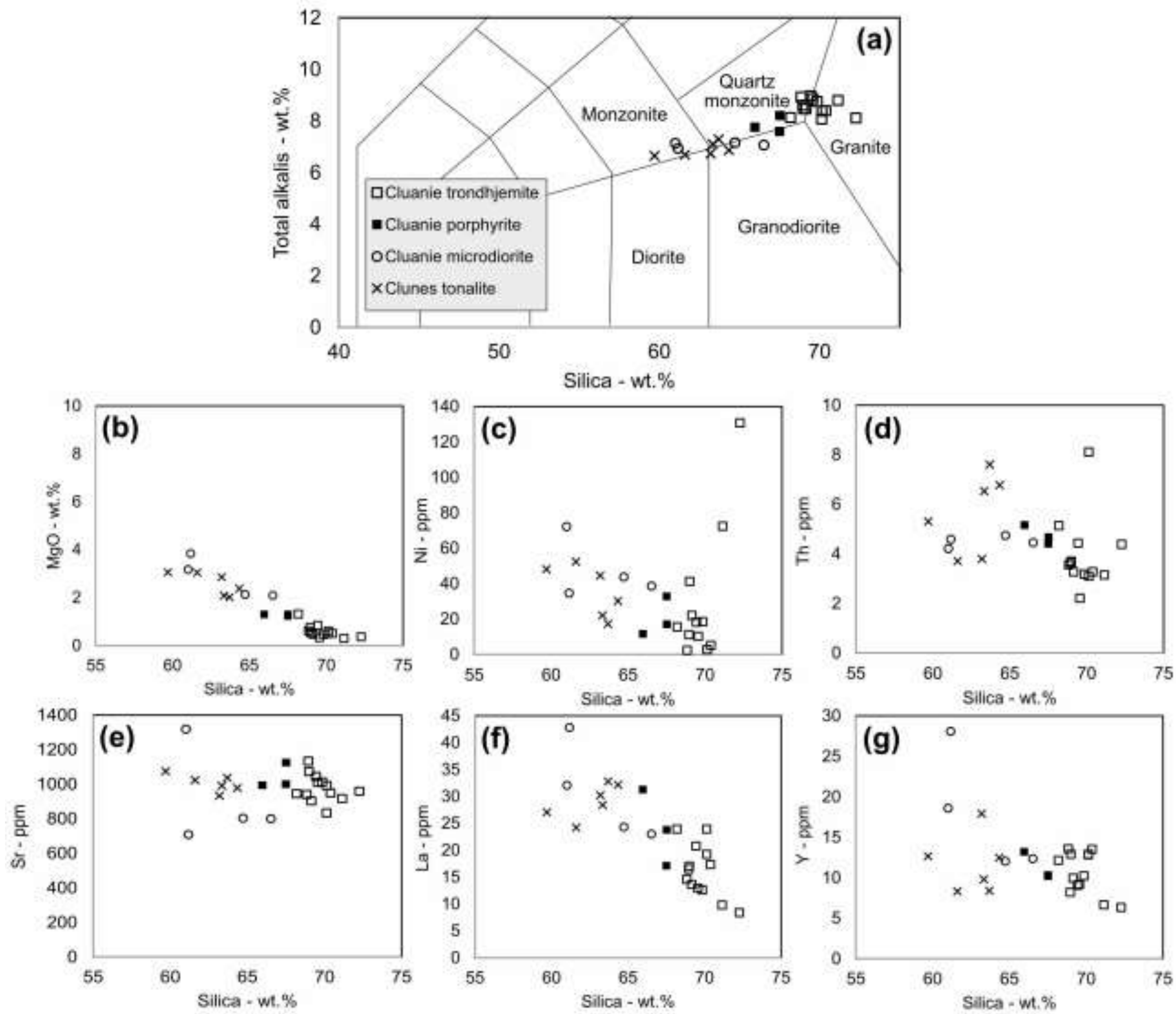

Figure 4. Chondrite- (McDonough & Sun 1995) and primitive mantle- (Sun & McDonough 1989) normalised plots for the Cluanie pluton and Clunes tonalite.

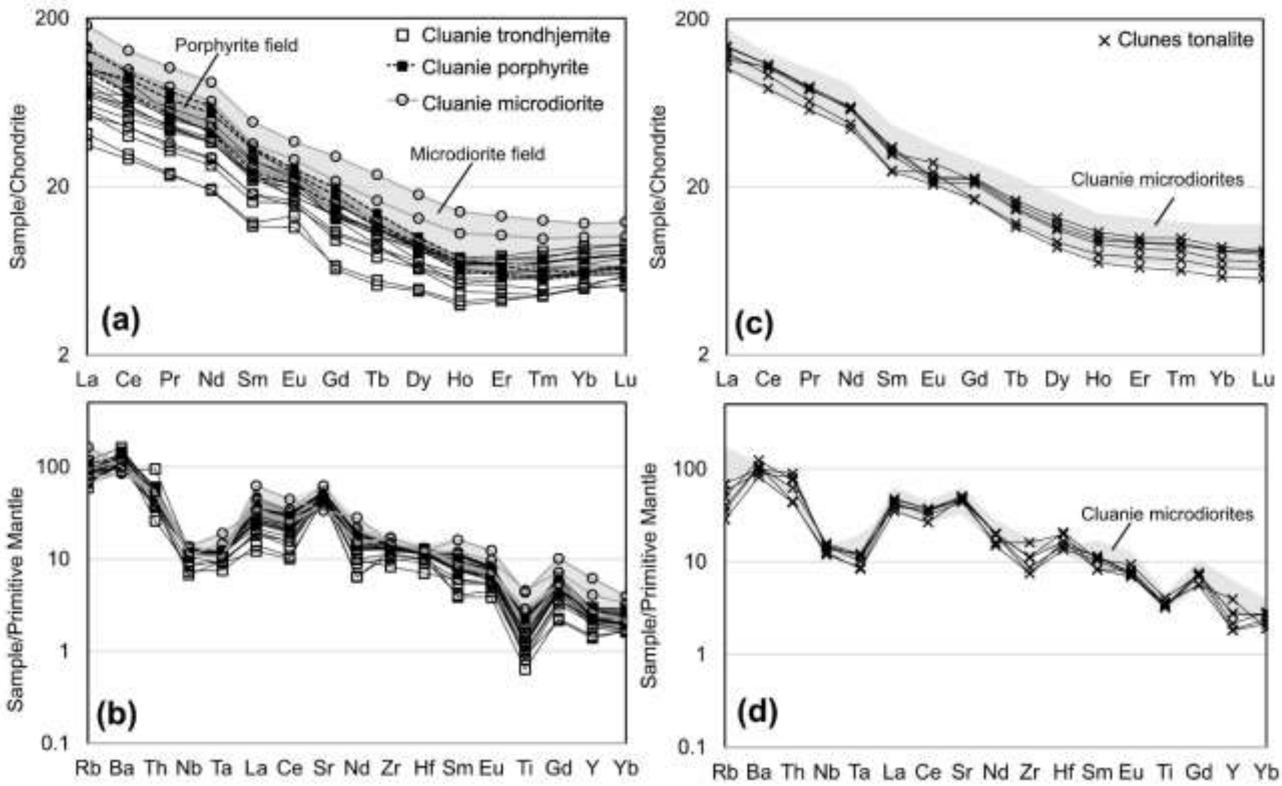

Figure 5. a-c) Adakite geochemical classification diagrams based on Martin (1999), with data from Fowler et al. (2008) for the Northern Highlands; d) Radiogenic isotope data for the Northern Highlands plutons from Fowler et al. (2008) including new results for the Cluanie pluton.

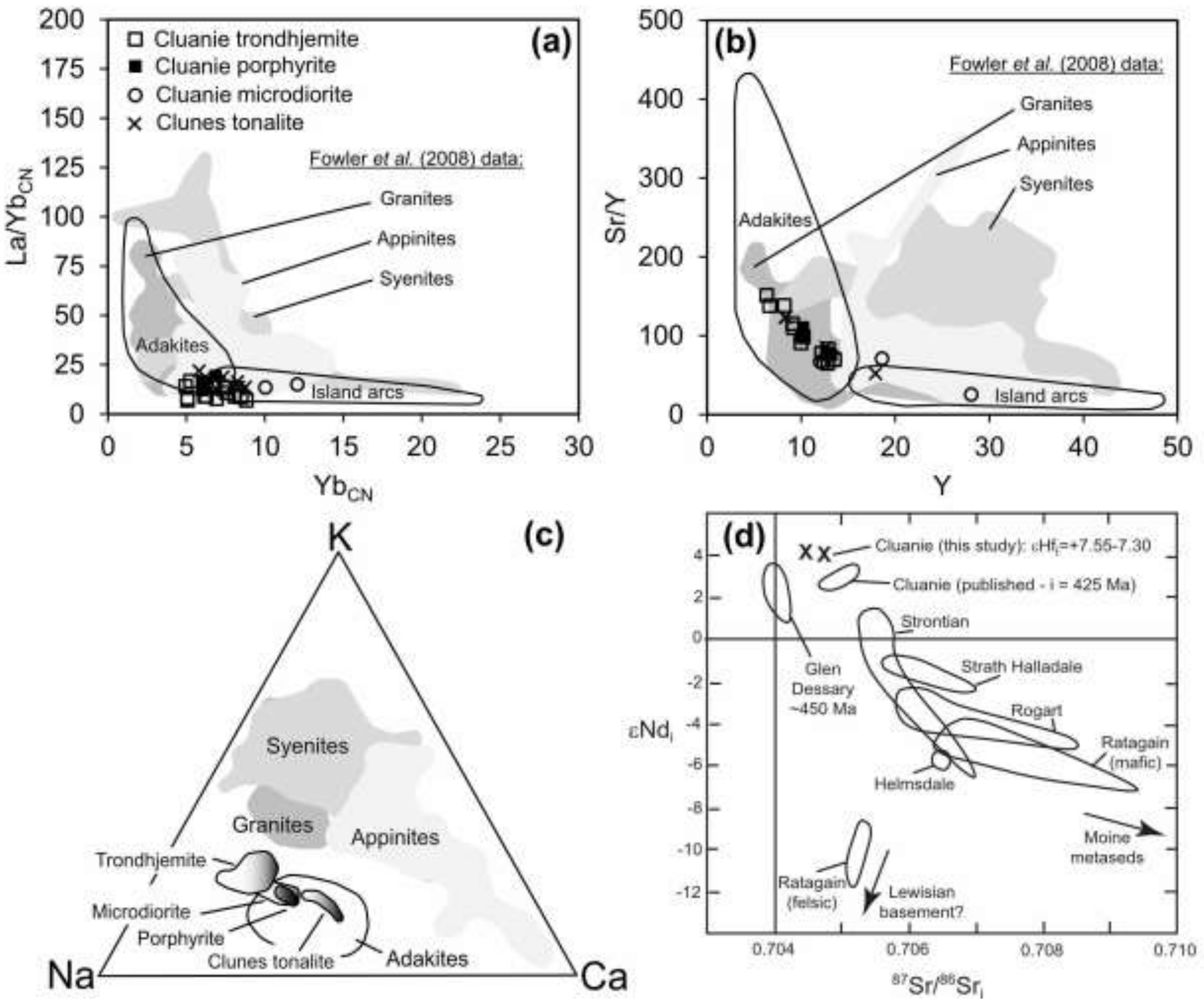

Figure 6. Trace element modelling of fractional crystallisation in the Cluanie pluton from a starting composition of felsic porphyrite minor intrusion IN/27-6/2. (a) La vs. Rb vector plot showing the strong influence of accessory minerals on the composition of the Cluanie pluton samples. (b) La vs. Yb vector plot. (c) Modelling of Rayleigh fractional crystallisation of the REE. See the Supplementary Item for modelling parameters.