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# *Testing petrogenetic models for contemporaneous mafic and felsic to intermediate magmatism within the “Newer Granite” suite of the Scottish and Irish Caledonides*

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## ABSTRACT

**Granitoid batholiths dominated by felsic to intermediate compositions are commonly associated with mafic plutons and enclaves; however, the genetic relationship between the apparently coeval but compositionally dissimilar magmas is unclear. Here, we reviewed the age and lithochemical and Nd-Sr isotopic compositions of some classic plutonic rocks emplaced in the Northern Highlands, Grampian and Connemara terranes of the Caledonide orogen of Scotland and Ireland. The Northern Highlands terrane consists mostly of Neoproterozoic metasedimentary rocks of the Moine Supergroup and is located north of the Great Glen fault. The Grampian terrane also consists of Neoproterozoic metasedimentary rocks (Dalradian Supergroup) and is located south of the Great Glen fault in both Scotland and Ireland. Amphibolite-facies metasedimentary rocks in the Connemara terrane are correlated with the Dalradian Supergroup, and the terrane is bounded by splays of the Highland Boundary and Southern Uplands faults. These three terranes were intruded by Silurian–Devonian mafic and felsic to intermediate plutonic rocks that display field evidence for mingling and mixing and have a similar range (between ca. 437 and 370 Ma) in emplacement ages. This range implies they were intruded during and after the late Caledonian Scandian orogenic event that resulted from the mid- to late Silurian collision of amalgamated Avalonia and Baltica with Laurentia and the final closure of the Iapetus Ocean. Our review supports the contention that the Great Glen fault represents a major compositional boundary in the Silurian lithosphere. Felsic to intermediate plutons that occur north of the Great Glen fault are more enriched in light rare**

earth elements and Ba-Sr-K compared to those to the south. Isotopic compositions of these late Caledonian plutonic rocks on both sides of the Great Glen fault indicate that metasomatism and enrichment of the subcontinental lithospheric mantle beneath the Northern Highlands terrane occurred just prior to emplacement of late Caledonian plutons. Within the same terrane, mafic and felsic to intermediate rocks display similar trace-element and rare earth element concentrations compatible with models implying that fractionation of a mafic magma played an important role in generating the felsic to intermediate magmas. The onset of slab failure magmatism may have been diachronous along the length of the collision zone. If so, slab failure may have propagated laterally, possibly initiating where promontories collided.

## INTRODUCTION

Many parts of the Lower Paleozoic Caledonian orogen of Scotland and Ireland are intruded by large, relatively undeformed granitoid bodies, which are spatially and temporally associated with mafic to ultramafic plutons, including a distinct suite of volumetrically subordinate, hornblende-rich intrusions termed appinites. Caledonian appinites are commonly associated with coeval lamprophyre dikes, and both preferentially occur along the periphery of the more voluminous granitoid plutons (Atherton and Ghani, 2002). Although some researchers propose a genetic relationship between the appinite suite, the lamprophyre dikes, and the granitoid batholiths (e.g., Pitcher and Berger, 1972; Fowler, 1988; Murphy et al., 2019), the nature of this relationship is unresolved and the subject of this contribution.

The Caledonian orogeny resulted from the closure of the Iapetus Ocean and the collision of three continental blocks, Laurentia, Baltica, and combined Ganderia-Avalonia (Fig. 1; Soper and Hutton, 1984; Pickering et al., 1988; Soper et al., 1992). In Britain and Ireland, the Iapetus suture divides fault-bounded terranes to the northwest that originated either on or along the periphery of Laurentia from those to the southeast, such as Ganderia and

Avalonia, that resided along the Gondwanan margin in the late Neoproterozoic (Figs. 1 and 2; Bluck et al., 1992; van Staal et al., 2012). In northwesternmost Scotland, the Hebridean terrane (Fig. 2) represents the foreland of the Caledonian orogen (Chew and Strachan, 2014). The structurally overlying Northern Highland and Grampian terranes represent the margin of Laurentia (Fig. 2). The Highland Boundary fault (Fig. 2) delimits Laurentian terranes to the northwest from oceanic terranes to the southeast. The Midland Valley terrane is largely overlain by Upper Paleozoic cover successions that are interpreted to unconformably overlie an Ordovician–Silurian magmatic arc assemblage (Badenzki et al., 2019). The Southern Uplands terrane comprises a Late Ordovician–Silurian accretionary prism (Leggett et al., 1979; Anderson and Oliver, 1986; Stone and Merriman, 2004).

The term “Caledonian orogeny” is a collective term used to refer to a number of distinct tectonic events that occurred between the Ordovician and the Devonian (McKerrow et al., 2000). In Scotland and Ireland, it comprises two main orogenic events: the Ordovician Grampian orogenic event and the Silurian to Devonian Scandian orogenic event (Chew and Strachan, 2014). Although alternative models exist (e.g., Searle et al., 2019), most models agree that the Grampian orogenic event resulted from the

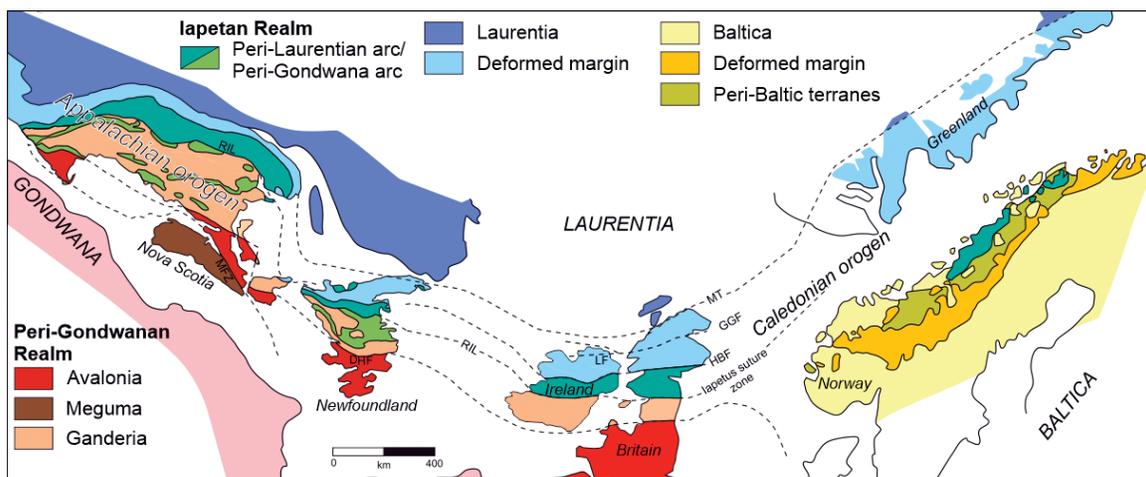


Figure 1. Paleogeographic reconstruction of the Appalachian-Caledonian orogen (modified after Pollock et al., 2012; Waldron et al., 2014). Abbreviations: DHF—Dover–Hermitage Bay fault; GGF—Great Glen fault; HBF—Highland Boundary fault; LF—Leannan fault; MFZ—Minas fault zone; MT—Moine thrust; RIL—Red Indian Line.

collision of the Lough Nafooye–Midland Valley island arc with the Laurentian margin during initial stages of ocean closure (e.g., Dewey and Shackleton, 1984; Ryan and Dewey, 1991; Chew et al., 2010; Chew and Strachan, 2014; Johnson et al., 2017; Dunk et al., 2019). These models invoke southward-dipping subduction

(present coordinates) away from Laurentia and ophiolite obduction and arc-continent collision followed by subduction polarity reversal in the Late Ordovician, resulting in a new subduction zone dipping to the northwest beneath Laurentia and the accreted arc (Dewey and Shackleton, 1984; Dewey and Mange, 1999).

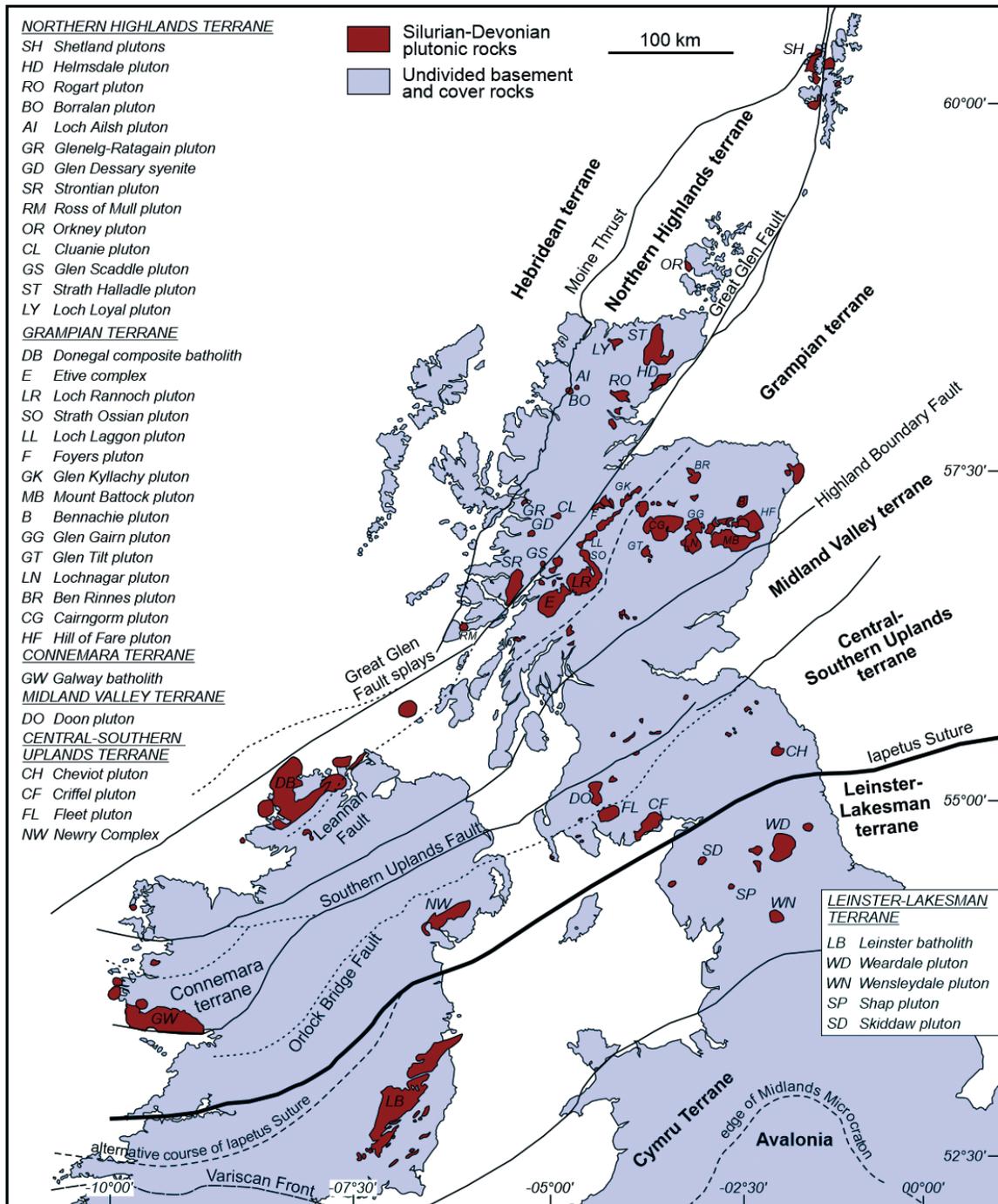


Figure 2. Terrane map of northern Britain and Ireland showing the locations of Silurian–Devonian plutonic rocks, modified from Chew and Strachan (2014), Dewey et al. (2015), and Miles et al. (2016). The location of the Moine thrust zone with respect to the Loch Borralan and Loch Ailsh plutons is after Dewey et al. (2015) and Fox and Searle (2021).

Independent evidence for the subduction polarity reversal is provided by the internal structure of the overlying Southern Uplands accretionary prism, and the NW dip of prominent mantle reflectors located along the Iapetus suture (Klemperer and Matthews, 1987). The Silurian Scandian orogenic event resulted from the mid- to late Silurian collision of a previously amalgamated Avalonia and Baltica with Laurentia and the final closure of the Iapetus Ocean (Coward, 1990; Dallmeyer *et al.*, 2001; Kinny *et al.*, 2003; Strachan *et al.*, 2020a). The overall tectonic environment from ca. 420–415 Ma onward was likely dominated by strike-slip faulting, because the Great Glen fault (Fig. 2) appears to truncate Caledonian thrusts (Snyder and Flack, 1990). An estimated 700–500 km of sinistral displacement along the Great Glen fault juxtaposed the Northern Highland and Grampian terranes by Early Devonian time (Dewey and Strachan, 2003).

The granitoids and associated mafic to ultramafic rocks belong to the “Newer Granite” suite, defined by Read (1961) to distinguish relatively undeformed Silurian–Devonian intrusive rocks from the “Older Granite” suite of mainly foliated plutons that were emplaced prior to, and during the early stages of, the Caledonian orogeny (Lambert and McKerrow, 1976; Oliver *et al.*, 2008). Here, we refer to the “Newer” granitoids and associated mafic rocks collectively as late Caledonian plutonic rocks.

The emplacement ages of late Caledonian plutonic rocks are constrained between ca. 437 and 370 Ma, but the majority were emplaced between ca. 425 and 415 Ma (Rogers and Dunning, 1991; Oliver *et al.*, 2008; Neilson *et al.*, 2009; Goodenough *et al.*, 2011; Lancaster *et al.*, 2017; Archibald *et al.*, 2021). In the Northern Highlands terrane, shallowly dipping, sheeted granitic plutons were emplaced during Scandian thrusting (e.g., Strath Halladale, ca. 426 Ma; Kocks *et al.*, 2006). In both the Northern Highlands and Grampian terranes, thrusting was followed by emplacement of steep-sided plutons (e.g., Strontian, Donegal, Rogart; Fig. 2) that are thought to have commonly exploited steep, strike-slip shear zones and faults (e.g., Hutton, 1982; Jacques and Reavy, 1994; Hutton and Alsop, 1995, 1996; Price, 1997; Stevenson *et al.*, 2006; Kocks *et al.*, 2014). Because pluton emplacement continued over a relatively restricted time range and mainly after Scandian ductile deformation and metamorphism, Atherton and Ghani (2002) interpreted the late Silurian to Early Devonian magmatism to be the result of “slab break-off” following Iapetus Ocean closure, an interpretation supported by more recent studies (Fowler *et al.*, 2008; Neilson *et al.*, 2009; Cooper *et al.*, 2013; Murphy *et al.*, 2019; Archibald and Murphy, 2021). According to this interpretation, rapid uplift following slab failure resulted in unroofing of late Caledonian plutons by the Early Devonian, and this was accompanied by deposition of siliciclastic sediments into extensional or transtensional basins (Atherton and Ghani, 2002; Brown *et al.*, 2008). Granite clasts in the Lower Devonian (Lochkovian) Crawton Group in the Midland Valley terrane in Scotland have lithochemical compositions that are similar to exposed late Caledonian granite plutons of the same age (Haughton and Halliday, 1991), supporting the model of rapid exhumation and exposure of granite plutons in the Early Devonian.

Despite these constraints, the genetic relationships between coeval mafic and felsic magmas within the orogen remain enigmatic. Although most studies invoke slab failure (also known as slab break-off) as the tectonic driver for the late Caledonian plutons (e.g., Atherton and Ghani, 2002; Neilson *et al.*, 2009; Cooper *et al.*, 2013; Miles *et al.*, 2016; Archibald and Murphy, 2021), Oliver *et al.* (2008) proposed an earlier phase of arc magmatism between ca. 430 and 420 Ma for Scottish granitoids. Field relationships show macroscopic mixing and mingling textures between appinites and coeval granitoid rocks (Fowler, 1988). Variations in the trace-element and isotopic compositions of the appinites that are mirrored by the coeval granitoid rocks are interpreted to reflect fractionation of mafic magmas to produce the felsic to intermediate magmas (Fowler and Henney, 1996; Fowler *et al.*, 2001, 2008). However, alternative models (Atherton and Ghani, 2002) suggest that mafic underplating may have advected heat into the crust, inducing anatexis and the production of the granitoid magmas. Other models suggest that the compositional diversity of various plutons results from partial melting of a mantle enriched in Ba-Sr and mixing and mingling with heterogeneous mafic to intermediate lower crust (Harmon *et al.*, 1984; Frost and O’Nions, 1985; Neilson *et al.*, 2009). Recent models for generating slab-failure magmas (Hildebrand *et al.*, 2018) instead invoke melting of the upper basaltic-gabbroic portions of the detached oceanic lithosphere and subsequent modification of the rising magmas by fractional melting of old and enriched subcontinental lithospheric mantle.

In this study, we reviewed U-Pb geochronological, lithochemical, and Nd-Sr isotopic data for late Caledonian plutonic rocks in northwestern Ireland and northern Scotland to evaluate the genesis of coeval mafic to ultramafic and felsic to intermediate rocks. To investigate possible variations in the potential source rocks for contemporaneous mafic and felsic to intermediate magmas, we focused on the geochemical and isotopic compositions of late Caledonian plutonic rocks located on opposite sides of the Great Glen fault and its splays (Fig. 2) in the Scottish Northern Highlands, Irish Grampian, Scottish Grampian, and Irish Connemara terranes, which have similar host metasedimentary rocks (Moine and Dalradian supergroups). On geochemical and isotopic plots, we subdivided the data geographically (Scotland and Ireland) and based on geological terrane (north or south of the Great Glen fault and its fault splays) to investigate variations in the potential source regions for contemporaneous mafic and felsic to intermediate magmas. We discuss the implications of alternative models for the genetic relationship between mafic and felsic to intermediate magmas during the slab-failure process.

## GEOLOGICAL CONTEXT

We review briefly the lithological characteristics and tectonic evolution of late Caledonian plutonic rocks emplaced into the Northern Highlands and Grampian terranes in Scotland, and the Grampian and Connemara terranes in Ireland (Fig. 2), as well as the country rocks into which these plutons were emplaced.

### Northern Highlands Terrane

The Northern Highlands terrane is bound by the Moine thrust to the northwest and the Great Glen fault to the southeast (Fig. 2). The terrane consists mostly of early Neoproterozoic metapsammite and metapelitic rocks, which are known collectively as the Moine Supergroup (Chew and Strachan, 2014), but which may in fact comprise two separate successions separated by a Tonian orogenic unconformity (Bird et al., 2018; Krabbendam et al., 2022). Local basement inliers of Archean to Paleoproterozoic orthogneiss have generally been correlated with the Lewisian basement of the Caledonian foreland but may constitute a separate basement terrane (Strachan et al., 2020b). The sedimentary precursors of the Moine successions were deposited along the eastern margin of Laurentia between ca. 1000 Ma, the age of the youngest detrital zircon grains, and ca. 870 Ma, the age of the oldest intrusive igneous rocks in the Northern Highlands terrane (Friend et al., 1997, 2003). Geochronological data indicate a complex history of polyphase metamorphism and deformation. Tonian (ca. 950–725 Ma) orogenic events likely resulted from accretionary tectonics along the Rodinian margin of eastern Laurentia (Cawood et al., 2010, 2015; Bird et al., 2018). The Moine rocks were reworked during the Caledonian orogeny in both the Grampian and Scandian events (Cutts et al., 2010; Bird et al., 2013; Cawood et al., 2015; Mako et al., 2019; Strachan et al., 2020a). The Northern Highlands terrane has no equivalent exposed in Ireland (Fig. 2).

### Grampian Terrane

The Grampian terrane in both Scotland and Ireland lies between the Great Glen fault and the Highland Boundary fault (Fig. 2). Most of the Grampian terrane is underlain by rocks of the Dalradian Supergroup, which comprises a sequence of lithologically diverse metasedimentary and mafic metavolcanic rocks that includes three distinct glaciogenic units correlated with widespread Neoproterozoic glaciations (Condon and Prave, 2000; McCay et al., 2006). Basement rocks to the metamorphosed cover include the Palaeoproterozoic Annagh Gneiss Complex in northwestern Ireland (Daly, 1996) and the Rhinns Complex in southwest Scotland (Muir et al., 1992). Metasedimentary rocks of possible Moine affinity and known as the Badenoch Group underlie the Dalradian rocks in the northern Grampian Highlands (Chew and Strachan, 2014). Rocks of the Dalradian Supergroup were deposited on the eastern margin of Laurentia between ca. 750 Ma and 510 Ma (Smith et al., 1999; Tanner and Sutherland, 2007). The Dalradian Supergroup underwent widespread ductile deformation and greenschist- to amphibolite-facies metamorphism between ca. 475 and 465 Ma during the Grampian orogeny (Friedrich et al., 1999; Soper et al., 1999; Flowerdew et al., 2000; Oliver et al., 2000; Baxter et al., 2002; Carty et al., 2012; Bird et al., 2013; Viete and Lister, 2017; Walker et al., 2021). Termination of the Grampian orogeny by ca. 460–455 Ma is indicated by a cluster of mineral cooling ages (Flowerdew et al., 2000; Viete et

al., 2013) and by the emplacement of posttectonic crustal melts (Flowerdew et al., 2000; Oliver et al., 2008).

### Connemara Terrane

The Connemara terrane is exposed only in western Ireland between splays of the Highland Boundary fault to the north and the Southern Uplands fault to the south (Fig. 2). It consists of ca. 475–463 Ma metagabbro and orthogneiss that intruded amphibolite-facies rocks correlated with the Dalradian Supergroup in the northern part of the terrane and mainly greenschist-facies rocks of the South Connemara Group to the south (Williams et al., 1988; Leake, 1989; Friedrich et al., 1999). The Dalradian Supergroup-equivalent rocks were thought to have been detached by strike-slip faulting from the margin of Laurentia at the end of the Grampian orogenic event and emplaced into their current, apparently anomalous, location south of the Lough Nafooy (= Midland Valley) arc (Fig. 2; Dewey and Shackleton, 1984; Hutton and Dewey, 1986). An alternative hypothesis is that the Connemara terrane is more or less in situ and was overthrust by the Lough Nafooy arc during the Grampian orogeny (Dewey and Ryan, 2015).

### Ordovician Magmatism in the Grampian and Connemara Terranes

Ordovician magmatism occurred mostly between ca. 470 and 460 Ma, although Johnson et al. (2017) obtained ca. 490 Ma U-Pb zircon ages from diorite and gabbro in the Scottish Grampian terrane. The Ordovician plutonic rocks are located in the northeast Grampian Highlands in Scotland and in the Connemara terrane (Chew and Strachan, 2014). Magmatic activity is recorded by mafic and intermediate intrusive rocks interpreted to represent components of a volcanic arc in the Connemara terrane (e.g., Leake, 1989; Tanner, 1990; Yardley et al., 1982). Mafic intrusions in the Connemara terrane yielded U-Pb zircon ages of  $470.1 \pm 1.4$  Ma and  $474.5 \pm 1$  Ma, and the posttectonic Oughterard granite yielded an age of  $462.5 \pm 1.2$  Ma (Friedrich et al., 1999). Mafic magmatism in northeast Scotland yielded similar ages (ca. 470 Ma; e.g., Carty et al., 2012). Oliver et al. (2008) reported 470 Ma ages for foliated granitoid rocks, but unlike plutonic rocks in the Connemara terrane, they are peraluminous two-mica granitoid rocks. Emplacement was coeval with regional peak metamorphism, constrained by Sm-Nd and Lu-Hf metamorphic garnet ages to between ca. 473 and 463 Ma in the Scottish Highlands (Oliver et al., 2000; Baxter et al., 2002; Bird et al., 2013) and ca. 460 Ma in the Irish Dalradian equivalents (Flowerdew et al., 2000). Thus, magmatism occurred between ca. 490 and 460 Ma, and Grampian metamorphism peaked ca. 475–465 Ma.

### Late Caledonian Mafic Rocks

The mafic and ultramafic rocks primarily occur as appinitic plutons, as microdioritic and lamprophyre dikes adjacent to granitoid batholiths, and as discrete facies and enclaves within

the granitoid plutons (Fig. 3; Pitcher and Berger, 1972; Rock *et al.*, 1986; Fowler, 1988; Murphy *et al.*, 2019). Rare gabbro-dioritic plutons (e.g., Glen Scaddle; Fig. 2), with no obvious spatial association with granitoids, occur in the Northern Highlands terrane.

The appinites are medium- to coarse-grained melanocratic rocks composed of large hornblende crystals in a matrix of feldspar and minor quartz, titanite, and zircon (e.g., Pitcher and Berger, 1972; Wright and Bowes, 1979; Fowler, 1988; Murphy *et al.*, 2019). The appinites are small bodies (typically <1 km in diameter) compared to the larger plutons and batholiths that they commonly border. Appinites intruded and were intruded by 1–10-m-wide porphyritic lamprophyre dikes that contain phenocrysts of olivine, phlogopite, hornblende, and clinopyroxene (Rock *et al.*, 1986; Elsdon and Todd, 1989; Murphy *et al.*, 2019). Lamprophyre dikes and sills also intruded the Dalradian metasedimentary rocks in the Irish Grampian terrane and have phenocrysts of hornblende and phlogopite with lesser amounts of clinopyroxene and olivine in a groundmass of plagioclase, biotite, pyroxene, and amphibole (Pitcher and Berger, 1972; Murphy *et al.*, 2019). Canning *et al.* (1996, 1998) described the lamprophyres in Scotland as porphyritic mica-lamprophyres with phenocrysts of olivine, clinopyroxene, and biotite-phlogopite, hosted in a felsic groundmass of K-feldspar and plagioclase. Hornblende lamprophyres are also porphyritic but with hornblende, clinopyroxene, and olivine as the main phases (Canning *et al.*, 1998). The lamprophyres commonly occur close to granitoid plutons (Pitcher and Berger, 1972; Fowler *et al.*, 2008).

In both the Northern Highlands and Grampian terranes, mafic enclaves within granitoid plutons are highly variable, with irregular, but mostly oblate, shapes that vary from a few millimeters to meters across (Figs. 3A, 3E, and 3F). Some enclaves are disaggregated and are likely the product of mafic magma mingling with more intermediate to felsic magmas (Figs. 3H–3K). Most of the mafic enclaves consist of biotite and amphibole partially replaced by chlorite and actinolite, with subordinate quartz, microcline, titanite, clinopyroxene, epidote, zircon, apatite, pyrite, and titanomagnetite. Some mafic enclaves contain plagioclase phenocrysts (e.g., Fig. 3E).

The contacts of the mafic enclaves with their host granitoids are generally irregular and cusped (Figs. 3C and 3G), which suggests that the mafic and felsic magmas coexisted during their emplacement, a relationship also supported in Donegal by the coeval U-Pb zircon ages of plutons and enclaves (Archibald *et al.*, 2021). However, in some locations, the granitoid magmatism clearly postdated the emplacement of appinite, as shown by the observation of angular appinite fragments (xenoliths) entrained in granitoid dikes and by the occurrence of narrow granite dikes that intrude the appinite (Fig. 3B).

### Late Caledonian Felsic to Intermediate Rocks

Late Caledonian felsic to intermediate rocks occur as dikes, plutons, and nested intrusions typically ranging from 0.5 to

10 km in width that coalesced to form composite batholiths (e.g., Donegal composite batholith) in Scotland and northwestern Ireland (Figs. 2 and 3). In the Grampian terrane, the felsic to intermediate magmas intruded mostly rocks of the Dalradian Supergroup (Pitcher and Berger, 1972; Neilson, 2008; Appleby *et al.*, 2010). In the Northern Highlands terrane, late Caledonian plutonic rocks intruded rocks of the Moine Supergroup (Fowler *et al.*, 2008). In the Connemara terrane, the Galway Granite Batholith intruded Ordovician metagabbro and gneiss (Friedrich *et al.*, 1999) as well as Lower Ordovician greenschist-facies metasedimentary and metavolcanic rocks (Williams *et al.*, 1988; Feely *et al.*, 2003).

These batholiths are composed of mostly medium- to coarse-grained quartz monzonite to granodiorite and typically K-feldspar porphyritic plutons. Some plutons display broad, concentric compositional and mineralogical zoning, with a quartz monzodiorite exterior grading inward to a granodiorite interior (e.g., Ardara pluton of the Donegal composite batholith; Pitcher and Berger, 1972). Felsic to intermediate rocks display a wide range in composition and texture across all three terranes. Although modal abundances vary significantly, most late Caledonian granitoid rocks are dominated by plagioclase, biotite, amphibole, microcline, and quartz, with accessory titanite, epidote, apatite, zircon, pyrite, and magnetite (see Pitcher and Berger, 1972). Some plutonic rocks younger than ca. 415 Ma, particularly in the Donegal composite batholith, have silica concentrations >70% and are peraluminous with magmatic muscovite and garnet (Ghani and Atherton, 2006; Archibald and Murphy, 2021).

The Northern Highlands terrane contains alkaline plutons, including shoshonitic, high-Ba-Sr syenite with high abundances of large ion lithophile elements (LILEs), depleted high field strength elements (HFSEs), and pronounced negative Nb-Ta anomalies (Thompson and Fowler, 1986; Thirlwall and Burnard, 1990; Fowler, 1992; Hughes *et al.*, 2013; Walters *et al.*, 2013; Bruand *et al.*, 2014, 2016). A diverse accessory mineral suite including apatite, allanite, zirconolite, zircon, and titanite is reflected in the significant trace-element variations recorded in these rocks (Fowler, 1992).

In the Northern Highlands terrane, coeval granitoid emplacement and regional-scale thrusting is supported by U-Pb data from concordant synkinematic sills (e.g., Strath Halladale, Assynt, and Naver intrusions; Kocks *et al.*, 2014; Strachan *et al.*, 2020a). These events overlapped and were succeeded by the development of a regional system of steep sinistral faults and shear zones that are interpreted to have acted as the conduits for felsic magmas across much of the Northern Highland and Grampian terranes (Jacques and Reavy, 1994). In the Northern Highlands terrane, plutons emplaced during this phase include Clunes, Ratagain, and Rogart (Rogers and Dunning, 1991; Stewart *et al.*, 2001; Kocks *et al.*, 2014). Offshoots of the Clunes pluton were emplaced syntectonically into sinistral mylonites developed along the Great Glen fault (Stewart *et al.*, 2001), and the sigmoidal geometry of magmatic fabrics within the Ratagain pluton is interpreted to reflect synkinematic emplacement along a ductile precursor to the

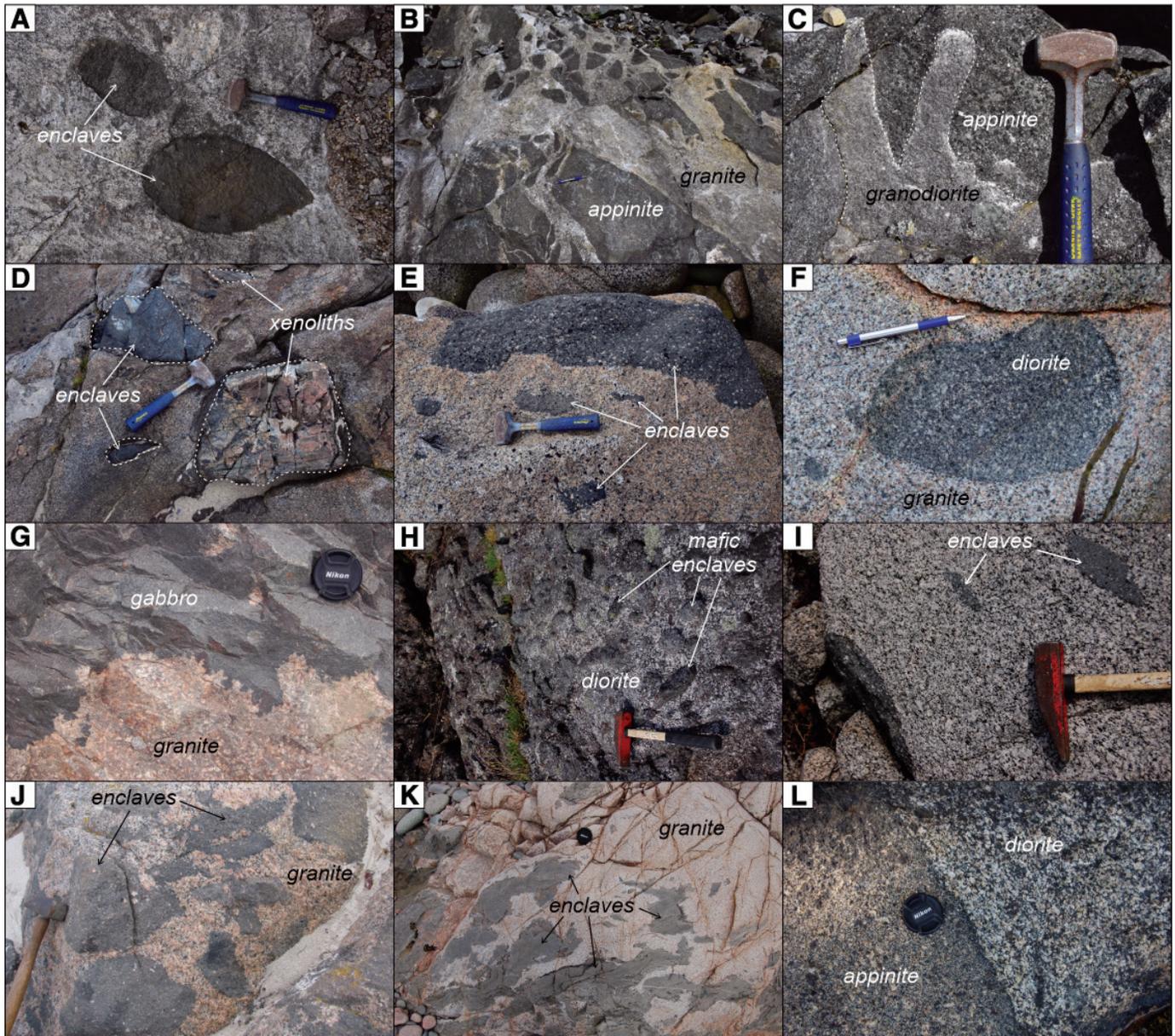


Figure 3. Representative photos showing field relationships between mafic and felsic to intermediate rocks in Ireland and Scotland: (A) oblate mafic enclaves in diorite-granodiorite in the Ardara pluton, Donegal composite batholith, Ireland; (B) breccia pipe with granite hosting appinite xenoliths in the Mulnamin More appinite complex, Donegal composite batholith, Ireland; (C) mingling of diorite with coarse-grained appinite, Mulnamin More appinite complex, Donegal composite batholith, Ireland; (D) Dalradian Supergroup metasedimentary xenoliths and appinite enclaves entrained in the Fanad pluton, Donegal composite batholith, Ireland; (E) plagioclase porphyritic mafic enclave in K-feldspar porphyritic granodiorite in the Thorr pluton, Donegal composite batholith, Ireland; (F) diorite enclave in granite in the Tullagh Point pluton, Donegal composite batholith, Ireland; (G) crenulated felsic-mafic contact from the eastern Granophyre north of Mavis Grind, Shetland Islands, Scotland; (H) disrupted synplutonic microdiorite from the north shore of Loch Sunart, west of Strontian, Strontian Complex, Scotland; (I) mafic enclaves in granite from the north shore of Loch Sunart, west of Strontian, Strontian Complex, Scotland; (J) mafic-felsic mingling relationship from Knockvollagan, Ross of Mull, Scotland; (K) mafic-felsic mingling from the Sandsting Complex, Shetland, Shetland Islands, Scotland; and (L) appinite (left) contact with granite (right) from the north shore of Loch Sunart, west of Strontian, Strontian Complex, Scotland. A–F are from the Grampian terrane in Ireland, and G–L are from the Northern Highlands terrane.

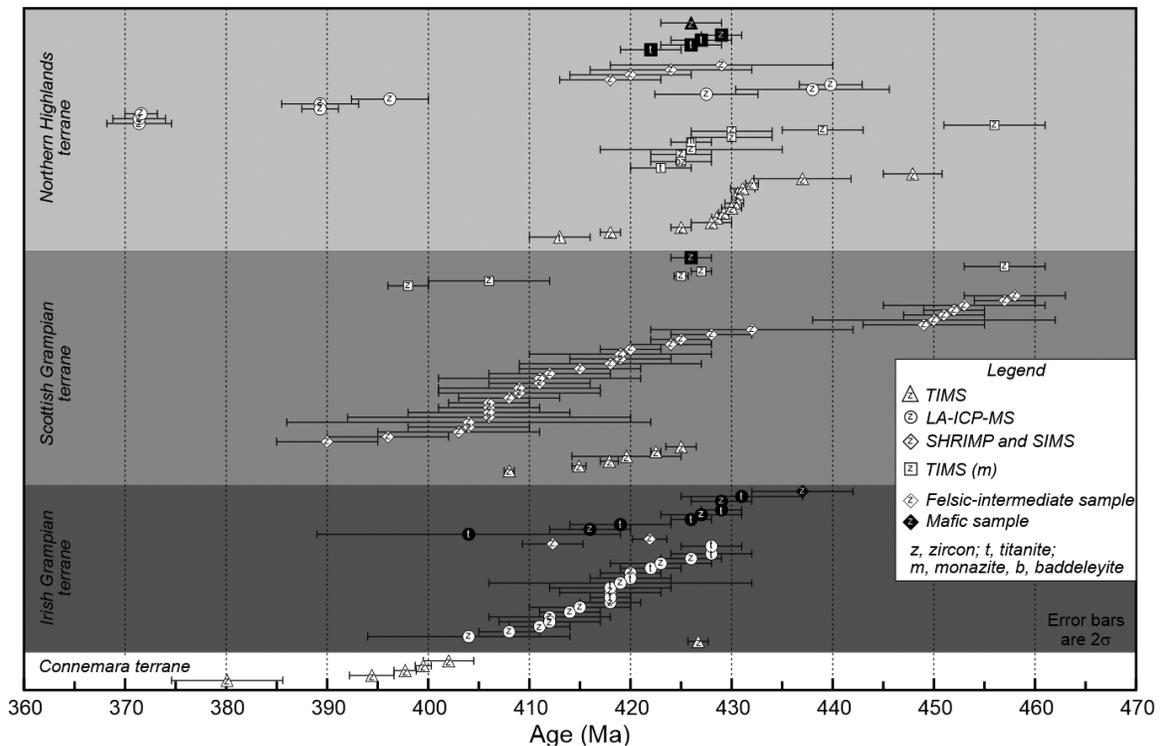


Figure 4. Summary of geochronological data for the late Caledonian plutonic rocks from the Grampian (Ireland and Scotland), Connemara, and Northern Highlands terranes (Fig. 2). U-Pb data are plotted as calculated individual sample ages. Subdivisions are based on terrane, and symbols represent the data collection method. Error bars are  $2\sigma$ . Abbreviations: TIMS—thermal ionization mass spectrometry; SHRIMP—sensitive high-resolution ion microprobe; SIMS—secondary ion mass spectrometry; LA-ICP-MS—laser ablation–inductively coupled plasma–mass spectrometry; TIMS (m)—TIMS multigrain fractions; z—zircon; t—titanite; m—monazite; b—baddeleyite. U-Pb data are from van Breemen et al. (1979); Halliday et al. (1987); Rogers and Dunning (1991); Paterson et al. (1993); Oliver et al. (2000); Stewart et al. (2001); Feely et al. (2003); Kinny et al. (2003); Millward and Evans (2003); Fraser et al. (2004); Goodenough et al. (2006); Kocks et al. (2006); Kirkland et al. (2008); Oliver et al. (2008); Strachan and Evans (2008); Neilson et al. (2009); Appleby et al. (2010); Goodenough et al. (2011); Cooper et al. (2013); Kirkland et al. (2013); Kocks et al. (2014); Lancaster et al. (2017); Lundmark et al. (2019); and Archibald et al. (2021).

Strathconan fault (Hutton and McErlean, 1991). In the Grampian terrane of NW Ireland, the broadly coeval, composite Main Donegal pluton (Hutton, 1982; Hutton and Alsop, 1996; Stevenson, 2009) contains evidence of internal deformation that is congruent with the sinistral deformation in the host rocks, which become increasingly deformed toward the margins of the pluton (Hutton and Alsop, 1995; Long and McConnell, 1997).

## DATA COMPILATION

To investigate the potential genetic relationships of late Caledonian plutonic rocks, we compiled U-Pb geochronological, lithochemical, and Nd-Sr isotopic data from late Caledonian plutonic rocks located north of the Highland Boundary fault with the addition of the Connemara terrane (Fig. 2). The data sources are listed in the figure captions. We subdivided the data into four groupings: (1) Grampian terrane in Ireland; (2) Connemara terrane in Ireland; (3) Northern Highlands terrane in Scotland; and (4) Grampian terrane in Scotland (Fig. 2). The terrane divisions

in Scotland are consistent with the isotopic composition of late Caledonian plutonic rocks across the Great Glen fault (Fig. 2; e.g., Canning et al., 1998). The Connemara and Grampian terranes reside on opposite sides of the extrapolation of the Highland Boundary fault across Ireland (Fig. 2; Chew and Strachan, 2014; Dewey et al., 2015; Miles et al., 2016), but it is uncertain whether this boundary coincides with significant compositional variations in late Caledonian plutonic rocks.

The geochronological data compilation preferentially includes published U-Pb data obtained using zircon, monazite, baddeleyite, and titanite (Figs. 4 and 5) (see Supplemental Material<sup>1</sup>). These data were obtained from a variety of analytical methods, such as single-grain thermal ionization mass spec-

<sup>1</sup>Supplemental Material. A: U-Pb geochronological data; B: Lithochemical data; C: Nd and Sr isotopic data; and D: Data sources used in the compilation of U-Pb, lithochemical, and Nd-Sr data. Please visit <https://doi.org/10.1130/SPE.SXXXX> to access the supplemental material, and contact [editing@geosociety.org](mailto:editing@geosociety.org) with any questions.

trometry (TIMS), TIMS multigrain fractions, secondary ion mass spectrometry (SIMS), sensitive high-resolution ion microprobe (SHRIMP), and laser ablation–inductively coupled plasma–mass spectrometry (LA-ICP-MS). Rocks excluded from the compilation included those dated by the K-Ar and Ar-Ar methods because they are commonly interpreted to represent cooling ages and not necessarily the magmatic crystallization age. However, given that the Ar-Ar closure temperature for hornblende is  $\sim 550$  °C (Harrison, 1982), and the host metasedimentary rocks

to plutons in the Grampian terrane are greenschist facies, the cooling ages obtained from some plutons may closely date igneous emplacement (e.g., Murphy et al., 2019). In addition, we excluded whole-rock Rb-Sr ages because of postemplacement processes that commonly result in erroneous ages. These processes include the potential relative mobility of Rb and Sr during secondary alteration, the probable isotopic heterogeneity of the source rocks, and field evidence for extensive mixing and mingling between mafic and felsic magmas that implies a potential

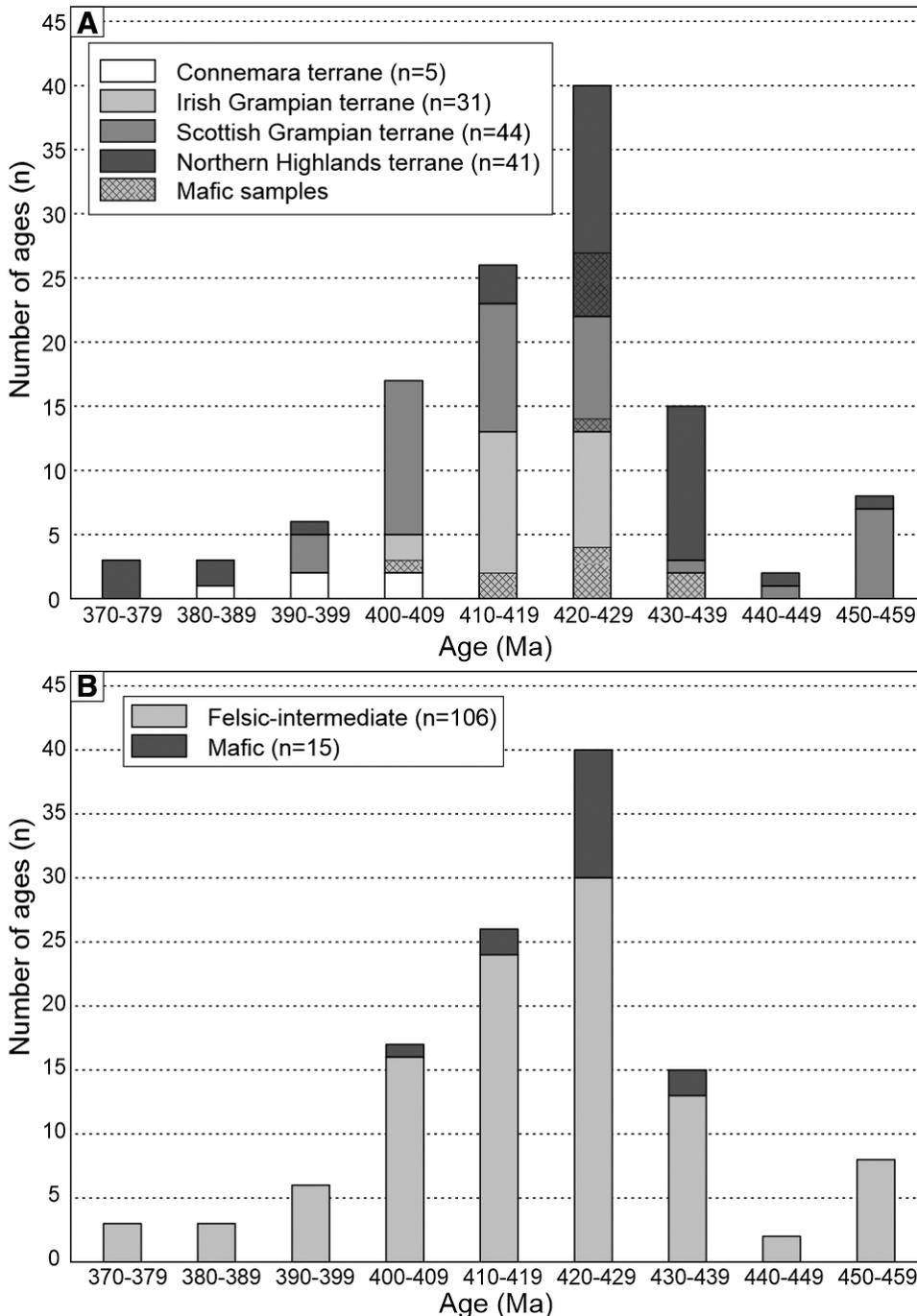


Figure 5. Histograms showing the distribution of U-Pb ages using a 10 m.y. bin size. (A) U-Pb ages subdivided based on geological terrane. (B) U-Pb ages subdivided based on rock type. Data sources are listed in the Figure 4 caption.

lack of Sr isotopic homogenization during emplacement (e.g., Chew and Schaltegger, 2005). We included only samples with crystallization ages younger than ca. 460 Ma, the interpreted end of the Grampian orogeny in Scotland and Ireland (Flowerdew et al., 2000; Chew and Strachan, 2014; Dewey and Ryan, 2015; Johnson et al., 2017).

The compilation of lithochemical data includes the petrogenetically significant trace elements and rare earth elements (REEs). Because many data sets do not include volatile compositions (resulting in low totals), the major-element data were normalized as volatile free (Fig. 6). Normalized (Sun and McDonough, 1989) REE and trace-element variation diagrams plot the average of all samples compiled from each terrane, and the numbers of samples used to calculate the average concentrations are shown with each plot (Fig. 7).

The compilation of isotopic data (Fig. 8) includes whole-rock Nd and Sr isotopic data. The initial Nd and Sr ratios for published data without reliable U-Pb age constraints were recalculated using the most recently published precise U-Pb ages. Initial Sr ratios for the Donegal composite batholith (O'Connor et al., 1987; Dempsey et al., 1990) and Tullagh Point pluton (O'Connor et al., 1987) were recalculated using the U-Pb ages in Archibald et al. (2021) and Kirkland et al. (2008), respectively.

## RESULTS

### U-Pb Data

The youngest U-Pb ages in our compilation (ca. 371 Ma,  $n = 3$ ) are from felsic to intermediate plutonic rocks in the Northern

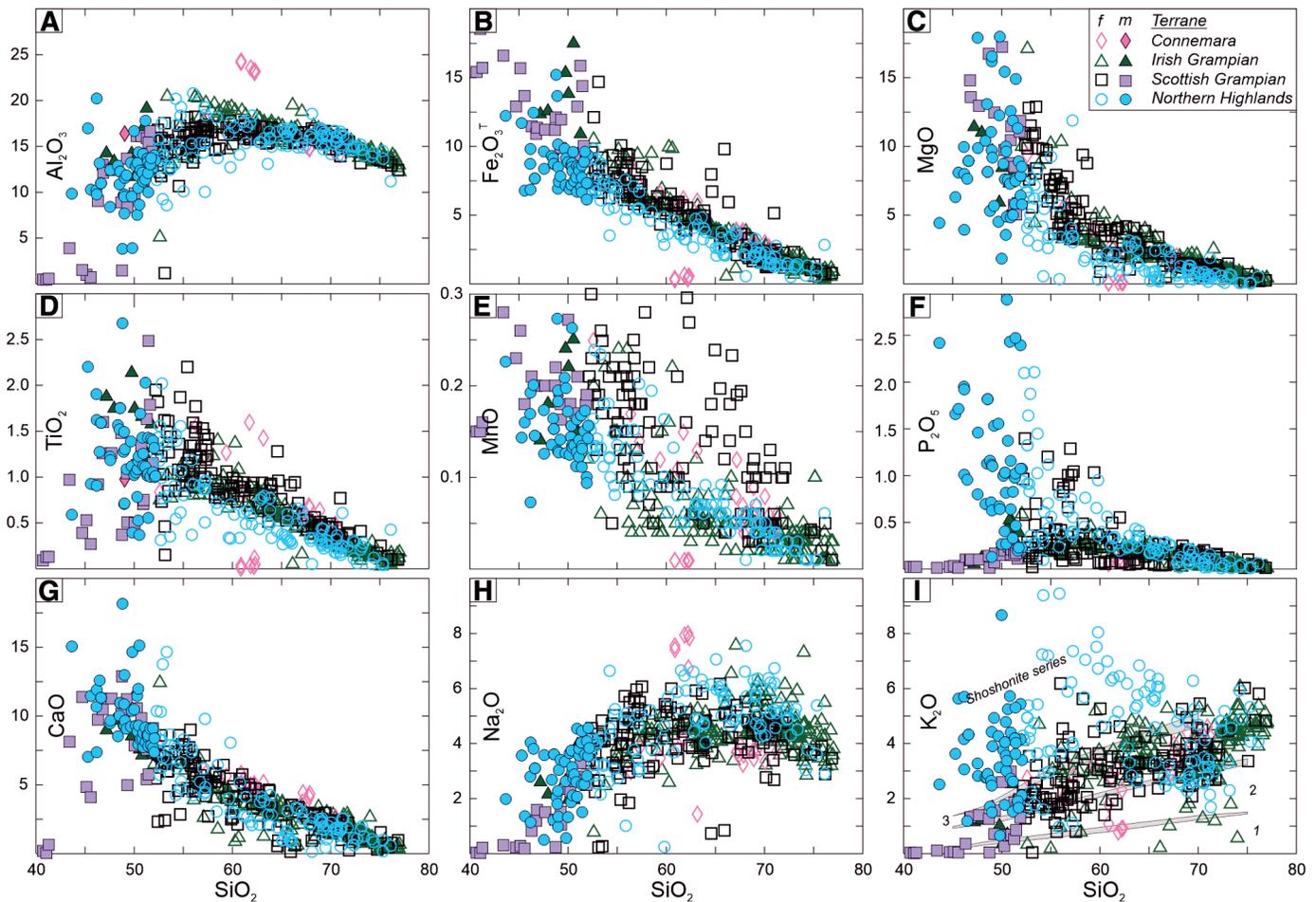


Figure 6. Variation diagrams for major-element oxides plotted against  $\text{SiO}_2$  (in wt%) for late Caledonian granitoids and associated mafic to ultra-mafic rocks. Fields in I are (1) low-K tholeiite series, (2) calc-alkaline series, and (3) high-K calc-alkaline series (Rickwood, 1989). Data sources are Thompson and Fowler (1986); Fowler (1988); Thirlwall and Burnard (1990); Fowler (1992); Canning et al. (1996); El Desouky et al. (1996); Fowler and Henney (1996); Ghani (1997); Graham et al. (2000); Fowler et al. (2001); Fowler et al. (2008); Ghani and Atherton (2006); Neilson (2008); Steinhöfel et al. (2008); Clemens et al. (2009a); Neill and Stephens (2009); Cooper et al. (2013); Walters et al. (2013); Lundmark et al. (2019); Murphy et al. (2019); Archibald and Murphy (2021); and B.P. Kokelaar (2020, personal commun.). Abbreviations: m—mafic rocks; f—intermediate to felsic rocks.

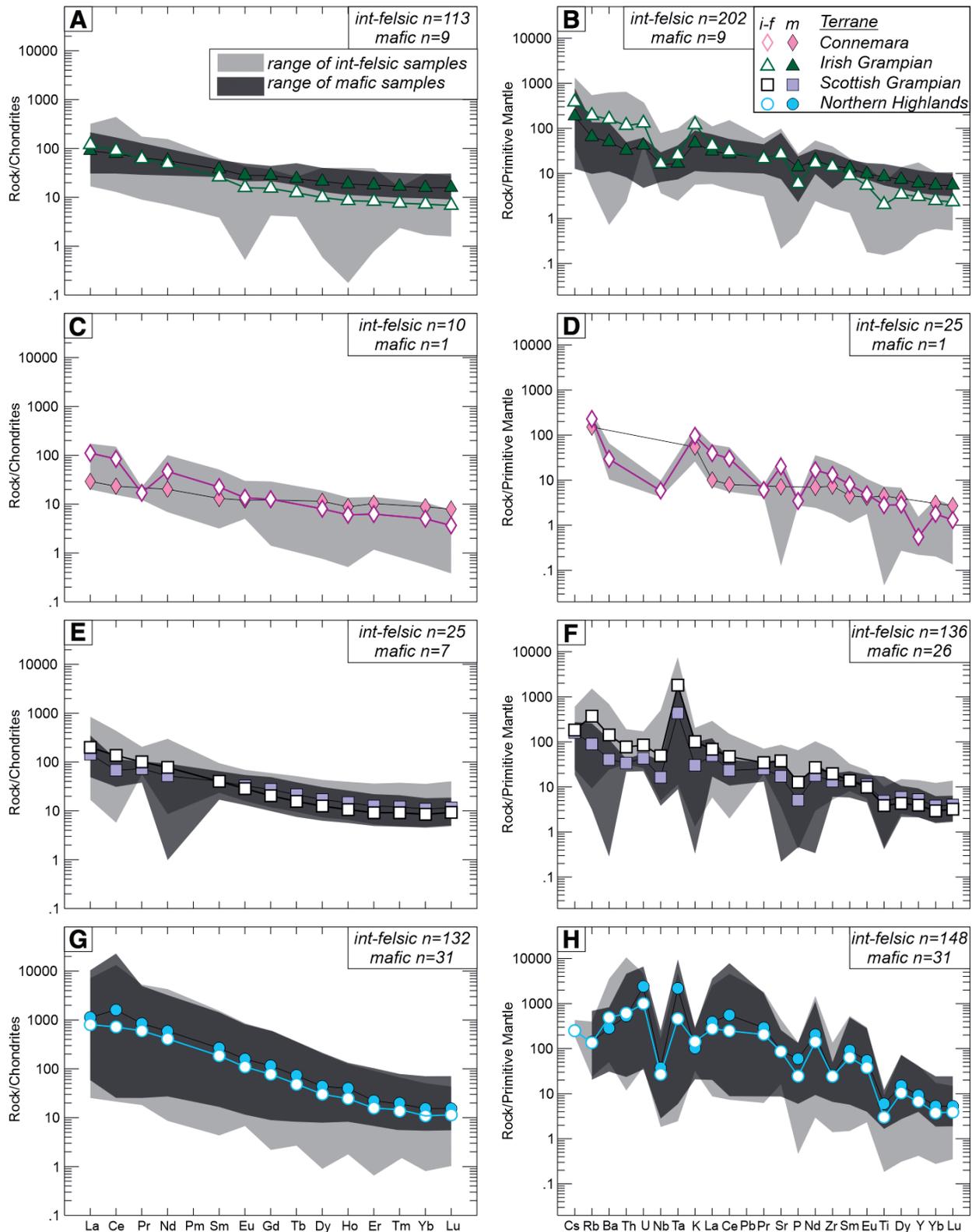


Figure 7. (A, C, E, G) Chondrite-normalized rare earth element (REE) diagrams and (B, D, F, H) primitive mantle-normalized trace-element variation diagrams for late Caledonian plutonic rocks: (A–B) Irish Grampian terrane, (C–D) Connemara terrane, (E–F) Northern Highlands terrane, (G–H) Scottish Grampian terrane. Plots show average values for ( $n$ ) samples from the data sources listed in the Figure 6 caption. Normalized (Sun and McDonough, 1989) REE and trace-element variation diagrams plot the average of all samples compiled from each terrane, and the numbers of samples used to calculate the average concentrations are shown with each plot (Fig. 7). Abbreviations: m—mafic rocks; int-felsic/i-f—intermediate to felsic rocks.

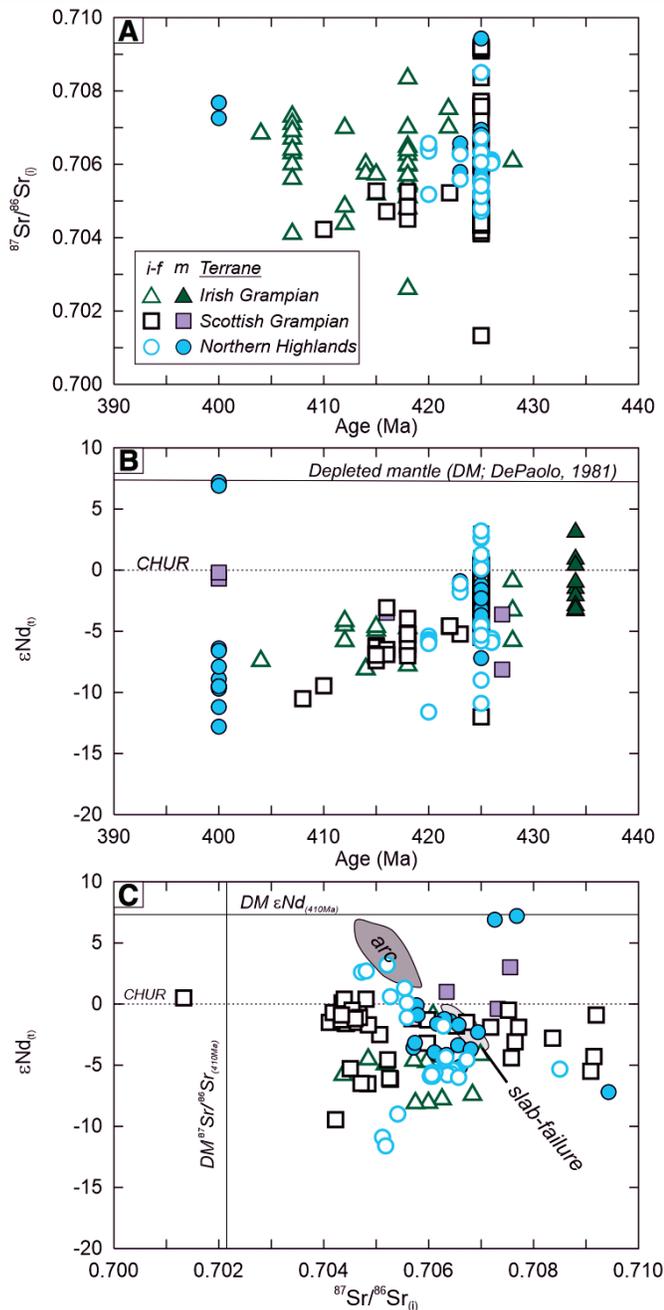


Figure 8. Isotopic data for late Caledonian granitoids and related rocks from the Grampian and Northern Highlands terranes: (A) age vs.  $^{87}\text{Sr}/^{86}\text{Sr}(t)$ , (B) age vs.  $\epsilon_{\text{Nd}}(t)$ , and (C)  $^{87}\text{Sr}/^{86}\text{Sr}(t)$  vs.  $\epsilon_{\text{Nd}}(t)$ . The depleted mantle (DM)  $\epsilon_{\text{Nd}}(t)$  curve in B and C was calculated using the values in DePaolo (1981). CHUR—chondritic uniform reservoir. The  $^{87}\text{Sr}/^{86}\text{Sr}_{(410\text{Ma})}$  ratio in C was calculated using the values in Salters and Stracke (2004). The fields for Peninsular Ranges and Sierran arc and slab-failure magmas are from Hildebrand et al. (2018). Data sources are O'Connor et al. (1982); O'Connor et al. (1987); Dempsey et al. (1990); Fowler (1992); Canning et al. (1996); Canning et al. (1998); Fowler et al. (2001); Fowler et al. (2008); Neilson (2008); Clemens et al. (2009a); Murphy et al. (2019). Abbreviations: m—mafic rocks; i-f—intermediate to felsic rocks.

Highlands terrane. The oldest ages are between ca. 460 and 450 Ma from the Northern Highlands and Grampian terranes in Scotland (Figs. 4 and 5). Most magmatic crystallization ages for felsic to intermediate rocks are between ca. 430 and 400 Ma (Fig. 4).

There are fewer U-Pb ages in our compilation for mafic to ultramafic rocks (Figs. 4 and 5). U-Pb data from zircon and titanite yielded ages ranging between ca. 437 and 415 Ma, which overlap with most of the age range of the felsic to intermediate rocks (Fig. 4). The  $437 \pm 5$  Ma age is a SIMS age from a lamprophyre dike (Kirkland et al., 2013). One outlier titanite date from a gabbroic enclave in the Irish Grampian terrane yielded an imprecise and unreliable date at ca. 404 Ma. This date likely represents postcrystallization Pb loss in titanite, because zircon from the same sample yielded a U-Pb age of  $416 \pm 4$  Ma (Archibald et al., 2021).

### Geochemistry—Major Elements

Samples showed a wide range of  $\text{SiO}_2$  concentrations (Fig. 6), varying from 46.4 wt% in the Glenelg-Ratagain pluton (Fowler et al., 2008) to 81.3 wt% in a sample from the Rosses pluton in the Donegal composite batholith (Archibald and Murphy, 2021). On all lithochemical plots, samples are divided based on their  $\text{SiO}_2$  concentrations. Mafic samples have  $\text{SiO}_2$  less than 52 wt%, and felsic to intermediate samples have  $\text{SiO}_2$  greater than 52 wt%. All late Caledonian plutonic rocks, regardless of terrane, share similar major-element compositions with only a few notable differences, as outlined below (Fig. 6). The spread of data shown by the mafic to ultramafic samples probably indicates cumulate compositions instead of liquid compositions (Fig. 6; Hildebrand et al., 2018).

The  $\text{Al}_2\text{O}_3$  concentrations in felsic to intermediate samples vary between 5.1 and 20.8 wt% (Fig. 6A), except for a suite of plagioclase-rich microgranular enclaves that have concentrations up to 24.3 wt%  $\text{Al}_2\text{O}_3$  (Graham et al., 2000). In mafic samples, the  $\text{Al}_2\text{O}_3$  concentrations are between 4.1 and 21.0 wt% (Fig. 6A). The  $\text{Fe}_2\text{O}_{3\tau}$  ( $1.1\text{FeO} + \text{Fe}_2\text{O}_3$ ) compositions in felsic to intermediate samples are between 0.3 and 12.2 wt%. Mafic samples have higher  $\text{Fe}_2\text{O}_{3\tau}$  concentrations (between 7.4 and 17.5 wt%; Fig. 6B).  $\text{MgO}$  in felsic to intermediate samples has a compositional range between 0.1 wt% and 10.2 wt%. Some appinite samples from the Northern Highlands terrane have  $\text{MgO}$  compositions up to 18.0 wt% (Fig. 6C).  $\text{TiO}_2$  concentrations are between 0.1 wt% and 2.8 wt% in felsic to intermediate samples and between 0.1 wt% and 3.6 wt% in mafic samples (Figs. 6D and 6E).  $\text{P}_2\text{O}_5$  (Fig. 6F) concentrations are low in both mafic and felsic to intermediate samples (both  $<0.5$  wt%), except for samples from the Northern Highland terrane (up to 3.1 wt%; Fig. 6F). Taken together,  $\text{Fe}_2\text{O}_{3\tau}$ ,  $\text{MgO}$ ,  $\text{TiO}_2$ ,  $\text{MnO}$ , and  $\text{P}_2\text{O}_5$  each display a negative correlation with increasing  $\text{SiO}_2$ .  $\text{CaO}$  concentrations in felsic to intermediate samples vary between 0.1 wt% and 12.4 wt%, and values in mafic samples vary between 7.0 wt% and 14.6 wt% (Fig. 6G). Felsic to intermediate samples have  $\text{Na}_2\text{O}$  of 0.8–11.1 wt% and  $\text{K}_2\text{O}$  of 0.2–9.4 wt%, and both elements

show a broad positive correlation with increasing SiO<sub>2</sub> (Figs. 6H and 6I). However, samples from the Northern Highlands terrane have higher K<sub>2</sub>O and more shoshonitic tendencies than samples from the other terranes (Fig. 6I). Mafic samples have lower Na<sub>2</sub>O (0.5–4.1 wt%) and K<sub>2</sub>O (0.3–8.7 wt%) than the felsic to intermediate samples (Figs. 6H and 6I).

### Geochemistry—Rare Earth Elements and Other Trace Elements

Felsic to intermediate samples from the Irish Grampian terrane display moderate light rare earth element (LREE) enrichment with average La<sub>N</sub>/Sm<sub>N</sub> of ~4.6 and heavy rare earth element (HREE) depletion (Gd<sub>N</sub>/Yb<sub>N</sub> ~2.2) with average La<sub>N</sub>/Yb<sub>N</sub> of ~21.0 (Fig. 7A). Samples generally lack a pronounced Eu anomaly, with Eu/Eu\* values of ~0.8. Primitive mantle-normalized trace-element plots exhibit enrichment in LILEs (Cs, Rb, and Ba) and depletion in both HFSEs (Nb, Ta, P, Ti) and HREEs (Fig. 7B). The average REE profiles of mafic and ultramafic rocks are similar, with La<sub>N</sub>/Sm<sub>N</sub> of ~2.3, Gd<sub>N</sub>/Yb<sub>N</sub> of ~2.2, and shallower REE slopes than the felsic to intermediate samples with La<sub>N</sub>/Yb<sub>N</sub> of ~6.9 (Fig. 7A). Eu/Eu\* values are ~0.9 (Fig. 7A). Trace-element plots for mafic rocks display similar enrichment in LILEs and depletion in HFSEs as the felsic to intermediate samples except for Ti, which is undepleted in the mafic to ultramafic samples (Fig. 7B).

Felsic to intermediate samples from the Connemara terrane display moderate LREE enrichment relative to HREEs, with averages of La<sub>N</sub>/Sm<sub>N</sub> ~6.3, Gd<sub>N</sub>/Yb<sub>N</sub> ~2.4, and La<sub>N</sub>/Yb<sub>N</sub> ~28.2 (Fig. 7C). These samples have Eu/Eu\* values that are ~1.1. Trace-element plots show enrichment in Cs and depletion in Nb, P, Ti, and HREEs (Fig. 7D). Data are available from only one mafic sample (Fig. 7C), which has La<sub>N</sub>/Sm<sub>N</sub> of ~2.2, Gd<sub>N</sub>/Yb<sub>N</sub> of 1.4, and La<sub>N</sub>/Yb<sub>N</sub> of ~3.3 (Fig. 7C) and lacks a Eu anomaly (Eu/Eu\* ~1.0). Trace-element plots indicate similar LILE enrichment and HFSE depletion compared to the felsic to intermediate samples (Fig. 7B).

In the Northern Highlands terrane, the felsic to intermediate samples exhibit steeper REE slopes than those from the Irish Grampian and Connemara terranes (Fig. 7). The Northern Highland samples are more enriched in LREE (average La<sub>N</sub>/Sm<sub>N</sub> ~6.3), are more depleted in HREE (average Gd<sub>N</sub>/Yb<sub>N</sub> ~4.8), and have steeper REE slopes with average La<sub>N</sub>/Yb<sub>N</sub> of ~48.8 (Fig. 7E). Samples lack a significant Eu anomaly, with average Eu/Eu\* of ~0.9. Trace-element plots show enrichment in LILEs and depletion in the HFSEs (Fig. 7F). Mafic to ultramafic rocks have a similar REE profile to the felsic to intermediate rocks but are more enriched in LREEs than the felsic to intermediate rocks (Fig. 7E). Average La<sub>N</sub>/Sm<sub>N</sub> is ~4.0, Gd<sub>N</sub>/Yb<sub>N</sub> is ~4.8, and average La<sub>N</sub>/Yb<sub>N</sub> is ~41.6 (Fig. 7E). Eu/Eu\* values are ~0.9 (Fig. 7E). Trace-element plots show the same enrichments in LILEs and depletions in HFSEs as the felsic to intermediate samples (Fig. 7F).

Felsic to intermediate samples from the Scottish Grampian terrane exhibit similar REE patterns to those from the Irish

Grampian terrane (Fig. 7). Average La<sub>N</sub>/Sm<sub>N</sub> is ~3.9, Gd<sub>N</sub>/Yb<sub>N</sub> is ~2.4, and average La<sub>N</sub>/Yb<sub>N</sub> is 18.9 (Fig. 7G). Samples also have similar Eu/Eu\* values of ~0.8. Trace-element plots show enrichment in the LILEs relative to HFSEs and HREEs (Fig. 7H). Compared to mafic to ultramafic rocks, the felsic to intermediate samples have similar REE profiles except for LREEs, which are more enriched. Average La<sub>N</sub>/Sm<sub>N</sub> is ~3.5, Gd<sub>N</sub>/Yb<sub>N</sub> is 2.2, and La<sub>N</sub>/Yb<sub>N</sub> is 12.6 (Fig. 7G). Eu/Eu\* values are ~1.0 (Fig. 7G). Trace-element plots show the similar enrichments in LILEs and depletions in HFSEs, but to a lesser degree than the felsic to intermediate samples (Fig. 7H).

### Isotopic Data

Felsic to intermediate samples in the Irish Grampian terrane have a wide range in initial <sup>87</sup>Sr/<sup>86</sup>Sr<sub>(i)</sub> (Fig. 8), mostly between 0.70260 and 0.70834, for samples with U-Pb ages between ca. 428 and 404 Ma (Fig. 8A). Initial <sup>87</sup>Sr/<sup>86</sup>Sr<sub>(i)</sub> for felsic to intermediate samples from the Scottish Grampian terrane are between 0.70133 and 0.71024 (Fig. 8A), and mafic samples have initial <sup>87</sup>Sr/<sup>86</sup>Sr<sub>(i)</sub> between 0.70634 and 0.71145 (Fig. 8A). Initial <sup>87</sup>Sr/<sup>86</sup>Sr<sub>(i)</sub> values for felsic to intermediate samples from the Northern Highlands terrane have a similar range between 0.70472 and 0.70850 (Fig. 8A), and mafic samples have initial <sup>87</sup>Sr/<sup>86</sup>Sr<sub>(i)</sub> between 0.70571 and 0.70943 (Fig. 8A).

The ε<sub>Nd</sub>(*t*) values (*t* = 434 Ma) from mafic samples of the appinite suite in the Irish Grampian terrane range from –5.8 to +3.1 (Fig. 8B). Coeval felsic to intermediate rocks have more evolved ε<sub>Nd</sub>(*t*) values between –8.1 and –0.9. Mafic samples from the Northern Highlands terrane have ε<sub>Nd</sub>(*t*) values mostly between –12.8 and –0.1. However, two samples have ε<sub>Nd</sub>(*t*) values of +6.9 and +7.2, which are similar to contemporary depleted mantle (Figs. 8B and 8C). Felsic to intermediate samples in the Northern Highlands terrane have a wider range of ε<sub>Nd</sub>(*t*) values (between –11.6 and +3.2). Felsic to intermediate samples from the Scottish Grampian terrane have ε<sub>Nd</sub>(*t*) values of +0.5 and –10.5, and mafic samples have ε<sub>Nd</sub>(*t*) values between +3.0 and –8.1 (Fig. 8B). When plotted together, initial <sup>87</sup>Sr/<sup>86</sup>Sr<sub>(i)</sub> and ε<sub>Nd</sub>(*t*) values from both mafic and felsic to intermediate samples show a similar range for the Northern Highlands and the Grampian terranes (Fig. 8C).

## DISCUSSION

### Late Caledonian Magmatism in Northern Scotland and Northwestern Ireland

The oldest magmatic crystallization ages in our compilation (460–450 Ma) are from felsic to intermediate rocks in Scotland (Fig. 4) and are probably related to processes occurring at the end of the ca. 465–455 Ma Grampian orogeny. The paucity of ages between ca. 450 and 440 Ma suggests an interval of quiescence in magma emplacement prior to the onset of the Scandian orogeny (Fig. 4). The Glen Dessary syenite in the Northern Highlands

terrane (Fig. 2) has a U-Pb zircon age of  $448 \pm 3$  Ma (Goodenough et al., 2011) and is the only pluton dated within this interval. Single zircon and titanite dates within this interval are reported from the Donegal composite batholith, but the geological significance of these dates is uncertain (Archibald et al., 2021). The lack of preserved plutonic rocks within this interval has been attributed either to low-angle subduction that suppressed magmatism or to erosional removal of a volcanic arc following collision (Oliver et al., 2008; Miles et al., 2016). Alternatively, according to Glazner (1991), subduction at a high angle to a continental margin could have suppressed emplacement of arc magmas if deformation failed to produce the favorable pathways required to facilitate magma ascent. Granite clasts in Silurian synorogenic conglomerate from the probable northern extension of the Grampian terrane in the Shetland Islands have U-Pb ages of ca. 440 Ma (Biejat et al., 2018). The clasts are interpreted to have been derived from thrust sheets located to the east composed of Dalradian (Neoproterozoic) metasedimentary rocks, igneous rocks from the Grampian orogeny, and late Caledonian plutons possibly emplaced in a Late Ordovician to early Silurian arc (Biejat et al., 2018).

Magma emplacement during the Scandian orogenic event began in the early Silurian. The oldest documented ages are a ca. 437 Ma U-Pb zircon date from a lamprophyre (Kirkland et al., 2013) and a ca. 434 Ma Ar-Ar date from hornblende in the appinite suite (Murphy et al., 2019) of the Irish Grampian terrane. These ages suggest that some mafic magmatism predated emplacement of the felsic to intermediate plutons. The youngest documented magmatic crystallization age for mafic to ultramafic samples is ca. 415 Ma (Archibald et al., 2021). Most U-Pb crystallization ages for felsic to intermediate samples are between ca. 430 and 405 Ma (Figs. 4 and 5). U-Pb data from the Donegal composite batholith in the Irish Grampian terrane support the coeval relationship between late Caledonian mafic-ultramafic and felsic-intermediate magmas (Archibald et al., 2021). The range of ages (ca. 428–415 Ma) obtained from the mafic enclaves hosted in the Ardara, Thorr, and Fanad plutons indicates ongoing mafic magmatism during sequential emplacement of the granitoid plutons (Archibald et al., 2021). Field observations of widespread mingling and mixing indicate that mafic and felsic magmatism were coeval during the emplacement of the Donegal composite batholith, and lithochemical data support a cogenetic relationship (Archibald and Murphy, 2021). Similar mafic-intermediate-felsic magma mixing and mingling relationships are observed elsewhere in the Northern Highlands, Grampian, and Connemara terranes (e.g., Ross of Mull—Zaniewski et al., 2006; McLeod et al., 2010; Garabal Hill—Clemens et al., 2009a; Loch Loyal Syenite Complex—Hughes et al., 2013; Cluanie et al., 2009; Strontian—Bruand et al., 2014; Galway—El Desouky et al., 1996; Graham et al., 2000). Taken together, these observations and data support a prolonged magma emplacement interval (between ca. 435 and 400 Ma). With the exception of the oldest rocks in the Northern Highlands terrane and the youngest rocks in Shetland, there are no significant differences between the terranes in the age of magmatism, and most of the ages over-

lap (Fig. 4). Lancaster et al. (2017) interpreted relatively young ages (ca. 390 and 370 Ma) in Shetland (Northern Highlands terrane) as discrete magmatic pulses emplaced along fault zones during late to post-Scandian sinistral displacement between Laurentia and Baltica.

The late Caledonian plutonic rocks have broadly similar major-element compositions. Intermediate to felsic compositions predominate over mafic compositions in all terranes, and there is significantly more variation in major-element contents at lower  $\text{SiO}_2$  values (Fig. 6), which is likely a result of crystal accumulation processes. However, there are some notable distinctions. For example, mafic and felsic to intermediate samples from the Northern Highlands terrane generally have higher concentrations of  $\text{P}_2\text{O}_5$  than samples collected in the other terranes (Fig. 6F). In addition, the mafic and the intermediate to felsic rocks in the Northern Highlands terrane have more shoshonitic tendencies (e.g., higher  $\text{K}_2\text{O}$ , higher  $\text{La}/\text{Yb}$ ) compared to the samples from the Grampian and Connemara terranes (Fig. 6I; Fowler, 1988). Overall, both mafic and felsic-intermediate samples from the Northern Highlands terrane have higher  $\text{La}_\text{N}/\text{Yb}_\text{N}$ ,  $\text{La}_\text{N}/\text{Sm}_\text{N}$ , Ba, and Sr concentrations than samples with equivalent  $\text{SiO}_2$  concentrations in the other terranes (Figs. 9C–9F).

Many late Caledonian plutons have high Sr/Y values, which are typical of adakitic magmas, but some late Caledonian plutons have higher Y concentrations, most notably in the samples from the Northern Highlands terrane (Fig. 9A). Elevated Sr/Y and high La/Yb can be generated by the presence of garnet in a source residue, because Y and Yb strongly partition into garnet (van Westrenen et al., 1999). High Sr/Y can also result from Sr enrichment in the source, possibly caused by sediment subduction and input into the mantle source (Plank and Langmuir, 1993; Fowler et al., 2008). The  $\text{La}_\text{N}/\text{Yb}_\text{N}$  values in both mafic and felsic to intermediate samples of the Northern Highlands terrane are higher than typical adakitic rocks (Fig. 9B). In contrast, mafic and felsic to intermediate samples from the Irish Grampian, Connemara, and Scottish Grampian terranes have a wide range of Sr/Y and  $\text{La}_\text{N}/\text{Yb}_\text{N}$  values that straddles the divide between arc and adakite fields (Figs. 9A and 9B).

All samples with  $\text{SiO}_2$  concentrations <70 wt% lack a significant Eu anomaly (Fig. 9G). However, samples with higher  $\text{SiO}_2$  (>70 wt%) have a pronounced Eu anomaly (Fig. 9G), which, together with their lower Sr and Ba concentrations (Fig. 9H), is consistent with plagioclase fractionation. For example, samples from the Donegal composite batholith in the Irish Grampian terrane (green triangles) show a strong correlation between increasing  $\text{SiO}_2$  and the magnitude of the negative Eu anomaly, attesting to the importance of plagioclase fractionation during later stages of batholith construction.

### Petrogenesis of Late Caledonian Plutons

Recently developed tectonic discrimination diagrams distinguish between arc, slab failure, and A-type granitoids for samples with  $\text{SiO}_2$  concentrations between 55 wt% and 70 wt%

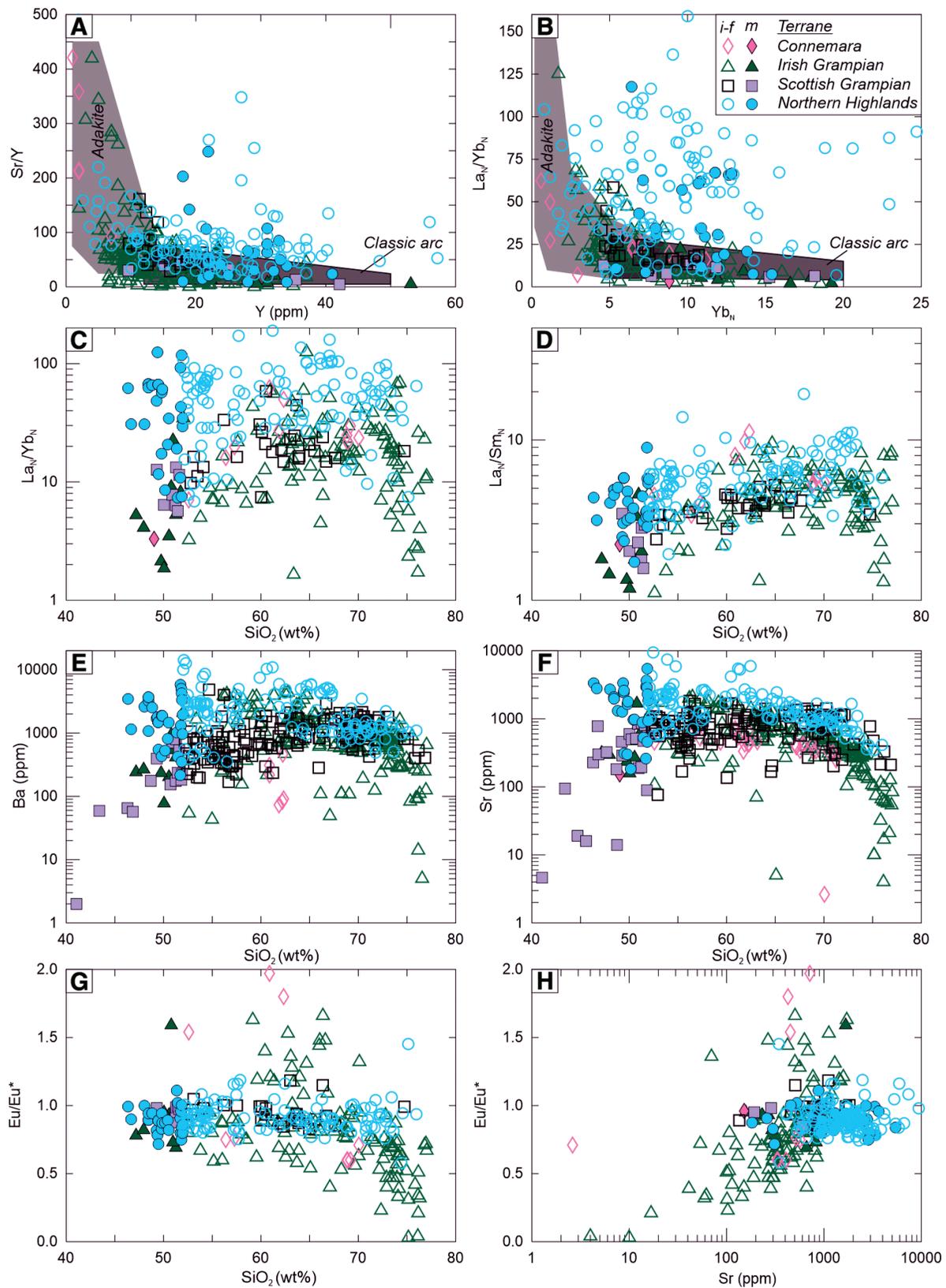


Figure 9. Comparison of late Caledonian rocks with adakites: (A) Sr/Y vs. Y, showing the classic arc and adakite fields; (B)  $\text{La}_N/\text{Yb}_N$  vs.  $\text{Yb}_N$ , showing the classic island-arc and adakite fields, with adakite fields from Martin (1986), Drummond and Defant (1990), and Martin (1999); (C)  $\text{La}_N/\text{Yb}_N$  vs.  $\text{SiO}_2$ ; (D)  $\text{La}_N/\text{Sm}_N$  vs.  $\text{SiO}_2$ ; (E) Ba (ppm) vs.  $\text{SiO}_2$ ; (F) Sr (ppm) vs.  $\text{SiO}_2$ ; (G)  $\text{Eu}/\text{Eu}^*$  vs.  $\text{SiO}_2$ ; and (H)  $\text{Eu}/\text{Eu}^*$  vs. Sr (ppm). Normalizing values in B–D and G–H are from Sun and McDonough (1989). Data sources are listed in the Figure 6 caption. Abbreviations: m—mafic rocks; i-f—felsic to intermediate rocks.

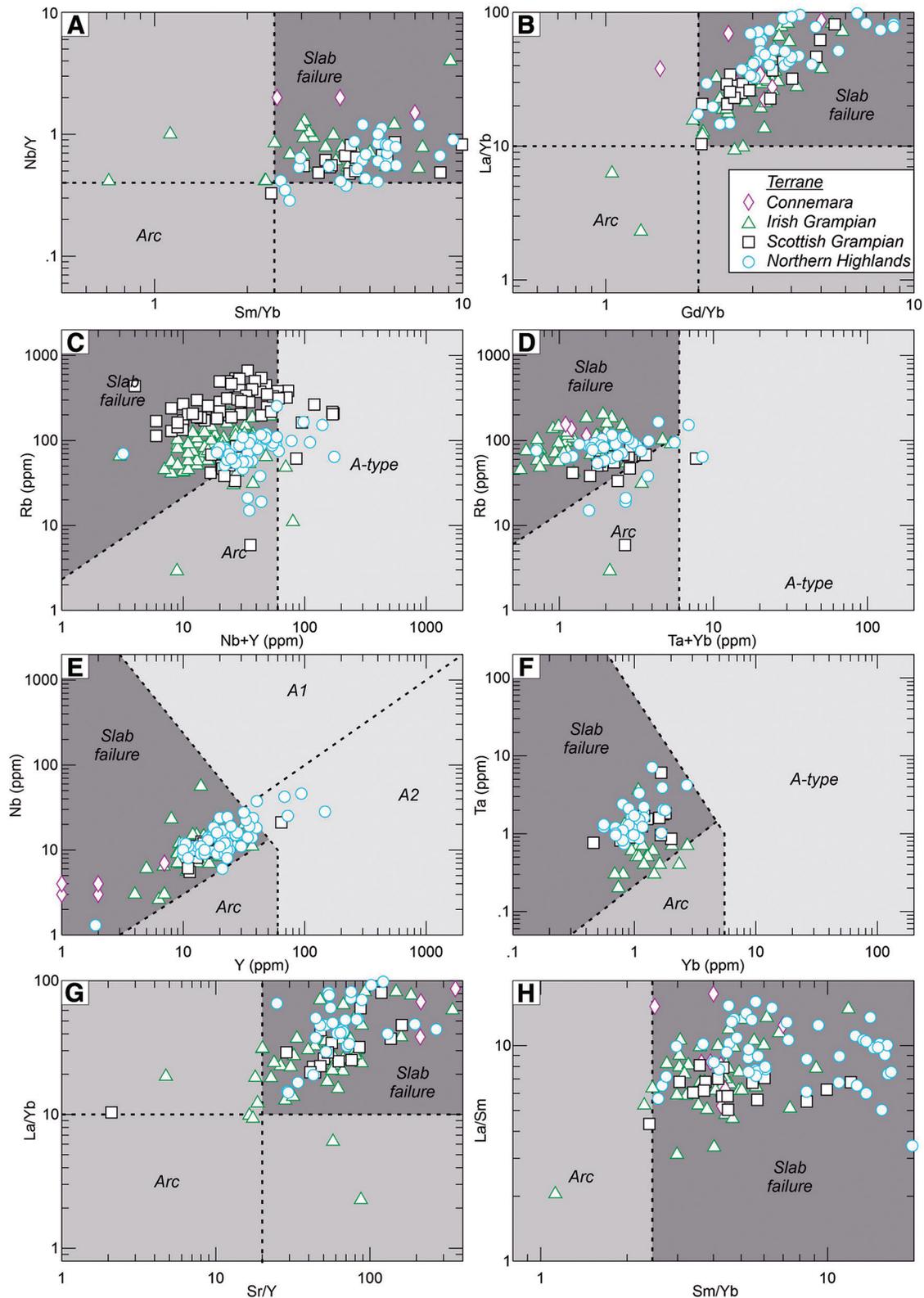


Figure 10. Selected tectonic discrimination diagrams (Hildebrand et al., 2018; Whalen and Hildebrand, 2019) plotting samples of late Caledonian intermediate to felsic rocks with  $\text{SiO}_2$  concentrations between 55 and 70 wt% and alumina saturation index (ASI) values  $<1.1$ : (A) Sm/Yb vs. Nb/Y; (B) Gd/Yb vs. La/Yb; (C) Nb + Y vs. Rb; (D) Ta + Yb vs. Rb; (E) Y vs. Nb; (F) Yb vs. Ta; (G) Sr/Y vs. La/Yb; and (H) Sm/Yb v. La/Sm. ASI was calculated using molar oxides and the formula  $\text{ASI} = \text{Al}/(\text{Ca} - 3.33 \times \text{P}) + \text{Na} + \text{K}$  (Frost and Frost, 2008). In H, nearly all samples have Sm/Yb  $>2.5$ , which is indicative of a slab-failure composition. Data sources are listed in the Figure 6 caption. Several samples that had anomalously high Ta values from the Northern Highlands and Grampian terranes in Scotland are not plotted on D and F.

and an alumina saturation index (ASI) less than 1.1 (Hildebrand and Whalen, 2014b, 2017; Hildebrand et al., 2018; Whalen and Hildebrand, 2019). Rocks generated by slab-failure processes are relatively enriched in Sr, Nb, Ta, and Eu and depleted in the HREEs and Y relative to arc rocks (Whalen and Hildebrand, 2019). The majority of samples that meet the criteria of Whalen and Hildebrand (2019) plot in the slab-failure fields on these diagrams. They have high Nb/Y > 0.4, Sm/Yb > 2.5, Gd/Yb > 2.0, and La/Yb > 10 (Figs. 10A and 10B). On plots of Rb versus Nb + Y and Ta + Yb, some samples from the Scottish Grampian terrane, the Irish Grampian terrane, and the Northern Highlands terrane have Rb concentrations lower than typical slab-failure rocks (Figs. 10C and 10D). However, Rb is highly mobile during alteration and is primarily hosted in K-feldspar, which exhibits petrographic evidence for alteration in many samples (e.g., Archibald and Murphy, 2021). On plots of Y versus Nb and Yb versus Ta, which emphasize the concentrations of less mobile elements, most samples plot in the slab-failure field (Figs. 10E and 10F). Plots of Sr/Y versus Nb/Y and La/Yb also show that late Caledonian plutonic rocks have ratios compatible with a slab-failure setting (Figs. 10G and 10H).

Recent literature (e.g., Clemens et al., 2009b; Clemens and Stevens, 2012; Brown, 2013) advocates that the chemical composition of magmas is inherited from the magma source, and that intracrustal processes such as assimilation or crustal contamination have little effect on the chemical composition of magmas. Two processes potentially control the critical petrogenetic-indicating ratios that determine whether a sample plots in the slab-failure field. La/Yb, Sm/Yb, Gd/Yb, and Sr/Y are higher than typical arc rocks. Given the partitioning of the HREEs and Y into garnet, these elevated ratios may indicate that garnet was a stable phase in the residue. Alternatively, LREE and Sr source enrichment in the absence of HREE depletion has the same geochemical effect. The lack of a prominent Eu anomaly in the low-SiO<sub>2</sub> (<70 wt%) granitoid rocks (Figs. 7, 9G, and 9H) has been interpreted to indicate derivation from a plagioclase-free source (Hildebrand et al., 2018), implying that the presence of such an anomaly in the high-SiO<sub>2</sub> granitoids likely reflects low-pressure fractionation.

The isotopic data imply heterogeneous magma sources for the late Caledonian plutonic rocks. Canning et al. (1998) examined lamprophyres (representative of mantle sources) and interpreted the variation in  $\epsilon_{Nd}(t)$  values to reflect two distinct lithospheric mantle domains with a boundary corresponding with the Great Glen fault (Fig. 2), i.e., the boundary between the Northern Highlands and the Grampian terranes. The trace-element compositions of the lamprophyre dikes in the Northern Highlands terrane require an enriched lithospheric mantle source (Canning et al., 1998). Strongly negative  $\epsilon_{Nd}(t)$  values in the granitoids in both the Northern Highlands and Grampian terranes indicate derivation by either (1) melting of enriched subcontinental lithospheric mantle to produce mafic magmas that subsequently underwent fractional crystallization, or (2) melting of older crust (Halliday, 1984; Harmon et al., 1984; Dempsey et al., 1990). The significant

range in  $\epsilon_{Nd}(t)$  values from evolved (−12.6) to depleted mantle-like compositions in the Northern Highlands terrane (Fowler, 1992; Fowler et al., 2001, 2008) suggests that their compositions reflect varying contributions from crustal and mantle sources. In the Grampian terrane, higher  $\epsilon_{Nd}(t)$  values recorded by plutons (such as the Ardara pluton) with abundant mafic intrusive rocks, including appinites and lamprophyres, are consistent with a contribution from juvenile magmas derived from a depleted mantle source (Dempsey et al., 1990; Murphy et al., 2019). However, the presence of inherited zircon cores in the Donegal composite batholith also indicates an important role for crustal contamination in their genesis (Archibald et al., 2021).

Models for the petrogenesis of the late Caledonian plutonic rocks depend on interpretations of the regional tectonic setting during the Silurian to Early Devonian. Traditional models show magma generation above a NW-directed subduction system beneath the Laurentian margin (Fig. 11) during the Scandian orogeny (Dewey and Shackleton, 1984; Strachan et al., 2002). In that context, Atherton and Ghani (2002) adapted the NW-directed subduction model to suggest that magmatism occurred in the aftermath of “slab break-off” following Iapetus Ocean closure. In their model, Scandian imbrication and slab break-off began at ca. 435 Ma (Atherton and Ghani, 2002). After slab failure, hot asthenospheric mantle impinged on the base of subcontinental lithospheric mantle, producing mafic magmatism to form a lamprophyric underplate, mafic dikes, and appinite complexes. Melting of the mafic underplate generated the felsic to intermediate magmas (Atherton and Ghani, 2002). Rebound and exhumation of the upper plate (Laurentia) occurred as the slab detached. Rapid exhumation followed slab detachment, caused by the buoyant rise of asthenosphere and the heating of the lithospheric mantle (Neilson et al., 2009). Further sinking of the detached oceanic lithosphere, partial melting of the LILE- and LREE-enriched mafic underplate beneath the subcontinental lithospheric mantle, and extensive partial melting of the lower continental lithosphere produced the felsic to intermediate magmas (Neilson et al., 2009).

Although our model shows magma generation in the context of the more widely accepted NW-directed subduction, alternative models have been proposed. For example, Searle et al. (2019) and Searle (2022) proposed SE-directed subduction based on the structural and metamorphic history of northern Scotland (Fig. 10), in which the Scandian orogenic wedge and the Moine thrust zone represent a classic fold-and-thrust belt tapering to the west. In that context, these authors proposed a SE-dipping subduction zone beneath the Avalonian margin, with a continuous thickening, regional metamorphism, and deformation history evolving from the Grampian through to Scandian orogenic events in the Northern Highlands terrane (Searle et al., 2019; Searle, 2022). Oliver et al. (2008) proposed bilateral subduction of Iapetus Ocean lithosphere under both Ganderia-Avalonia and the Laurentian margin. In this model, slab roll-back at ca. 420 Ma was followed by bilateral slab failure at ca. 410 Ma, when Baltica hard-docked against the Northern Highlands terrane, and Ganderia-Avalonia

soft-docked against the Grampian terrane (Oliver et al., 2008; see also Miles et al., 2016; Miles and Woodcock, 2018). This bilateral slab failure effectively delaminated the lithosphere on both sides of the Iapetus suture zone, allowing hot asthenosphere to ascend under both the Midland Valley and Southern Uplands terranes (Oliver et al., 2008).

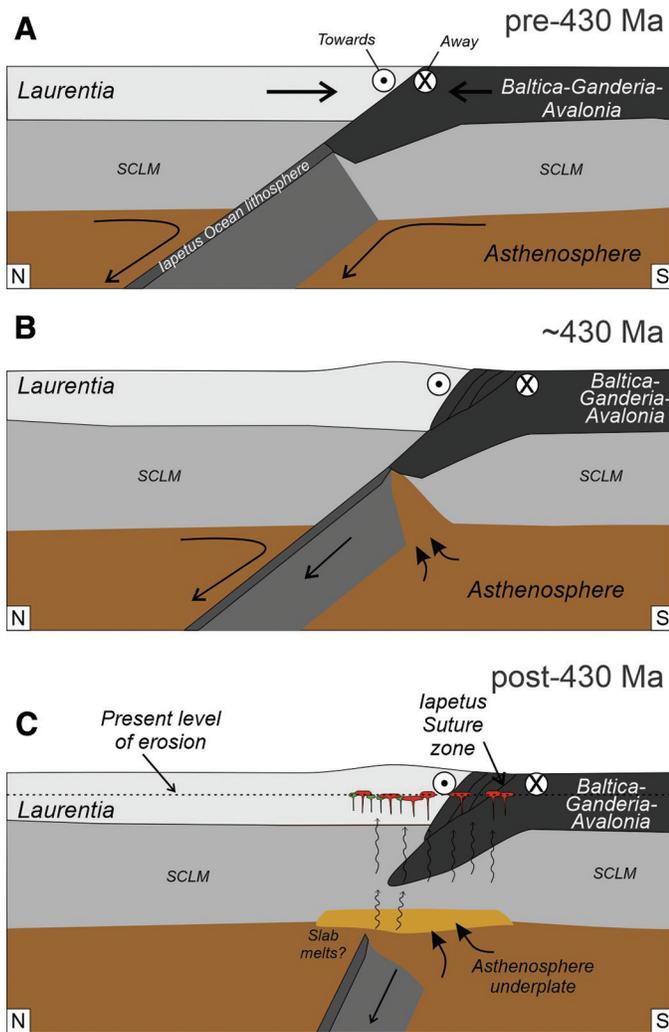


Figure 11. Simplified cartoon depicting the Silurian–Devonian tectonic evolution of the Irish and Scottish segments of Caledonian orogeny modified after Atherton and Ghani (2002), Neilson et al. (2009), and Archibald et al. (2021). (A) Beginning of subduction cessation and Scandian imbrication after closure of the Iapetus Ocean in the Silurian. (B) Oblique convergence at ca. 430 Ma accompanied slab weakening and incipient slab failure. (C) Slab failure after ca. 430 Ma, underplating of subcontinental lithospheric mantle by asthenosphere, and intrusion of appinite, lamprophyre, and the felsic to intermediate plutons. Upwelling asthenosphere underplated and melted the subcontinental lithospheric mantle to generate the mafic magmas. Alternatively, the melts could have been derived from melting of the detached oceanic lithosphere. Felsic to intermediate plutons were derived from either fractionation of the mantle-derived mafic magmas or by melting of the lower lithosphere. Abbreviations: SCLM—subcontinental lithospheric mantle.

In recent studies of postcollisional plutons, Hildebrand et al. (2018) and Whalen and Hildebrand (2019) proposed that magmas with  $Sm/Yb > 2.5$  were derived by melting at depths greater than arc magmas. According to their model, during arc-continent collision, the continental margin of the lower plate is pulled beneath the arc to create thickened lithosphere. The effect of pulling the cratonic margin beneath the arc on the petrogenesis of magmas produced in the aftermath of the Scandian orogeny was not considered in the slab break-off models of Atherton and Ghani (2002) and Neilson et al. (2009), or in the petrogenetic model of Fowler et al. (2008), who attributed the magmatism to be triggered by asthenospheric upwelling. However, Hildebrand et al. (2018) argued that the subducted cratonic edge underthrust the arc and isolated it and any lower arc material from the mantle. They suggested that magmatism related to rebound after slab failure was emplaced into rising, tectonically thickened crust, which contrasts in chemistry with precollisional arc magmas, which were emplaced into the relatively thin crust of the upper plate. After the oceanic lithosphere detached, rocks of the partially subducted continental margin rose rapidly due to buoyancy forces, causing exhumation and erosion of the tectonically thickened collisional hinterland. The distinctive slab-failure geochemical and isotopic signatures of the late Caledonian plutonic rocks (Archibald and Murphy, 2021) are hypothesized to be derived from the basaltic-gabbroic part of the subducted oceanic slab (Fig. 10; Hildebrand et al., 2018). When the slab fails deeper in the mantle, the asthenosphere, instead of upwelling, flows laterally to fill the gap (Freeburn et al., 2017; Hildebrand et al., 2018). In this scenario, the late Caledonian slab failure magmas would be mostly derived from the melting of garnet-bearing, plagioclase-absent metabasalt and gabbro. As these magmas ascended, fractional melting of the overlying old, enriched subcontinental lithospheric mantle would have modified their source-derived Sr and Nd isotopic ratios and LILE concentrations.

Taken together, the generation of the parental magmas for the late Caledonian plutonic rocks resulted from a thermal disturbance in lower lithosphere source regions, probably because of slab failure (Fig. 11; Atherton and Ghani, 2002; Neilson et al., 2009; Archibald and Murphy, 2021). The lower crust of the Grampian terrane had previously produced peraluminous granite magmas at ca. 470 Ma during the Grampian orogeny (Oliver et al., 2008). Therefore, more intermediate and mafic compositions likely dominated the lower-crustal hot zone (Annen et al., 2006) during generation of the late Caledonian plutonic rocks. These lower-crustal compositions would have yielded I-type magmas after the addition of mantle magmas or melts from the detached oceanic lithosphere. Granitoid magmas with an I-type mineralogy and shoshonitic geochemical affinities can be derived from fractional crystallization of subcontinental lithospheric mantle-generated mafic magmas with concomitant crustal contamination (Fowler et al., 2008), by the partial melting of hydrous, calc-alkaline, garnet-bearing and plagioclase-absent lithosphere (Roberts and Clemens, 1993), or by the contamination of partially melted, subducted oceanic lithosphere (including its sedimentary

components) by fractional melting of old, enriched subcontinental lithospheric mantle (Hildebrand et al., 2018). The abundance of hornblende in these calc-alkaline magmas, like those of the appinite suite, indicates water-saturated conditions (e.g., Murphy et al., 2019).

### Constraints on the Timing of Slab Failure

The precise timing of Scandian deformation is an important temporal constraint because convergent tectonic processes end when the oceanic slab detaches and slab pull no longer affects the lower plate (e.g., van Hinsbergen et al., 2014; Fig. 11). However, the best constraints on the timing of slab failure are from rocks preserved in the upper plate. A U-Pb zircon age of  $431.1 \pm 1.2$  Ma obtained from the syntectonic early phases of the Loch Borralan pluton dates brittle thrusting within the Moine thrust zone (Goodenough et al., 2011; Searle et al., 2019; Searle, 2022), although it does not constrain final displacement on the Moine thrust itself. Further east, the late- to post-tectonic Loch Loyal syenite is poorly constrained isotopically, but mineral ages of ca. 426–425 Ma provide an indicator that ductile deformation within host Moine rocks must have ceased by that stage (Goodenough et al., 2011; Walters et al., 2013). These constraints are compatible with U-Pb monazite ages of ca. 430–425 Ma obtained from adjacent upper-amphibolite-facies gneisses of the Naver nappe (Mako et al., 2019; Strachan et al., 2020a) and with U-Pb zircon ages of ca. 432–426 Ma obtained from syntectonic granites that were intruded along the Naver thrust (Strachan et al., 2020a). Accordingly, high-grade metamorphism and contractional deformation within the central Scandian nappes in northern Scotland persisted until ca. 425 Ma, and it is possible that marginal thrusting did not terminate until as late as 420–415 Ma, implying a protracted evolution and diachronous termination of Scandian deformation (Strachan et al., 2020a, and references therein). After ca. 420–415 Ma, the tectonic environment is thought to have switched from a dominantly convergent setting to a dominantly strike-slip regime (Snyder and Flack, 1990).

Geochemical data from the apparently syn-Scandian (ca. 430 Ma) Loch Ailsh and Borralan granitoid plutons and apparently post-Scandian granitoid plutons (younger than ca. 425 Ma) in the Northern Highlands terrane are indistinguishable and plot in the slab-failure field on discrimination plots (Fig. 10). Litho-geochemical comparisons of pre-, syn-, and post-slab failure plutons from the Grampian and Connemara terranes are more challenging because the timing of pluton emplacement relative to Scandian deformation is not well constrained. In the Irish Grampian terrane, a ca. 437 Ma lamprophyre dike containing a magmatic foliation parallel to a country rock foliation is interpreted to reflect synkinematic emplacement during NW-SE (Scandian) compression (Kirkland et al., 2013). This dike predates the ca. 430 and 400 Ma emplacement of the Donegal composite batholith (Archibald et al., 2021). Movement on the Leannan fault (Fig. 2), a major splay of the dominantly strike-slip Great Glen fault, is associated with the emplacement of granitic magmas at

ca. 412 Ma (Kirkland et al. 2008). Based on these constraints, the cessation of deformation, interpreted to constrain the timing of slab failure, occurred at ca. 430 Ma, and the Borralan and Loch Ailsh plutons are the oldest slab-failure plutons.

The transition from contractional to strike-slip tectonics in Scotland occurred at ca. 425 Ma, although partitioned thrusting and strike-slip movements above a regional décollement may have occurred between ca. 420 and 415 Ma (Strachan et al., 2020a). The timing of this transition is unconstrained in Ireland. Unlike intrusions from the western United States, which display an abrupt change from arc to slab-failure geochemical signatures over a few million years (Hildebrand and Whalen, 2014b, 2014a, 2017; Hildebrand et al., 2018), there is no a precise temporal constraint for an equivalent change in the Caledonian orogeny, implying this transition was likely more protracted. Unequivocal early Silurian arc rocks are not preserved on either side of the Iapetus suture in Britain or Ireland (Chew and Strachan, 2014). However, these arc rocks could be preserved below the present level of exposure in the Midland Valley terrane, in conglomerate clasts or xenoliths hosted in Permian to Carboniferous dikes (Biejat et al., 2018; Badenszki et al., 2019). In addition, provenance studies indicate a possible arc terrane outboard of the Laurentian margin in the Silurian (Biejat et al., 2018).

Slab failure can be diachronous, initiating at the point of initial collision along irregular continental margins and propagating laterally along the slab (van Hunen and Allen, 2011; Boutelier and Cruden, 2017; Fernández-García et al., 2019). Such a scenario is consistent with models for highly oblique, sinistral, and diachronous closure of the Iapetus Ocean in the Silurian (Dewey and Strachan, 2003). Additional litho-geochemical and high-precision geochronological data from plutons coupled with the ages of fabrics in their country rocks are required to investigate the potential diachronous timing of slab failure, the rates of slab failure, and its tectonothermal consequences.

The more shoshonitic compositions in mafic rocks in the Northern Highlands terrane relative to the Grampian terrane (Figs. 6 and 9) suggest these terranes were underlain by a compositionally distinct subcontinental lithospheric mantle in the early Silurian, with the subcontinental lithospheric mantle beneath the Northern Highlands being comparatively older, more extensively metasomatized, and more enriched in LILEs. The presence of appinites on both sides of the Great Glen fault indicates that water was important in the petrogenesis of the mafic magmas. However, the Nd-Sr isotopic compositions of coeval plutons on both sides of the Great Glen fault are indistinguishable (Fig. 8), indicating that the metasomatic event likely occurred during subduction of Iapetan oceanic lithosphere immediately prior to slab failure. The subcontinental lithospheric mantle of the upper plate was likely metasomatized by the subduction that preceded slab failure, and the subducting slab was likely metasomatized before and during subduction. Taken together, these data suggest fundamental differences in the composition of the mantle lithosphere or source region on either side of the Great Glen fault, which separates the Northern Highlands and Grampian terranes.

### **Relationship between Mafic and Felsic to Intermediate Magmatism**

In the Irish and Scottish segments of the Caledonian orogen, mafic and felsic to intermediate rocks display intimate field relationships suggestive of mingling and mixing of coeval mafic and felsic magmas. In addition, mafic magmatism both predated and continued during emplacement of the felsic to intermediate plutons, as shown by the presence of mafic enclaves in granitoid plutons and by geochronological data that demonstrate mafic enclaves are the same age as their felsic to intermediate host rocks (Archibald et al., 2021). The litho-geochemical and isotopic signatures of mafic intrusive rocks are similar across the Grampian terrane of Scotland and Ireland but differ from mafic rocks in the Northern Highlands terrane, which were derived from a relatively enriched mantle. Similarly, plutons are geochemically indistinguishable across the Grampian and Connemara terranes but differ from plutons in the Northern Highlands terrane, which, like their mafic counterparts, are more enriched in the LREEs and LILEs. Three models have been proposed for the genetic relationship between coeval mafic and felsic to intermediate magmatism. They are not mutually exclusive, but the relative balance between them requires further detailed study.

### **Fractionation with Crustal Contamination**

The similarity in trace-element and REE compositions in the coeval mafic and felsic to intermediate rocks (Fig. 7) has been interpreted to reflect crystal fractionation in which the distinctive metasomatic signatures in the subcontinental lithospheric mantle-derived mafic magmas were inherited by the felsic to intermediate magmas (Fowler, 1988; Fowler and Henney, 1996; Fowler et al., 2001, 2008). For example, in the Northern Highlands terrane, Fowler et al. (2008) suggested that mafic (appinitic) magmas were derived from the melting of a mantle source variably enriched by subducted sediment and fractionated to produce the high-Ba-Sr granitoids, with the wide range of silica saturation reflecting significant crustal input. If so, mantle metasomatism may have had a fundamental influence on the trace-element and isotopic composition of the felsic to intermediate plutons. If crystal fractionation was the dominant process responsible for generating the felsic to intermediate magmas, then large volumes of mafic to intermediate rocks possibly exist below the current level of exposure (Murphy, 2020).

### **Melting of the Detached Oceanic Lithosphere**

The distinctive slab-failure geochemical signatures may have been generated by melting of a steeply dipping subducted oceanic slab (Hildebrand et al., 2018). In this model, (1) melting of the upper layers of subducted oceanic lithosphere generated mafic magmas that were chemically modified as they ascended through the old and enriched continental lithosphere; (2) fractional melting of old, metasomatized subcontinental lithospheric mantle generated the felsic to intermediate magmas (Hildebrand et al., 2018; Whalen and Hildebrand, 2019);

and (3) alkalic rocks (such as the Borralan and Loch Ailsh plutons) in Scotland may represent melting of subcontinental lithospheric mantle contaminated by rift-facies continental crust derived from the ancient, rifted portion of the subducted continental margin that included evaporites and alkaline basalt (Burke et al., 2003; Hildebrand et al., 2018). However, because the alkali rocks are silica undersaturated, the crustal contamination must have been minor.

### **Asthenospheric Upwelling**

A third model invokes asthenospheric upwelling following slab failure, in which the upwelling would have provided sufficient heat to melt metasomatized subcontinental lithospheric mantle and mafic underplate to trigger the late Caledonian magmatism (Atherton and Ghani, 2002; Neilson et al., 2009). In this model, after slab failure, hot asthenospheric mantle advected heat to the base of subcontinental lithospheric mantle, causing the melting that generated the mafic magmas that were emplaced as lamprophyric dikes and appinite complexes. Partial melting of the LILE- and LREE-enriched subcontinental lithospheric mantle mafic underplate (Atherton and Ghani, 2002) or extensive partial melting of the lower continental lithosphere (Neilson et al., 2009) produced the felsic to intermediate magmas.

## **CONCLUSIONS**

The published geochemical data used many different techniques and approaches, making it challenging to compare data from late Caledonian plutonic rocks across the region. Our review highlights the need for a systematic study of felsic to intermediate and mafic plutons in Scotland residing on both sides of the Great Glen fault. Such a study should include carefully selected samples for strategic U-Pb dating and petrochronologic and litho-geochemical analysis to precisely constrain the timing of slab failure and determine whether discrete plutons represent arc, transitional, or slab-failure magmas. Constraining the timing of slab failure would help to resolve the issue of diachroneity and the potential role of promontories. Resolving the contrasting models would also require a detailed study of the age of antecrysts (magma incubation) and autocrysts (magma emplacement). Coupling these age data with isotopic data could resolve the issue of whether slab-failure magmas were derived by melting of oceanic lithosphere, metasomatized subcontinental lithospheric mantle, or crust and the extent of chemical modification of these magmas as they ascended.

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