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Garnet stability during crustal melting: Implications for chemical mohometry and secular change in arc magmatism and continent formation

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A R T I C L E I N F O

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A B S T R A C T

Understanding how new felsic crust is formed and subsequently evolves through time is critical to identifying the geodynamic regimes that have dominated various parts of Earth history, and have important implications for feedbacks between the lithosphere and biosphere, such as controlling the influx of continental detritus into the oceans. In recent years, several trace element-based geochemical proxies have been proposed to allow determination of paleo-crustal thicknesses, which have been calibrated primarily using data collected from modern-day arcs. The application of these proxies through deep time has revealed surprising results, including the suggestion that the mid-Proterozoic continents were atypically thin compared to those in the Archean and the Phanerozoic; however, a range of factors may influence commonly cited trace element ratios (e.g. Sr/Y) rather than just crustal depth, leading to additional and unexpected magnitudes of uncertainty. Here we perform geochemical modelling to deduce the effect of variable bulk-rock composition and geothermal gradient on the trace element signature of felsic melts generated in arc systems. Using a range of protoliths representative of deep arc crust, the results show that considerable care must be taken when analysing simple trace element ratios of granitoid melts and making direct interpretations of the pressure of crystallisation. In particular, changes in geothermal gradients and differences in arc basalt composition impart strong controls on the relative stability of garnet and plagioclase during metamorphism and partial melting, and wide ranges of Sr/Y and La/Yb may be produced in derivative felsic melts produced at the same crustal depth. The interpretation of mid-Proterozoic continental arcs being atypically thin may instead be an artefact of underestimation of the active geothermal gradient at the time of magma formation, which acts to reduce Sr/Y and La/Yb ratios, even in normal thickness (~35–40 km) crust. Furthermore, we argue that the potentially garnet-free residua during the formation of mid-Proterozoic felsic magmas points to crust formation without lower crustal foundering, and thus, that this commonly invoked paradigm for formation of the continental crust may only be applicable to certain periods of Earth history.

1. Introduction

Metamorphism and partial melting within the continental crust are key petrological processes that allow chemical differentiation and drive its long-term stabilisation (Clemens, 2006). As a result, the architecture and composition of Earth's continents have changed significantly through geological time (Rollinson, 2008). A key paradigm within the community is that Phanerozoic continents are typically thicker and more silicic than immature, undifferentiated, and thinner Archean equivalents (see Cawood and Hawkesworth, 2019 and references therein); however, this has been recently questioned by several lines of evidence that point to Archean continental crust having a composition (Keller and Harrison, 2020; Lipp et al., 2021; Ptáček et al., 2020) and thickness (Tang et al., 2021b) similar to that of Phanerozoic continental crust. A range of geochemical proxies have been developed in recent years to estimate crustal thickness, now informally known as chemical mohometers (Fig. 1: Luffi and Ducea, 2022), and include bulk-rock Sr/Y ratios (Chapman et al., 2015) and Eu anomalies in zircon (Tang et al., 2021a). Such proxies exploit the varying stability of key metamorphic minerals as a function of depth within the crust, such as garnet and plagioclase, which are known to stabilise at relatively high and low pressures, respectively, in a wide range of rock types (Ferry, 2000).

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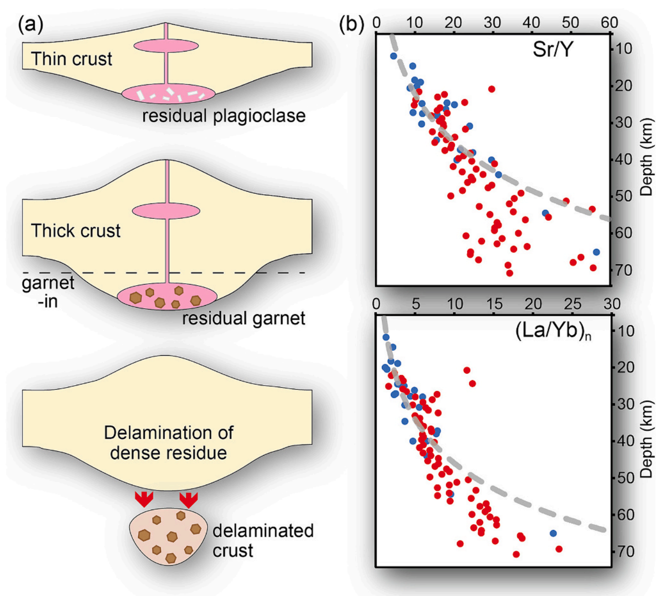

Fig. 1. Simplified conceptual model of chemical mohometry. (a) Magmatism at the base of thin continental crust occurs within the plagioclase stability field, but outside that of garnet. Magmatism in thick crust occurs within the garnet stability field, forming dense residues that can founder or delaminate into the mantle, thereby increasing the bulk SiO_2 content of the remaining continental crust. (b) Sr/Y and (La/Yb)n ratios in modern volcanic arc whole-rock compositions correlate with depth to the Moho, forming the basis of these ratios being chemical mohometers. These arguably relate to residue minerals sequestering these elements from the rising melts, such as Y and Yb in garnet, and Sr in plagioclase. Grey dashed lines show the calibrations of Sundell et al. (2021); blue and red dots are from Profeta et al. (2015) and Farner and Lee (2017), respectively. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

However, with secular changes in average continental composition, hydration state, and structure, it is not straightforward how to interpret changes in these geochemical tracers through time, and equate them directly to variation in crustal thickness or depth of melting.

The stability of minerals and melts in metamorphic rocks is independently controlled by pressure (*P*), temperature (*T*), and bulk composition (*X*) (Essene, 1989). Scatter in the above-mentioned geochemical proxies for crustal thickness for any period of Earth history may therefore alternatively record changes in bulk composition or changes in the crustal geotherm, or both. Examining the importance of these variables is key to understanding crustal evolution through time, and deciphering gradual and abrupt changes in the metamorphic rock record. The major rock-forming minerals commonly used for investigating variation in crustal thickness and/or depth of melting include garnet, plagioclase, hornblende, and rutile, all of which are common in meta-mafic bulk compositions, are readily involved in subsolidus and suprasolidus metamorphic reactions, and are sensitive to magmatic crystallisation processes (Martin, 1987). Such studies have been performed for the growth of Archean and Phanerozoic continental crust via both experimental (e.g. Moyen and Stevens, 2006) and modelling (e.g. Palin et al., 2016a) approaches; the Proterozoic, however, has received much less attention with these methods. In particular, secular trends in average crustal Sr/Y ratios (and other proxies for plagioclase/garnet behaviour) in newly formed felsic magmas show a conspicuous drop during the mid-Proterozoic (Tamblyn et al., 2022; Roberts et al., 2023), which may indicate that continents formed during Earth's middle age were atypically thin (Tang et al., 2021b), or that magma generation occurred under higher geothermal gradients (Tamblyn et al., 2022).

To address this issue, and to place new constraints on the potential thickness of average Proterozoic continental crust, we have modelled the stability of major rock-forming minerals in four meta-mafic compositions that represent a broad range of lower continental crust lithologies that may undergo partial melting. Building on previous studies, we demonstrate that the stability of garnet and plagioclase are sensitive to secular changes in bulk maficity, and the compositions of partial melts generated are controlled as much by the characteristics of the protolith as they are by the *P-T* conditions of anatexis. We then compare predicted trace element ratios in modelled melt fractions generated in arc-like settings against natural data, which show that interpretation of crustal thickness or paleo-elevation from Sr/Y or La/Yb data is overly simplistic without considering the influence of bulk-rock composition on the stability of rock-forming minerals. This is particularly important when interpreting changes in crustal architecture through time. We argue that mid-Proterozoic continental crust may have exhibited a 'normal' thickness of 35–40 km, with these conditions prohibiting the formation of dense residues. Importantly, our data imply that the model of continent formation involving crustal foundering during arc magmatism may only have been possible during some periods in Earth history.

2. Materials and methods

2.1. Petrological modelling

Petrological phase equilibrium modelling was performed to determine mineral and melt stability in several rock types at *P–T* conditions spanning the upper crust to a Moho depth of ~75 km (2–22 kbar and 500–1050 °C). We also calculated bulk-rock density, volumetric phase proportions, and major and trace element composition of melt generated along linear geotherms of 50, 75, and 100 °C/kbar, which cover a range of thermal states that occur at convergent margins. We considered three mafic lithologies as analogues for lower continental crust in arc-like settings: a medium-K, high-alumina Cascades basalt 87S35A from Sisson et al. (2005), the statistically determined 'deep crust' (DC) from Sammon and McDonough (2021), and the average Cascades 'arc basalt' composition from Schmidt and Jagoutz (2017). We also considered the average enriched Archean tholeiite (EAT) basalt composition from Condie (1981) to compare between modern-day and ancient mafic compositions. All modelling was performed in the $Na_2O–CaO–K_2O–FeO–MgO–Al_2O_3–SiO_2–H_2O–TiO_2–O_2$ (NCKFMASHTO) compositional system using the Gibbs free energy minimization software Theriak–Domino (de Capitani and Petrakakis, 2010) and the internally consistent thermodynamic data set ds62 (Holland and Powell, 2011), with further details provided in the Supplementary Information. The results of each petrological model are thought to be relatively accurate to around 0.2 kbar and 10 °C (2σ) (Worley and Powell, 2000).

2.2. Trace element modelling

Geochemical modelling was performed to calculate the concentrations of trace elements in melt fractions generated from each protolith as a function of depth within the crust. All calculations considered equilibrium partitioning between minerals in the residuum and coexisting silicate melt fractions using the equation of Shaw (2006). Our modelling considered closed-system conditions and the continuous in-situ accumulation of melt with increasing depth in order to examine the change in trace element ratio of partial melts formed along certain geotherms in arc environments. While experimental and field studies have shown that partial melt can form a connected network along grain boundaries at small volumes (~7 vol%; Rosenberg and Handy, 2005), which leads to a dramatic drop in rock strength, this rheological transition does not necessarily allow for voluminous melt escape. As noted by Vigneresse et al. (1996), the major structural change corresponding to breakdown of the solid (crystalline) framework in a rock undergoing anatexis, which then allows large-scale melt escape, is likely to depend on additional factors, such as melt viscosity and strain rate. This melt-escape threshold (MET), which is of more relevance for the generation of granitic intrusions at higher crustal levels, occurs at a melt fraction of at least 25 vol% (Sawyer, 1994; Vigneresse et al., 1996). Previous modelling of partial melting in mafic rocks in the middle to deep crust has shown that these volumes are typically reached at around ~900 °C (Palin et al., 2016b), which is towards the upper limit of our modelling. Consequently, we avoid complications related with intermittent melt escape in this work, and focus solely on the relationships between *P-T* and composition (*X*) with the aim of determining how the trace element profile of a melt and residue change with respect to crustal depth.

2.3. Secular change in granite geochemistry

Secular variations in granite geochemistry were assessed using the compilation of Roberts et al. (2023), which builds on the database of Gard et al. (2019) by the addition of data from Proterozoic units (Fig. 2). Following Sundell et al. (2021), we filtered all analyses to ensure SiO_2 = 57–68 wt%, MgO = 0–4 wt%, and bulk Rb/Sr = 0.05–2.0. Data are displayed as scatter plots for Sr/Y (Fig. 2a) and $(La/Yb)_n$ (Fig. 2b) with the addition of locally weighted smoothing (LOWESS) curves with 95% confidence intervals (calculated with a 10% moving window).

In general, these values show notable scatter through time, with episodes of supercontinent amalgamation recording the highest concentrations of data. The LOWESS curve for Sr/Y (Fig. 2a) ranges between values of ~5 and ~ 30, with minima documented during the Mesoproterozoic to Neoproterozoic (~5–10). Direct application of these values to determine equivalent crustal thicknesses using the regression of Sundell et al. (2021) produces a range of ~10–45 km. By contrast, the LOWESS curve for $(La/Yb)_n$ exhibits a fairly narrow range of values through time, from ~10 to ~20 (Fig. 2b), correlating to approximate crustal thicknesses of 40–60 km. Notably, unlike for Sr/Y, there is no pronounced period of low values during the Proterozoic.

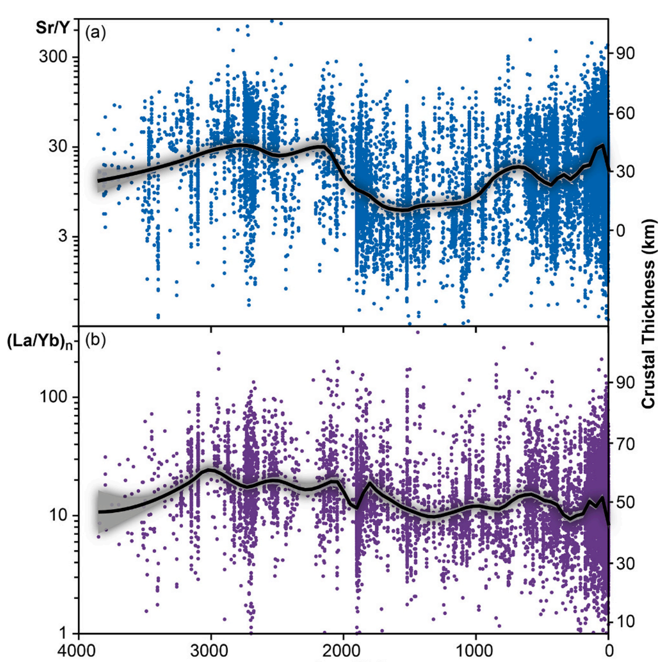

Fig. 2. Global whole-rock geochemistry (data from Roberts et al., 2023), filtered to 57–68% SiO_2, 0–4% MgO and 0.05–2.0 Rb/Sr, and plotted as Age (Ma) vs Sr/Y (a) and chondrite-normalised $(La/Yb)_n$ (b). Crustal thickness calculated using the regressions of Sundell et al. (2021). Black band is a locally weighted smoothing (LOWESS) curve with 95% confidence intervals in grey.

3. Results

Calculated *P-T* pseudosections for all four mafic bulk compositions are shown in the Supplementary Information. In Fig. 3a, we summarise the main features of these phase diagrams by showing isopleths for the volumes of garnet and plagioclase (vol%) for each lithology; these plots demonstrate the significant variability in garnet and plagioclase stability across the P-T conditions of the continental crust, which in turn affect Sr/Y and La/Yb ratios of melts derived from different residua.

In general, all four mafic lithologies exhibit a fluid-saturated solidus that lies at similar P-T conditions, with the onset of melting beginning at ~610–620 °C and ~ 10–14 kbar. Plagioclase stability in the suprasolidus field shows an inverted-U shape for all samples except high-alumina Cascades basalt 87S35A from Sisson et al. (2005) (Fig. 3a). Isopleths for plagioclase volume resemble the geometry of its limit of stability ("Pl-out"), with feldspar consumed during melting up-temperature through the granulite facies. Garnet stability varies with protolith composition, with its calculated minimum pressure of formation varying between 8 and 14 kbar (EAT), 10–16 kbar (DC), 10–17 kbar (87S35A), and 12–16 kbar (Cascades). These differences are independent of MnO content, which was excluded as a component in our modelling.

The relative stability of plagioclase and garnet in the presence of melt for mafic bulk-rock compositions can be used to divide the phase diagram into four discrete regions: (1) a low-temperature region where neither plagioclase nor garnet are stable, typically immediately suprasolidus at P ~ 12–16 kbar; (2) a region where both garnet and plagioclase are stable in varying proportions; (3) a region where only garnet is stable (high pressure); and (4) a region where only plagioclase is stable (low pressure). The morphology, position, and size of region (2) where both plagioclase and garnet are stable varies considerably between protoliths, indicating that calculated Sr/Y and/or La/Yb ratios of melts derived from different residua at a lower crustal pressure of 12 kbar, for example, would be notably different.

For comparison with the results of our modelling, Fig. 3b shows experimentally derived stability curves for garnet and plagioclase in arc basalt and tholeiite with varying proportions of H_2O (Blatter et al., 2023; Loucks, 2021). We note that these experiments cover a broader range of temperatures than our calculations, and are relevant also to crystallisation of basaltic melts at high pressures rather than only examining partial melting behaviour. Nonetheless, the patterns recorded in these experimental data are similar to our results, with three of the four above-mentioned mineralogical domains identified here. In particular, the absolute P-T conditions over which garnet and plagioclase may coexist in the presence of melt are shown to vary considerably, ranging from as low as ~800 °C (H_2O-saturated tholeiite; Loucks, 2021) to up to 1100 °C (2 wt% H_2O basalt; Blatter et al., 2023).

4. Discussion

4.1. Compositional control on mineral stability

Since the most commonly used proxies for depth of melting and crustal thickness draw upon the variable partitioning of trace elements between plagioclase and garnet (e.g. Tang et al., 2021a), we focus on the stability of these phases. The results of our modelling show significant variability in the stability of both garnet and plagioclase across the four compositions. Garnet is suppressed to the deepest pressures in the Cascades arc basalt, and stable at the lowest pressures in the EAT. These results are comparable to the modelling of Kendrick and Yakymchuk (2020) and Yakymchuk et al. (2023), who both compared the EAT starting composition with that of a depleted Archean tholeiite (DAT) and a high MgO basalt; although a notable difference is seen in the latter, which has a pronounced deficit in both garnet and plagioclase at typical mid- to lower-crustal conditions.

Since the experimental work of Alonso-Perez et al. (2009), garnet

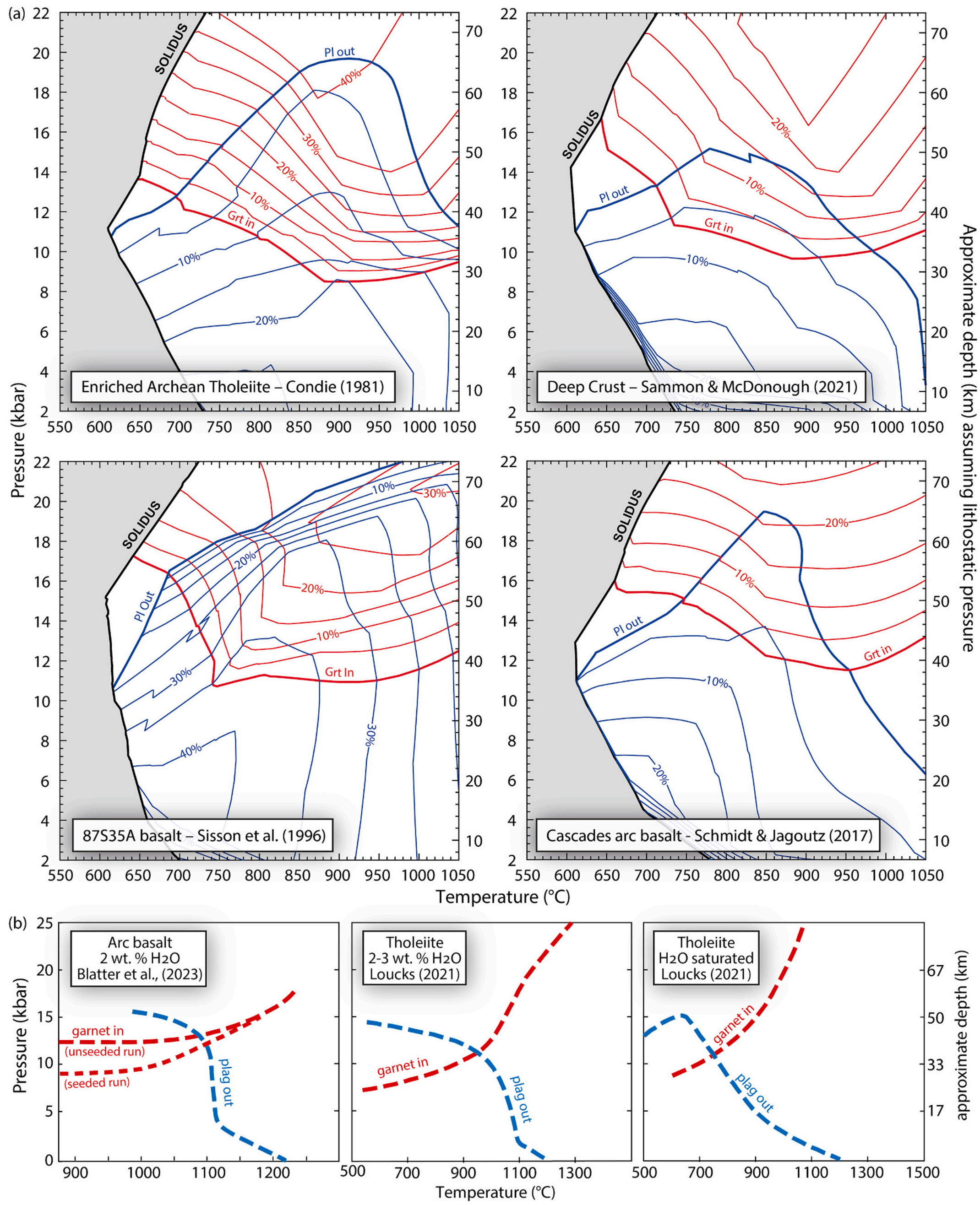

Fig. 3. (a) Results of thermodynamic modelling of four mafic starting compositions, plotted as garnet (red) and plagioclase (blue) isopleths at 5 wt% volume intervals; see text for full methodology. (b) Garnet-in (red) and Plagioclase-out (blue) isopleths based on compilations of experimental melting for three different conditions; see text for full description. Isopleths are estimated from data presented in Loucks (2021) and Blatter et al. (2023). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

stability has commonly been cited as occurring at pressures >12 kbar, equivalent to ~35 km. The negative slope of the garnet-in isopleth indicates a temperature dependence to garnet stability in the region of dehydration melting. Crustal temperatures, particularly those of the mid- to lower-crust during active arc magmatism vary greatly; however, felsic magmas formed by crustal anatexis within lower arc crust are likely to be in the range 750–900 °C (e.g. Collins et al., 2020). At this temperature range, the depth of the garnet-in isopleth has significant variation, falling between 30 and 50 km. Plagioclase is well known to be stable at shallow crustal pressures, and absent at high pressures; however, as with garnet, our modelling shows significant variation. The plagioclase-out isopleth at temperatures of 750–900 °C ranges from 40 to 70 km.

Our results use thermodynamic modelling to assess mineral stability. An alternative method is to use experimental petrology to melt specific starting compositions at a range of P-T conditions, and then measure the composition and modal proportions of the minerals and quenched melt. The topology of mineral isopleths can be estimated from multiple experiments across a range of P-T space, but overall, volumetric isopleths are less well constrained. In Fig. 2b, we present garnet and plagioclase-in isopleths for three compilations: tholeiitic starting compositions under wet (2–3 wt% H_2O) and 'damp' (H_2O saturated) conditions based on the compilations of Loucks (2021), and a basalt under wet conditions (2 wt % H_2O) based on the experiments of Blatter et al. (2013, 2023). The P-T range of these plots covers a different range to those of our pseudosections, but the broad shape of the garnet- and plagioclase-in isograds can be compared. The garnet-in isopleths have a shape that is markedly different to those of the thermodynamic modelling, with a particularly strong temperature dependence in the tholeiitic compilations. At 750–900 °C, the garnet-in isopleth broadly corresponds to depths of 30 to 40 km. The plagioclase-out isopleth has a strong temperature dependence, and lacks the positive slope at low temperatures; at high temperatures the shape has some similarity to all but one composition (that of 87S35A).

The primary factors known to control mineral stability in metamorphic rocks include *P*, *T*, and the composition of the protolith itself (*X*), including its hydration state. Similar to our study, Kendrick and Yakymchuk (2020), Triantafyllou et al. (2022) and Yakymchuk et al. (2023) modelled a variety of mafic compositions and discussed the variable stability of garnet and/or plagioclase based on pressure and temperature. Triantafyllou et al. (2022) used compositions that were chosen to mimic average mafic compositions evolving through Earth history. Kendrick and Yakymchuk (2020) and Yakymchuk et al. (2023) used compositions that are typically associated with the formation of Archean TTGs. Our study builds upon these previous works, but we focus on the diversity of lower crustal mafic compositions that may exist today; and thus, the variability in mineral stability that may exist in modern environments.

Given garnet's primary role in thermobarometry and as a metamorphic timekeeper (Baxter and Scherer, 2013) means that the compositional factors that promote garnet growth in metabasic rocks are of key importance for investigating the growth and evolution of the continental crust. Our results suggest that the minimum pressure at which garnet can stabilise in the presence of melt is correlated with total alkalis; specifically, mafic protoliths with higher $Na_2O + K_2O$ suppress the onset of garnet growth to higher pressures (cf. ~11–12 kbar for 87S35A and the cascades basalt, compared to ~8–9 kbar for DC and EAT), reflecting the expansion of the plagioclase stability field. The volume of garnet that is predicted to stabilise also increases with bulk-rock FeO/MgO ratio, with model results for EAT indicating up to 40 vol% garnet present at >18 kbar (~60 km) (Fig. 3a) compared to ~15–25 vol% for the other three protoliths.

Regarding hydration state, previous studies have quantified the role of variable water content on the stability of plagioclase and garnet (Pourteau et al., 2020; Hernández-Montenegro et al., 2021; Triantafyllou et al., 2022; Wang et al., 2022). For example, Pourteau et al. (2020) studied a suite of Proterozoic TTGs in NE Australia, and demonstrated high Sr/Y ratios despite only moderate depths of melting. These high ratios resulted from fluid-fluxed melting where sufficient water infiltrated the system to cause oversaturation, causing the stability of plagioclase to decrease and also for it to be consumed during prograde mineral reactions. This suppression of plagioclase at high water contents and its effect on trace element contents of the melt has been further modelled in other recent studies, including Hernández-Montenegro et al. (2021), Triantafyllou et al. (2022) and Wang et al. (2022). Collectively, our results along with those of previous studies demonstrate that variations in the composition and hydration state of the lower crust play a significant role on mineral stability during melt formation.

4.2. Implications for chemical mohometry

It has long been demonstrated that certain elements or elemental ratios in arc rocks correlate with the depth to the Moho, i.e. the crustal thickness (e.g. Plank and Langmuir, 1988; Mantle and Collins, 2008). These include both whole-rock and mineral-based compositional proxies, and have recently been reviewed by Luffi and Ducea (2022), who also introduced the term 'chemical mohometry'. Although there are a range of individual mohometers, two of the most commonly used whole-rock proxies are the trace element ratios La/Yb and Sr/Y. These have been calibrated against crustal thickness in several studies (Profeta et al., 2015; Chapman et al., 2015; Hu et al., 2017; Lieu and Stern, 2019; Sundell et al., 2021; Luffi and Ducea, 2022). The cause of the correlations between La/Yb and Sr/Y with depth is generally ascribed to increasing stability of garnet with depth, which has greater compatibility for HREEs than most other common rock-forming minerals, and to decreasing plagioclase stability with depth. It should be noted, however, that the cause of high Sr/Y in magmatic rocks—one of the primary signatures of adakites—has been extensively discussed with multiple hypotheses beyond simple fractionation of garnet that sequesters Y and Yb from partial melts (Moyen, 2009; Hernández-Uribe et al., 2020; Wang et al., 2022).

Fig. 4 shows calculated $(La/Yb)_n$ and Sr/Y ratios of melts generated from various protoliths along 50, 75, and 100 °C/kbar geotherms, shown as a function of depth below the Earth's surface. These data show a broad increase in both ratios with depth, although the extent of the grey region, which delimits the total range of values for all scenarios, indicates the large potential ranges that may occur in melts produced in arc systems at the same crustal depth. Alongside the factors already discussed, differences in individual protolith (i.e. pre-melting) trace element characteristics will have a noticeable effect on these results. For example, Sr/Y ratio in the starting compositions ranges from ~7.8 to 49; this variation in initial composition will clearly influence the range of ratios in the subsequent melts, and indicates that these starting compositions have evolved significantly from primary mantle-derived melts. The initial melts produced have significant variations, ranging from ~25 to 166 at 50 °C/kbar, ~6.8 to 59 at 75 °C/kbar, and ~ 3.6 to 19 at 100 °C/kbar. In addition, modelled ratios for each individual lithology do not evolve consistently along or between each geotherm (e.g. with progressive increases in *P* and *T*). For example, the La/Yb ratios of melts at the highest geothermal gradients (100 °C/kbar) tend to progressively decrease with depth; however, for intermediate (75 °C/kbar) and cool (50 °C/kbar) geothermal gradients, most protoliths produce melts fractions that exhibit increasing and then decreasing ratios with depth. Similar complexities are present in some Sr/Y results (Fig. 4). These results therefore support the key observation that a single elemental ratio cannot be reliably used to infer crustal depth; for example, according to our calculations, a $(La/Yb)_n$ ratio of 70 could represent depths of anywhere between 22 and 44 km, depending on protolith composition and geothermal gradient.

We note there are several limitations to our modelling, such as uncertainties in the phase equilibrium models, the partition coefficients used, and the assumption of a closed system. Kendrick and Yakymchuk (2020) conducted similar modelling to that presented here, including the same EAT starting composition. In addition, these authors compared the influence on trace element ratios of different model types, a closed system scenario, an open system scenario, and one in which garnet is fractionated. The results show that the overall trace element ratios are similar, but that the rate of enrichment (in e.g., Sr/Y and La/Yb) varies across these model types. We plot their results of the EAT starting composition in Fig. 5a to demonstrate this; again, the variability demonstrates that specific trace element ratios should not directly be tied to specific depths in a generic sense. Another limitation of our modelling is that we have only considered fluid-saturated melting, and as discussed in the previous section, oversaturation leading to fluid-fluxed melting will significantly affect mineral stability, notably the suppression of plagioclase. The effect of this plagioclase suppression leading to higher melt Sr/Y ratios has been modelled by several previous studies, which we demonstrate with the examples in Fig. 5b.

Despite significant complexity and variation, our model results show broadly increasing elemental ratios (La/Yb and Sr/Y) with depth when viewed as a whole, which is in agreement with the concept of these ratios being used as chemical mohometers. Nonetheless, published calibrations have large associated uncertainties within the range ± 5–10 km (e.g. Sundell et al., 2021; Luffi and Ducea, 2022), which may be attributed to 'geological noise' in the calibration procedures, such as taking the mean of several analyses from specific regions and/or comparing measured ratios to typical levels (depths) of exposure in a terrain. Our modelling, along those of previous studies, demonstrates

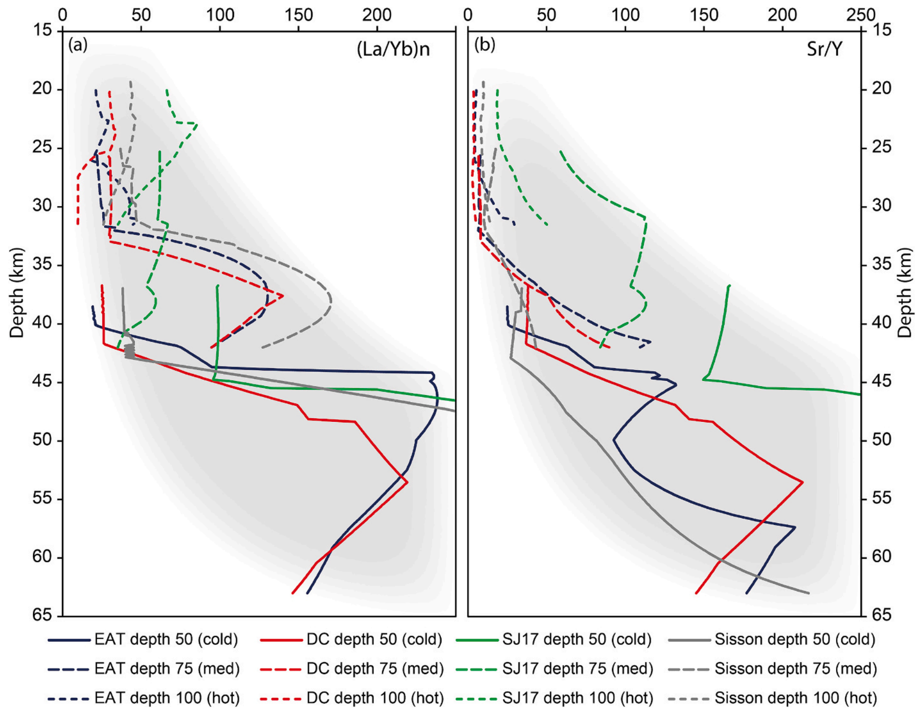

Fig. 4. Plot of calculated $(La/Yb)_n$ (left) and Sr/Y (right) ratios of melts generated from various protoliths along 50, 75, and 100 °C/kbar geotherms, shown as a function of depth below the Earth's surface. Grey field represents an envelope that brackets all values, illustrating the potential ranges that may occur in melts produced in arc systems at the same crustal depth. X-axis is truncated at 250, full plots in Supplementary Information.

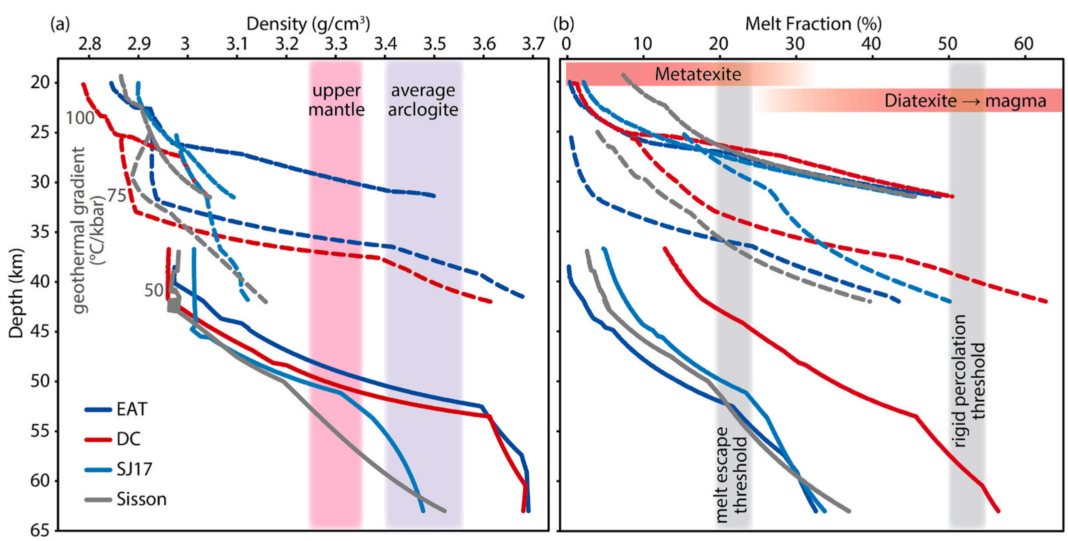

Fig. 5. Variation in (a) bulk-rock density and (b) calculated melt fraction as a function of depth along geotherms of 50, 75, and 100 °C/kbar for a medium-K, high-alumina Cascades basalt 87S35A from Sisson et al. (2005) (Sisson), 'deep crust' (DC) from Sammon and McDonough (2021), the average arc basalt from Schmidt and Jagoutz (2017) (SJ17), and the enriched Archean tholeiite (EAT) composition from Condie (1981). Arclogite density range from Ducea (2002) and Ducea et al. (2021a), and rheological thresholds are after Bea et al. (2021).

that these uncertainties may underestimate the range of crustal thicknesses that could lead to such ratios. Critically, current chemical mohometry calibrations are based on modern arc settings, which focuses on melting that occurs under a 'Phanerozoic' range of geothermal gradients; however, this range of geothermal gradients and P-T-H_2O conditions is biased towards those present in modern arcs, and not representative of conditions found in other modern tectonic settings or those present earlier in Earth history. Issues with this approach were already found by Hu et al. (2017), who found different ratio vs. depth correlations in a collisional setting. Furthermore, as demonstrated by previous authors, average geothermal gradients in arc environments may have changed through Earth history (Tamblyn et al., 2022), as may the water contents (Triantafyllou et al., 2022). As such, the calibration of chemical mohometers to modern arc settings limits their use when applied to other geological settings and/or throughout other periods in Earth history.

4.3. Implications for crustal refinery via delamination

Delamination/foundering of unstable dense lower crust that arises from melt-depleted residua and cumulate phases produced during partial melting and fractional crystallisation is a widely accepted phenomenon that contributes to the creation and geochemical maturation of Earth's continental crust (e.g. Kay and Kay, 1993; Rudnick, 1995; Lee et al., 2006; Ducea et al., 2015). By inference, this process has likely occurred throughout Earth's history for as long as continental arcs have been a locus of crust formation (e.g. Polat, 2012; Gazel et al., 2015), and even perhaps in stagnant lid environments that characterised the Earth before the onset of plate tectonics (Johnson et al., 2014). The crustal foundering model requires the formation of dense restite/cumulates, which can be achieved by producing significant volumes of garnet. As noted above, garnet is commonly cited as first stabilising at depths greater than ~35 km, but as we have demonstrated with our modelling, along with others recently (Johnson et al., 2017; Kendrick and Yakymchuk, 2020; Pourteau et al., 2020; Hernández-Montenegro et al., 2021; Tamblyn et al., 2022; Triantafyllou et al., 2022; Wang et al., 2022; Yakymchuk et al., 2023), the volume of garnet created during lower crustal melting depends strongly on the geothermal gradient, hydration state and bulk composition of the source lithology.

To assess the role and likelihood of crustal foundering, we converted mineral proportions calculated in our models to bulk-rock densities of the restite using typical individual mineral densities (Fig. 6). We consider the upper mantle to have an average density of ~3.3 g/cm^3. Lower crustal cumulates ('arclogites'; see Ducea et al., 2021a, 2021b) have densities ranging from 3.2 to 3.6 g/cm^3 (Ducea, 2002; Ducea et al., 2021a), and averaging 3.4 to 3.55 g/cm^3 (Bowman et al., 2021). Lower crust is assumed to be unstable when it has a notably higher density than the underlying upper mantle (Bowman et al., 2021), although there is no formal definition of the minimum difference that will allow dripping or delamination. Nonetheless, our results show that formation of unstable dense residues is highly dependent on the source composition that undergoes partial melting. At the coolest geothermal gradient (50 °C/kbar), all four compositions produce dense garnet-bearing residues with melt fractions >20%. These geothermal gradients are equivalent to melting at depths >50 km, i.e. in substantially thickened crust. At the intermediate geothermal gradient (75 °C/kbar), only two of the four compositions produce dense residues (EAT and DC). Finally, at the highest gradient of 100 °C/kbar, only one composition produces a dense residue (EAT), and this requires high degrees of melting to do so.

Our results corroborate previous hypotheses that the formation of dense residues is limited to thick continental arcs with cooler geothermal gradients (Ducea et al., 2015). Interestingly, changing thermal conditions related to secular cooling of the mantle infer lower geotherms in Proterozoic and Phanerozoic arc systems than Archean arc systems, while the changing bulk compositions of basalts through time also promotes foundering in ancient arcs. Although the results of modelling should not indiscriminately be applied directly to nature without careful discussion of their limitations, we argue that substantially over-thickened arcs >45 km thickness can readily produce dense restites, but that moderately thickened arcs >35 km will have a strong dependence on the compositions and conditions. For direct comparison with natural examples, the Kohistan Arc in north Pakistan is considered

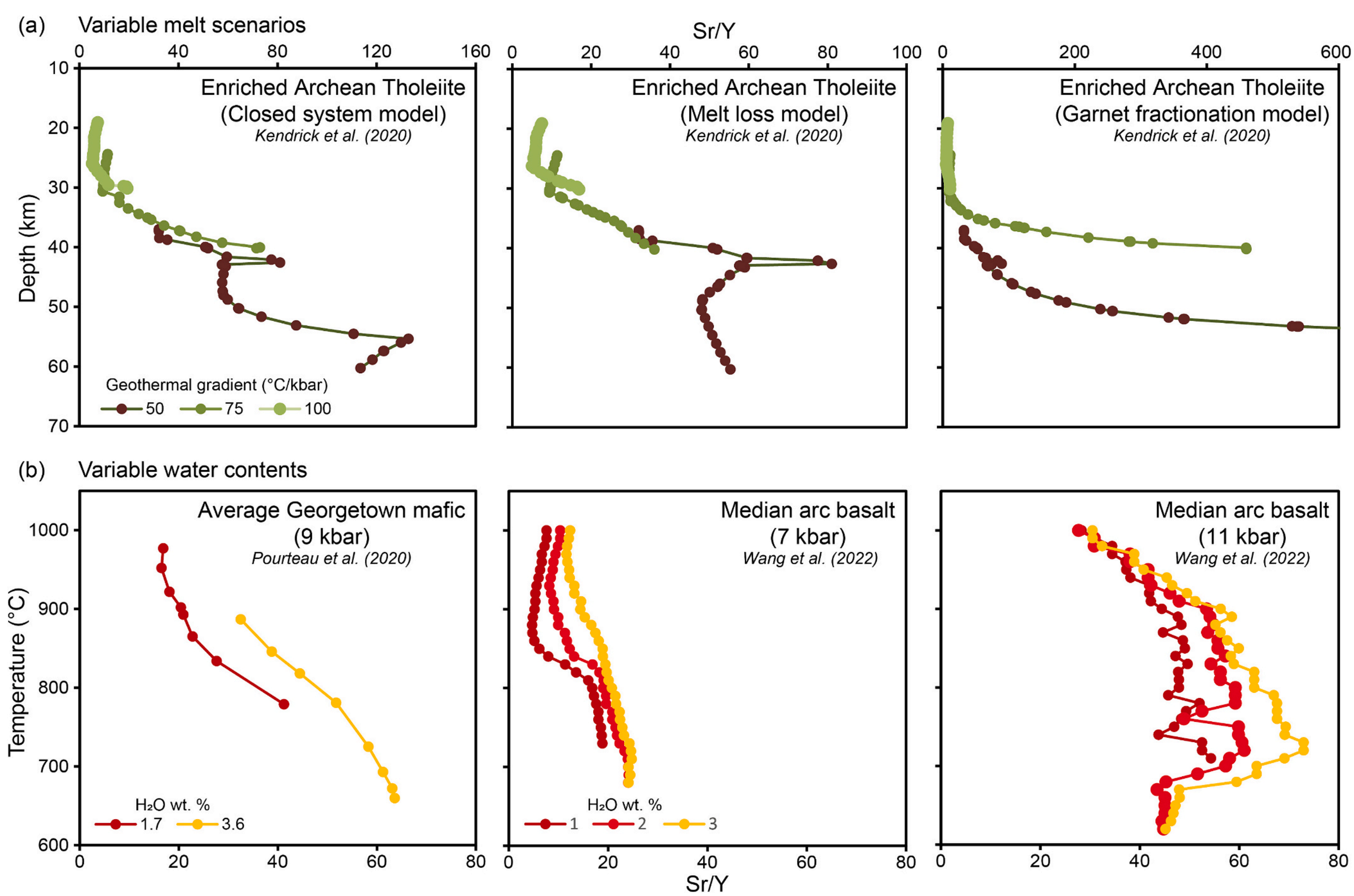

Fig. 6. (a) Plots of calculated Sr/Y ratios of melts generated from an Enriched Archean Tholeiite along 50, 75, and 100 °C/kbar geotherms, shown as a function of depth below the Earth's surface; based on the model results of Kendrick and Yakymchuk (2020). The results are based on three scenarios, closed system behaviour (similarly to Fig. 4), open system behaviour with loss of the partial melt, and open system behaviour with loss of elements into garnet porphyroblasts (details provided in Kendrick and Yakymchuk, 2020). (b) Plots of calculated Sr/Y ratios of melts generated at fixed pressure but with varying water content, shown as a function of the model temperature at melt extraction; based on the model results of Pourteau et al. (2020) using the Average Georgetown mafic protolith, and Wang et al. (2022) using an estimate Median arc basalt from a global compilation.

an exhumed island arc section, but has a reconstructed Moho depth of 50 km, with dense material occurring at depths >40 km (Jagoutz and Schmidt, 2013). Our results build upon the work of Bowman et al. (2021) who demonstrated that: 1) incorporation of melt from sedimentary protoliths will lead to stable restites (i.e. low those of low density); and 2) any melt included in the restite will lower the density and therefore also increase stability. In our modelling of restite density, we assume that the melt is extracted efficiently, leaving behind a pure residual mineral assemblage.

4.4. Crustal thickness during Earth's Middle Age

Secular trends in whole-rock geochemistry show temporal changes in the composition of granitic rocks through Earth history (e.g. Keller and Schoene, 2012; Condie et al., 2023). These changes include variations in the range of Sr/Y and La/Yb ratios (Tamblyn et al., 2022; Roberts et al., 2023). The origin of variable Sr/Y ratios for Archean rocks (TTGs) has been widely discussed (e.g. Moyen and Martin, 2012; Huang et al., 2020), with a traditional interpretation being a simple correlation of Sr/Y with pressure of magmatic differentiation (Moyen, 2011). Similar concepts have equally been applied to modern adakites (Condie, 2005; Moyen, 2009). More recent interpretations have moved away from focusing on the importance of pressure when interpreting the geochemistry of TTGs, with models invoking enriched source materials (Smithies et al., 2019), crystal fractionation and accumulation (Laurent et al., 2020; Kendrick et al., 2022), and water-fluxed melting (Pourteau et al., 2020) having been proposed instead. Tamblyn et al. (2022) identified low Sr/Y and La/Yb ratios in the mid-Proterozoic (ca. 1.85–0.85 Ga) and linked them to warmer geothermal gradients in the mantle (Brown and Johnson, 2018). Roberts et al. (2023) additionally argued that such conditions were related to granite formation in hot, wide back-arcs that would have been prevalent during the mid-Proterozoic.

Several studies have attempted to estimate average continental crustal thicknesses from the Archean to the present. Using correlated Rb/Sr ratios with total SiO_2 content, Dhuime et al. (2015) argued that the thickness of newly formed juvenile crust peaked in the Late Proterozoic, then decreased to modern-day values. Latterly, Keller and Harrison (2020) demonstrated that secular changes in the degrees of mantle melting through time can adequately explain these ratios, thus questioning the crustal thickness trend of Dhuime et al. (2015). Tang et al. (2021a) argued that Eu/Eu* ratios in zircon can be calibrated to depth, using the La/Yb host rock values and the La/Yb versus depth calibration of Profeta et al. (2015). Using this proxy, Tang et al. (2021b) subsequently arguing for a decrease in crustal thickness during the mid-Proterozoic, with a minima at ca. 1 Ga, in direct contrast to the model of Dhuime et al. (2015). As we highlight above, applying geochemical crustal thickness proxies calibrated using modern tectonic settings to Precambrian rocks or minerals carries additional uncertainties, such as global changes in geothermal gradient, crustal composition and/or hydration state (Tamblyn et al., 2022; Triantafyllou et al., 2022; Yakymchuk et al., 2023).

The deficiencies of simple whole-rock calibrations are exemplified when they are applied to large datasets of global geochemistry. Using the database of Roberts et al. (2023), we calculated the crustal thickness from ca. 4 Ga to present (Fig. 2) using Sr/Y and $(La/Yb)_n$ ratios and the calibrations of Sundell et al. (2021). Based on the Sr/Y calibration, many data points have negative crustal thicknesses in the mid-Proterozoic, indicating a severe limitation of these proxies. This result begs the question: what was the likely crustal thickness during the mid-Proterozoic and why does this calibration fail to provide sensible values? It remains a well-understood fact that direct or indirect means of establishing past crustal thickness are tricky. Although thicknesses of Proterozoic crust can be gleaned from seismically determined thickness estimates of Proterozoic terranes (e.g. Mooney et al., 1998), these crustal columns may grow or shrink due to erosion, sedimentation and/or magmatic underplating. Independent means can include P-T estimates of equilibration of metamorphic or magmatic rocks. One such constraint is the thermobarometric estimates of anorthosite generation. Massif anorthosite provinces are most prevalent during the mid-Proterozoic, and are thought to form at the Moho or within the very shallow levels of the lower crust. Thermobarometry performed on such units has indicated a somewhat consistent range of depths of formation between 35 and 40 km (Ashwal and Bybee, 2017). Although this is only one line of evidence, it implies that Proterozoic crustal thicknesses may have been similar to the average modern-day continental crust. For comparison, the mean Sr/Y of granitoids formed during the Mesoproterozoic is 12.6 (± 35.1 2SD; see Fig. 2), equivalent to a Moho depth of 25.7 ± 10.8 km using the calibration of Sundell et al. (2021).

These discussions show that the thickness of the mid-Proterozoic continental crust remains debated (see also Roberts et al., 2024), and we argue that methods calibrated specifically for modern day crust generation should be applied with caution to older geological terranes. Low Sr/Y and La/Yb ratios prevalent through this period may correlate with higher geothermal gradients (Tamblyn et al., 2022) and/or lower water contents during arc magma formation (Triantafyllou et al., 2022), but importantly, may comprise magma formation in crust of similar average crustal thickness to that of today (30–40 km). This provides an interesting question for further investigation: is over-thickening of crust (> ~ 50 km) within continental arcs limited to Phanerozoic Cordilleran-style arcs? Based on published numerical models of convergent margins, Roberts et al. (2023) argued that mid-Proterozoic continental arcs formed under elevated ambient mantle temperatures would indeed be thinner than Phanerozoic equivalents.

4.5. Crust production and the efficacy of foundering through time

As mentioned previously, we postulate that intracrustal magmatism and differentiation in mid-Proterozoic arcs dominantly occurred in continental crustal columns of 'normal' thickness (i.e. 30 to 40 km), as indicated by low average Sr/Y and La/Yb ratios. A corollary of this notion is that the generation of new felsic melts presumably occurred at pressures below those required for garnet to stabilise within the protolith. As such, the formation of dense residues within the lower crust during partial melting was likely to be significantly hampered. The foundering of dense crustal roots is one of the paradigms of Earth's continental crust formation that is built upon observations of recent to modern-day environments. Since this paradigm was established, it has been built into models of coupled crust-mantle evolution, for example, mass balance modelling of recycling into the mantle (Jagoutz and Schmidt, 2013; Lee and Anderson, 2015; Tilhac et al., 2022). We argue that the paradigm of lower crustal foundering may not be applicable throughout Earth history, not least since the onset of plate tectonics when convergent margins likely became the major locus of continental crust formation, probably since at least 2 Ga (Condie, 2021; Cawood et al., 2022). Therefore, future models of both crust formation and mantle evolution must consider the potential for secular changes in arc magmatism that may result from variable geothermal gradients, water contents and source compositions (i.e. Keller and Schoene, 2018; Triantafyllou et al., 2022). This is particularly relevant, given the prevalence of numerical modelling studies that provide insight into the geodynamics of the early Earth and the mechanisms of crustal growth and evolution. A key parameter that controls whether subduction and/or extensive arc systems may develop is mantle potential temperature, although this remains a debated variable of the Archean (and Proterozoic) Earth, with recent studies suggesting that ambient Archean mantle was likely ~100–200 °C cooler than previously suggested (cf. Ganne and Feng, 2017). Continued work is needed to place tighter constraints on such geophysical parameters that may be fed into geochemical and petrological models to produce more realistic simulations of the behaviour of the Precambrian continental lithosphere.

5. Conclusions

Trace element characteristics of igneous rocks are commonly used to interpret the petrological characteristics of the anatectic source region, including its depth within the crust. Two ratios commonly applied as barometers or 'chemical mohometers' are whole-rock Sr/Y and La/Yb, which themselves have been used to calibrate mineral-scale barometers, i.e. Eu anomalies in zircon. We have performed petrological modelling of the relative stability of major minerals that stabilise in mafic lithologies of varying composition and hydration state, which indicates that nuance is needed when using measured values from derivative melts to estimate the paleo-depth of formation. In particular, we show that compositional variability of the protolith has a non-negligible effect on major mineral stability during lower crustal melting, specifically garnet and plagioclase, which can lead to partial melts acquiring a range of trace element characteristics, even when produced at the same pressure.

These results, which build upon the work of others, imply that regional and/or secular changes in granitoid trace element ratios do not necessarily indicate a dramatically thinner crust in Proterozoic orogens; they may equally reflect varying water content in the lower crust (i.e. an increase or reduction in the flux of volatiles from slab to arc), changes in ambient geothermal gradient (i.e. isobaric temperature changes resulting from magma flux or asthenospheric/radiogenic heating), or secular variation in arc crust composition through time. Whilst such chemical mohometers have much use for the interpretation of crust formation and reworking processes on the Precambrian Earth, we urge caution in their application without consideration of all relevant sources of uncertainty.

Finally, we argue that the low Sr/Y and La/Yb ratios that typify the mid-Proterozoic correlate to garnet-free residua during the formation of felsic magmas, and thus point to crust formation without lower crustal foundering. This commonly invoked paradigm for formation of the continental crust may only be applicable to certain periods of Earth history, which has implications for mass balance of the crust-mantle system through time.

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CRediT authorship contribution statement

Nick M.W. Roberts: Writing – review & editing, Writing – original draft, Visualization, Investigation, Formal analysis, Conceptualization. **Juan David Hernández-Montenegro:** Writing – review & editing, Visualization, Methodology, Investigation, Formal analysis. **Richard M. Palin:** Writing – review & editing, Writing – original draft, Visualization, Methodology, Investigation, Formal analysis.

Declaration of competing interest

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

Data availability

Modelling software, thermodynamic database, and activity–composition relations used for phase equilibrium calculations can be downloaded from http://www.rocks.uni-kiel.de/theriakd/html/down_en.html

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Appendix A. Supplementary data

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