

A mantle-source solution to the enigma of bimodal arc volcanism

Nathan Daczko (✉ nathan.daczko@mq.edu.au)

Macquarie University <https://orcid.org/0000-0002-3737-3818>

Timothy Chapman

University of New England <https://orcid.org/0000-0002-4821-6420>

Michael Foerster

Macquarie University <https://orcid.org/0000-0002-0778-5811>

Heather Handley

Macquarie University

Stephen Foley

Macquarie University <https://orcid.org/0000-0001-7510-0223>

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Abstract

Silicate melts in arc environments are dominated by mafic (low-silica) and silicic (high-silica) compositions, often generating a characteristic bimodal pattern. We investigate the whole arc crust and show that the plutonic lower crust shares the bimodal pattern of melts from volcanoes. This key observation reveals that, contrary to some explanations of bimodal volcanism, variation in mantle source and mantle processes must fundamentally control bimodalism. We also recognise bimodalism in Th/La composition of the whole arc crust and suggest a new working hypothesis: bimodalism originates by melting of distinct sub-arc mantle sources, one dominated by relatively dry peridotite and the other by hydrous pyroxenite. The two groups of primary melts fractionate along distinct liquid lines of descent that lead to relatively dry mafic melts ($\text{Th/La} \sim 0.1$) versus hydrous silicic melts ($\text{Th/La} > 0.2$) by 65–80% fractional crystallisation. Common crustal processes such as crystal fractionation, assimilation, reactive flow and/or magma mixing may also lead to differentiation of both groups.

Full Text

Subduction zones are the most tectonically active regions on Earth. They produce more variable volcanic rock compositions than any other tectonic environment and are crucial to understanding many fundamental geological processes, such as continental growth, geochemical cycles, tectonic-climate coupling, ore genesis, earthquakes, volcanism, and other natural hazards¹. A basic characteristic of melts in arc environments is that they are bimodal², meaning that mafic and silicic compositions are common, whereas intermediate melts are relatively rare. Although this bimodality has been recognised and debated widely for nearly 100 years, it is yet to be convincingly explained³. Here, we use whole-rock and isotope compositions of plutonic rocks in deep crustal arc sections to develop a new hypothesis for bimodality in arcs. We show that the bimodal pattern extends throughout the entire arc crust from volcanoes to the lower crust, indicating that the ultimate cause of bimodality cannot be a crustal phenomenon. Instead, we propose that bimodalism originates by melting of distinct sub-arc mantle sources dominated by relatively dry peridotite or hydrous pyroxenite. Crustal processes that were previously invoked to explain bimodality in igneous systems also occur, but their influence is limited to secondary modification of bimodal melts that originate in the mantle.

A long-recognised pattern of bimodal distribution in the composition of igneous rocks, known as the Bunsen-Daly or Daly gap, has puzzled geologists for over 100 years, producing many extant and contradictory models for its origin that fuel ongoing debate²⁻⁴. Suggested causal processes include liquid immiscibility, stalling of intermediate compositions, crystallization and/or separation of particular minerals, and partial melting of the crust⁵⁻⁷, but consensus is lacking.

In subduction zone settings, mafic and silicic melt inclusions dominate, as opposed to whole rock compositions where intermediate rocks are very common² (Fig.1); the whole rocks comprise mostly crystal-rich units with up to 90% of minerals in disequilibrium with their bulk-composition⁸. Recently,

Meade et al.³ returned to crustal assimilation as a major factor in the explanation of bimodality in continental volcanism. However, Keller et al.⁹ argued for progressive fractional crystallization during magma ascent to produce intermediate and silicic magmas, emphasizing the fractionation of mafic cumulates. Similarly, numerical considerations of the thermal effects of mafic intrusions support melting of the crust or, at smaller scales, reactive flow through crystal mushes to create bimodal suites containing low degrees of magma mixing¹⁰⁻¹³. The breadth of these studies shows that the bimodal geochemical pattern is well documented, but its origin may be multifaceted.

The mechanisms that have been proposed to create the bimodal geochemical pattern are complicated and invariably assigned to the crustal component of the arc system (e.g., deep hot zone, MASH [melting, assimilation, storage, homogenization] zone, shallow hot zone, cold storage and remobilisation^{11,13,14-15}). Most of these models are based on inferred magma chamber or crystal mush processes occurring dominantly in the upper crust⁴ (<15 km) but some studies suggest that the fundamental bimodal character had already been generated below this level². An additional limitation to these models is the common assumption of a relatively homogeneous peridotite mantle source for the initial arc magmas. However, the first stages of mantle melting almost universally occur in a mixed source region consisting of a minimum of two distinct rock types¹⁶. In the case of the mantle wedge above subducting slabs, these comprise a mixture of peridotite and metasomatised components involving serpentinite through to hydrous pyroxenite^{17,1} with increasing temperature deeper into the mantle wedge above the subducting slab (Fig.1).

The fundamental bimodal character of volcanism in arcs² is investigated here by using a new compilation of whole-rock geochemical data from eroded Mesozoic arcs: Fiordland (New Zealand), Kohistan (Pakistan) and Talkeetna (Alaska, USA), which are the Earth's premier deep arc sections (Fig.1). This compilation presents consistent peaks in composition at 50–54 wt.% and 70–75 wt.% SiO₂ (Fig.1; complete references and compiled data in Supplementary Table 1). The pattern is the same as that identified in melt inclusions from volcanic rocks (red line in Fig.1; ref.2), demonstrating that the bimodal character applies to the entire arc crust from volcanoes down to the lower crust. Thus, a deeper origin within the mantle is implicated as the ultimate source of the bimodal geochemical pattern, discounting all explanations that rely on crustal processes alone.

The formation of hydrous pyroxenite by interaction between slab-sediment-derived partial melt and peridotite in the mantle wedge above a subducting plate is predicted by experiments from temperatures as low as 675°C^{17,18}. Partial melting experiments on dry pyroxenites show that they may produce primary melts that overlap or are up to 5–15 wt.% higher in SiO₂ than melts of peridotite¹⁹ (B and A, respectively on Fig.1). However, experiments show both source types are capable of forming a range of primary melt compositions. While current models focus on flux melting of peridotite hydrated by water released from the subducting slab (A, Fig.1), our hypothesis also invokes flux melting of hydrous pyroxenite (B, Fig.1), as return flow in the mantle wedge may draw pyroxenite to depths and temperatures where it will partially melt²⁰. Large areas of hydrous pyroxenite occur in arc-related ultramafic massifs and are often thought

to originate as cumulates²¹; these may become involved in melting above the subducting slab. Phlogopite pyroxenite rocks are expected to be common products of melt-rock reaction in the mantle wedge^{17,18}. However, most experiments on pyroxenite have been aimed at the deep recycling of dry rocks in the oceanic environment^{19,22}, and so may be of limited relevance to the hydrous conditions above subducting slabs. Melting points of hydrous pyroxenites^{23–24} can be 150–200°C lower than dry pyroxenites²⁵ at 30–50 km depth, depending on the mineral assemblages present. This highlights a need for future focussed experiments to establish the full range of melt compositions formed beneath arcs.

Regardless of the possibility that primary melts sourced from hydrous pyroxenite may begin with higher SiO₂ (up to 55 wt.% SiO₂), experiments show that the water content of mafic melts is a critical determinant of the degree to which SiO₂ increases in the fractionated magma^{26–28}. At 80% fractional crystallisation, dry mafic magmas evolve to ~56 wt.% SiO₂, whereas more hydrous magmas evolve along much longer trajectories to ~77 wt.% SiO₂^{27,28} (green and blue vertical dashed lines on Fig. 2). The short, dry liquid line of descent (green arrow, Fig.2) forms fractionated magmas with low-SiO₂ and high MgO, FeO and CaO contents. In contrast, the long, wet liquid line of descent (blue arrow, Fig.2) forms fractionated magmas with high-SiO₂, but low MgO, FeO and CaO.

Our hypothesis of a fundamentally bimodal sub-arc mantle is supported by bimodal Th/La in arc crust, approximately distinguished by Th/La=0.2 (Fig.3). Mafic plutons and melt inclusions share the low Th/La (Fig.3) characteristic of mid-ocean ridge basalt and most peridotite xenoliths (green star; Supplementary Table 2); the mafic compositions form an array at nearly constant Th/La~0.1 (Fig.3, dashed black arrow). Pyroxenite xenoliths cluster at either high Th/La (~0.3; Fig.3, left blue star) or at similar Th/La to peridotite (Fig.3, right blue star). Both Th and La are enriched in subducted sediments and the enrichment is imparted to metasomatised sub-arc mantle components (hydrous pyroxenite) compared to peridotite²⁹. The felsic plutons and melt inclusions form steep arrays on Figure 3, increasing in Th/La (Fig.3, black arrows). We require new experiments to better understand partitioning of Th and La during flux melting of peridotite and hydrous pyroxenite, and how these elements behave during magmatic fractionation. These details will help explain the bimodalism of Th/La in arc crust (Fig.3).

The observation of bimodal compositions in both surface volcanoes and the plutonic base of arcs (Fig.1) indicates that significant magmatic evolution occurs by melt-rock reaction during transport through the sub-arc mantle (arrows A to C or B to D, Fig.1). Additionally, some evolution may occur in the lower to middle crust via combinations of crystal fractionation, assimilation, reactive flow and/or magma mixing (C to E and D to F, Fig.1; ^{13, 30–31}).

In our case study of the lower and middle crust of Fiordland, New Zealand, the mafic and silicic plutonic components are spatially distributed into paired inboard (Western Fiordland Orthogneiss, WFO, dry, mafic, rear arc) and outboard (Separation Point Suite, SPS, wet, silicic, front arc) belts (Fig.2). Our new and compiled published isotopic data (Supplementary Table 2) shows that the paired plutonic belts in Fiordland have similar mantle-like Sr-Nd isotopic compositions to each other (Fig.4), precluding

significant ancient crustal contributions in the petrogenesis of the silicic suite^{3,15} and relating the two mantle sources. The distinction in major element compositions (Fig.2) and the simple spatial pattern in Fiordland likely reflects well-separated relatively dry peridotite- and hydrous pyroxenite-dominated sub-arc mantle sources (Fig.1). These relationships are difficult to explain with the other proposed causes of bimodality^{2-8,10-15}, though additional isotopic systems (e.g., Pb) could be analysed to investigate this further. An equally heterogeneous sub-arc involving two mantle sources is inferred for paired volcanic belts, such as NE Japan, Kamchatka, Kurile and Chile³²⁻³⁴. We envisage that the distribution and degree of mixing of the sub-arc mantle endmembers controls the presence or absence of such spatial geochemical patterns. In contrast, relatively homogeneous volcanic and plutonic crust in many arcs is produced by melting of a uniform sub-arc mantle source.

In summary, a heterogeneous distribution of variably hydrous source rocks in the sub-arc mantle imparts a fundamental bimodal geochemical signature to magmatic arcs. Relatively dry peridotite and hydrous pyroxenite endmembers form primary melts that follow very different fractionation paths. This is essential to explain the existence not only of bimodal volcanism at Earth's surface, but also of bimodal plutonism in the lower and middle crust (Fig.1). It places the solution to the enigma of arc bimodalism at sub-crustal depths within the sub-arc mantle (Fig.1) and attributes it to reactions between peridotite of the mantle wedge and fluids or melts from the subducting slab to form bimodal source rocks comprising less-reacted peridotite and metasomatised components such as hydrous pyroxenite. This working hypothesis provides a critical foundation for future research of melting processes, the causes of the bimodal composition of arc volcanism, and how and where primary melts are modified on their way to the surface.

Methods

Geochemical databases: The geochemical compilation of whole-rock data from (1) deep-arc sections for Kohistan and Talkeetna, (2) peridotite and pyroxenite xenoliths, and (3) melt inclusions compositions are from the Georoc database (<http://georoc.mpch-mainz.gwdg.de/georoc/>). The data was refined by removal of altered samples (Supplementary Table 1). The new compilation for Fiordland is based on extensive published literature across the entire Cretaceous arc sequence (Supplementary Table 1).

Whole-rock Sr-Nd isotopes: Selected samples of the Western Fiordland Orthogneiss (WFO) and Separation Point Suite (SPS) in Fiordland, New Zealand were analysed for whole-rock Rb-Sr and Sm-Nd isotopes. The samples selected include the deepest exposed portion of the Fiordland arc, the Breaksea Orthogneiss³⁵ together with mid-crustal components of the SPS in the Grebe Valley in Eastern Fiordland³⁶ (Supplementary Table 2). The samples were analysed on a Thermo Finnigan Triton Thermal Ionisation Mass Spectrometer (TIMS) housed at Macquarie GeoAnalytical, Macquarie University, Sydney. Sample powder weights of 0.1–0.15 g were digested in Teflon beakers with 2 mL concentrations of HNO₃ and HF, together with 10–15 drops of HClO₄, followed by HCl (4 ml) and H₂O₂. The samples were then dissolved in HCl and dried, before being added to an HCl and HF solution and then placed in a

centrifuge. The samples were then eluted through chromatographic columns containing a wet powder resin. A 29.5 mL solution of 2.5N HCl/0.1N HF was added before collecting Sr. The remaining Nd sample was dried before processing through a second ion exchange column containing Ln.Spec resin. The collected Sr and Nd separates were dried before being re-dissolved with 1 μ L activator solution and loaded onto Rhenium filaments. The filaments were heated to a current of 2 A for drying purposes. Sr was processed by TIMS on single filaments producing between 1.5–6 volts of signal at currents of 2700 mA. Nd was processed on double filaments producing between 2–10 volts of signal at currents of 4500 mA (ionisation) and 1200–1800 mA (evaporation). Up to 200 readings were taken in each run, together with BHVO-2 and synthetic standards. Absolute standard errors for Sr were between 3×10^{-6} and 5.3×10^{-5} and for Nd between 2×10^{-6} and 2.8×10^{-5} .

Declarations

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Author contributions

N.D, T.C, M.F., H.H and S.F initiated the project, contributed to analysis of the data and the writing of the manuscript.

Competing Interests

The authors declare no competing financial interest. N.D. was an editorial board member for Scientific Reports at the time of submission.

Materials and Correspondence

Requests should be addressed to the corresponding author: nathan.daczko@mq.edu.au.

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Figures

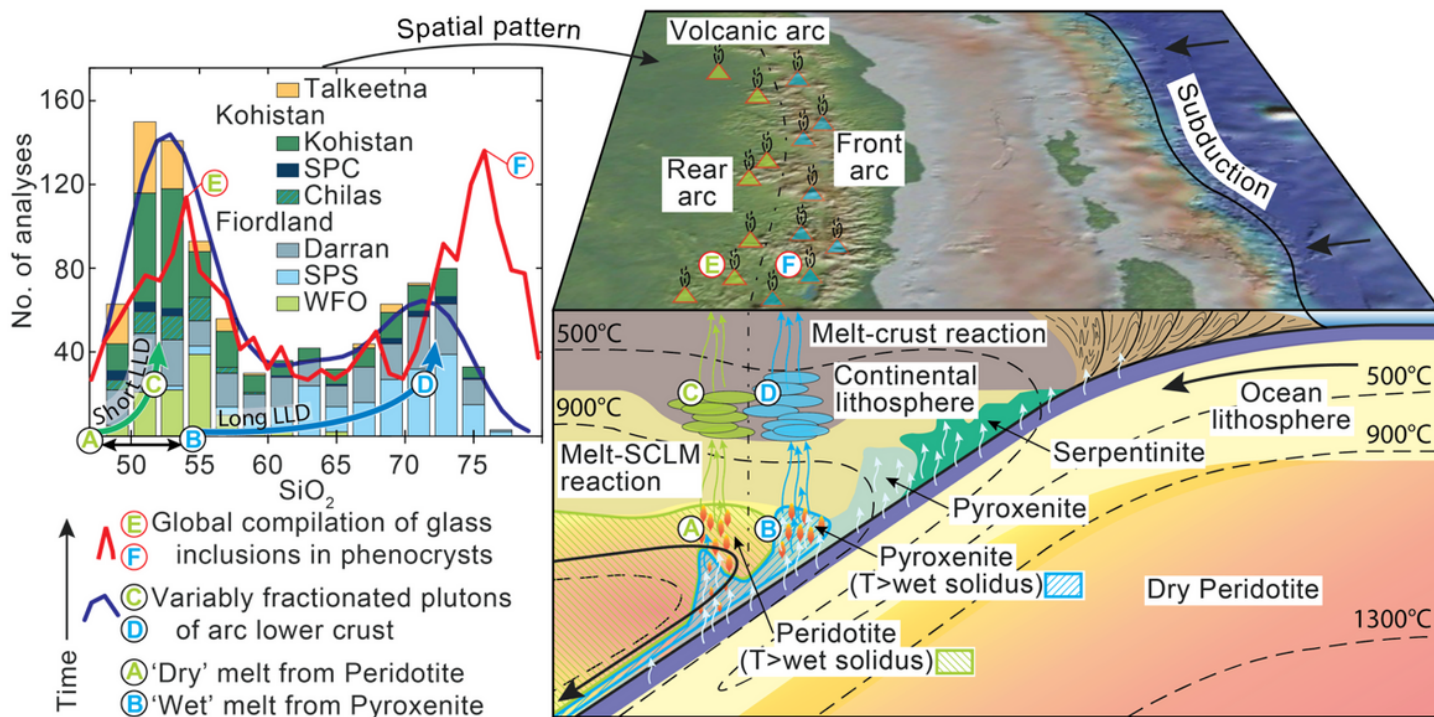


Figure 1

Three-dimensional working hypothesis of a subduction zone and volcanic arc. Bimodality originates at source in a heterogeneous mantle due to flux melting of distinct sources dominated by relatively dry peridotite (A) and metasomatised hydrous pyroxenite (B). Sources are both above the subducting oceanic lithosphere, and melts evolve to bimodal suites observed in both the lower crust (C, D) and volcanoes (E, F). Stacked histogram and probability density function of a global compilation of SiO_2 contents of rocks ($n=1,399$) from Earth's premier exposed deep arc sections -- Fiordland (New Zealand [Darran Suite, Separation Point Suite (SPS), Western Fiordland Orthogneiss (WFO)]), Kohistan (Pakistan [Kohistan

Batholith, Southern Plutonic Complex (SPC), Chilas Complex]) and Talkeetna (Alaska, USA); red probability density function is an updated global compilation of glass inclusions in phenocrysts from volcanic arcs (n=7,875), confirming the research of ref.2. Probability density functions show the bimodal pattern in both volcanic (red line) and plutonic (blue line) crust but are not shown at the same vertical scale. Sources of data in Supplementary Table 1 and GEOROC (See Methods).

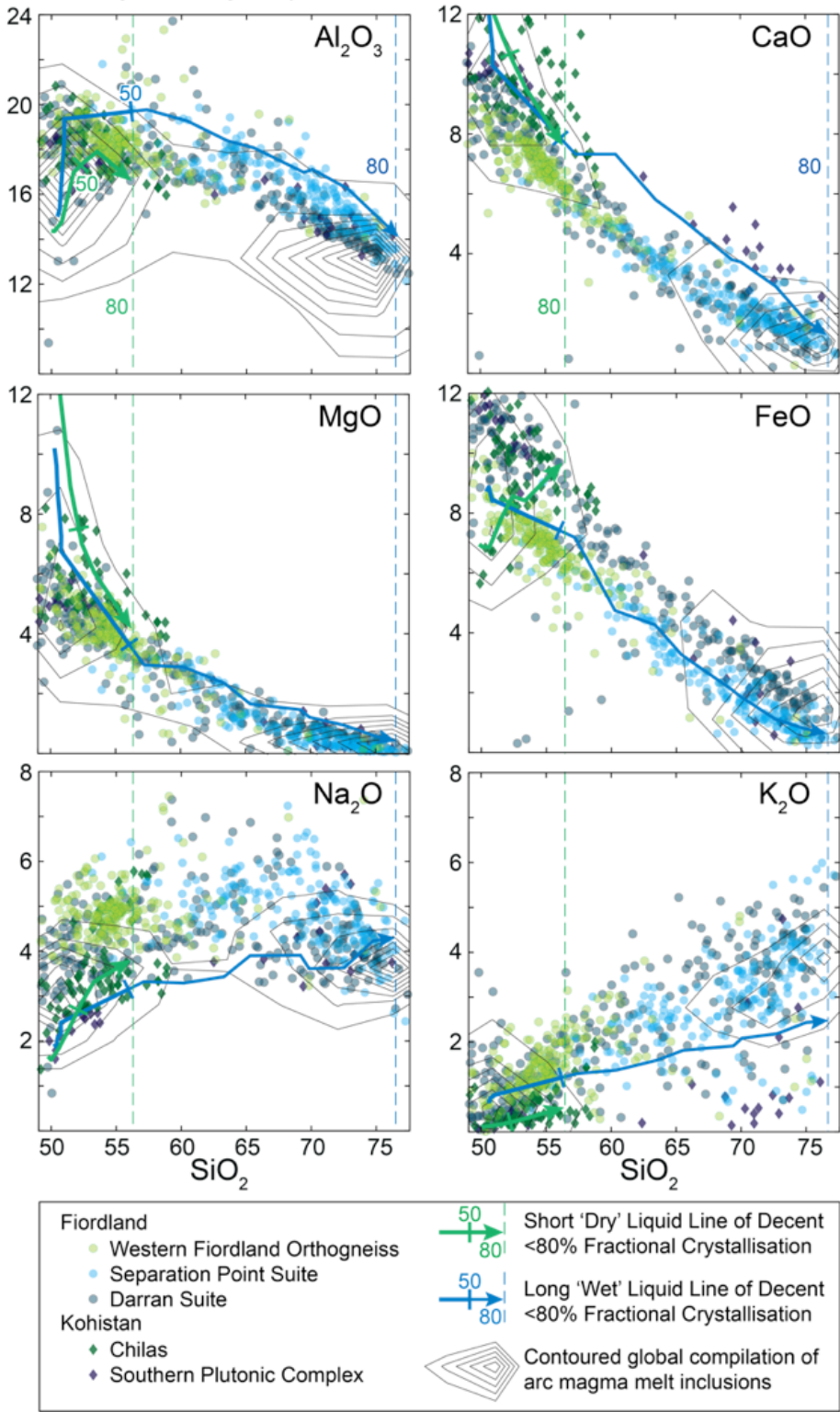
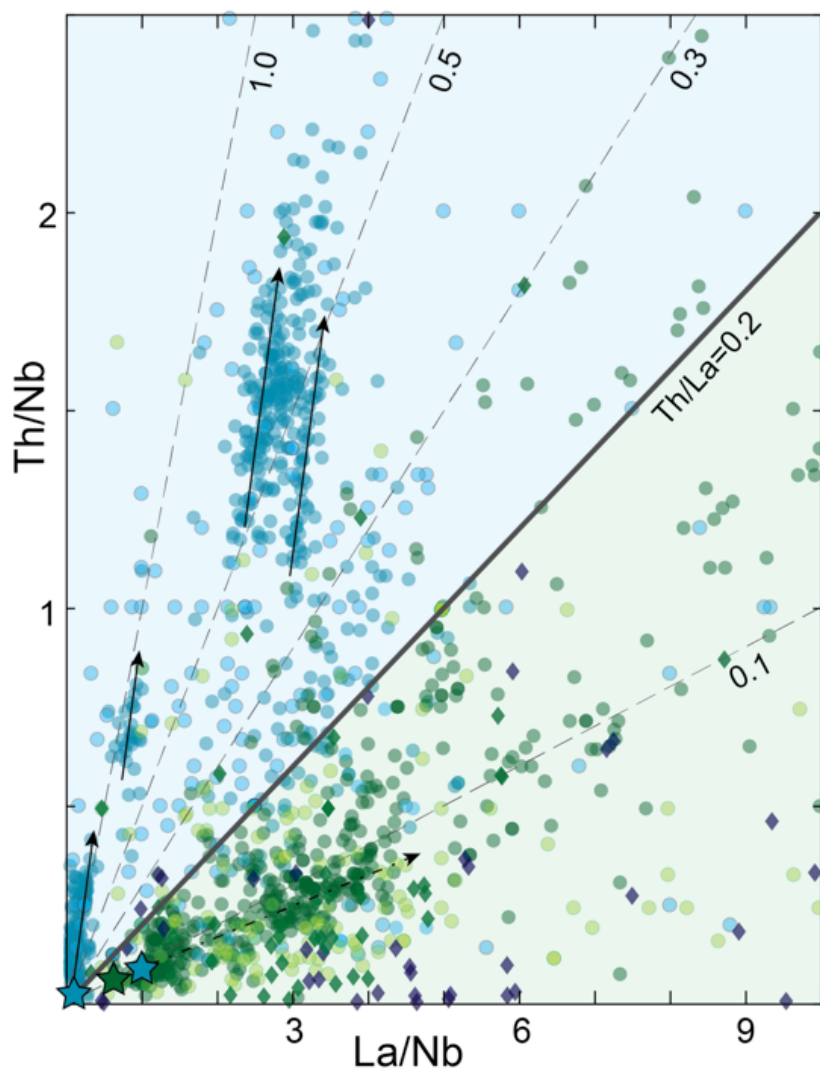


Figure 2

Whole rock major elements versus SiO₂ showing contoured global compilation of arc magma melt inclusions (Supplementary Table 1, updated from ref.2; See Methods) overlaid with fractionation patterns of bimodal arc plutonic crust for Fiordland and Kohistan (Supplementary Table 1; the Talkeetna data set is not shown as it contains only mafic samples and shares compositions overlapping the Chilas Complex, Kohistan). Liquid lines of descent are shown from relatively dry (green arrow²⁷) and hydrous (blue arrow²⁸) mafic magmas. A tick mark on the lines indicates 50% fractional crystallisation and each line ends with an arrowhead at 80% fractional crystallisation, marked by a vertical dashed line on each plot. Note that 80% fractional crystallisation of the relatively dry magma only evolves to ~56 wt.% SiO₂, whereas more hydrous magmas evolve along much longer trajectories to ~77 wt.% SiO₂^{27,28}.



- ★ peridotite xenoliths, Th/La~0.1
- ★ pyroxenite xenoliths, Th/La~0.3

High Th/La (>0.2)	
95%	● Melt inclusions, SiO ₂ > 60
74%	● Separation Point Suite
Low Th/La (<0.2)	
88%	● Melt inclusions, SiO ₂ < 60
77%	● Western Fiordland Orthogneiss
90%	◆ Chilas
87%	◆ Southern Plutonic Complex

Figure 3

Bivariate plot of Th/Nb versus La/Nb showing arc magma melt inclusions and selected deep arc plutonic rocks (Supplementary Table 1). High proportions of silicic data sets plot at Th/La>0.2 and form steep arrays (black arrows), while the majority of mafic data sets plot at Th/La<0.2 and form an array a nearly constant Th/La. High Th/La is consistent with a greater component of subducted sediment in

metasomatic mantle (e.g., hydrous pyroxenite), whereas low Th/La is typical of MORB-like peridotite sources.

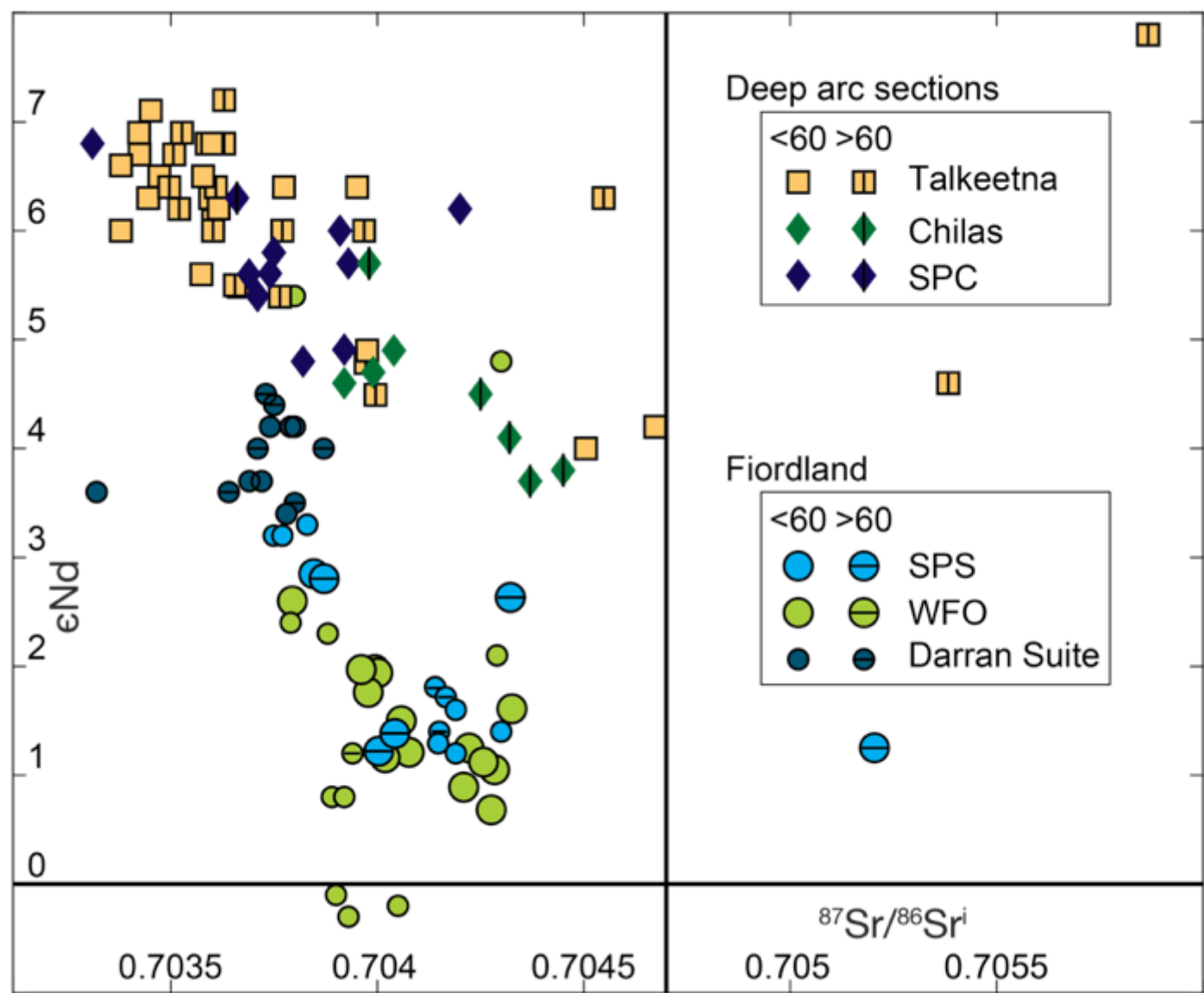


Figure 4

Epsilon Nd versus initial $^{87}\text{Sr}/^{86}\text{Sr}$ ratio at crystallisation ages for Earth's premier exposed deep arc sections – Fiordland (New Zealand), Kohistan (Pakistan) and Talkeetna (Alaska, USA). The Fiordland data includes the older long-lived Darran Suite and two Cretaceous suites comprising inboard (Western Fiordland Orthogneiss, mafic, rear arc) and outboard (Separation Point Suite, silicic, front arc) belts. Smaller circles and diamonds, this study; larger circles and other symbols are from literature (Supplementary Table 2).

Supplementary Files

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