

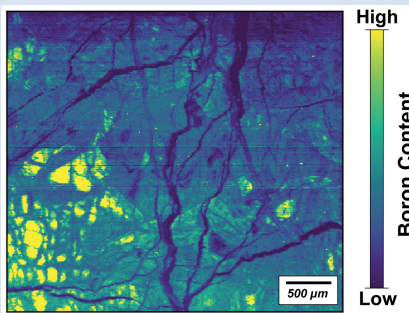
Imaging of boron in altered mantle rocks illuminates progressive serpentinisation episodes

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Abstract



Serpentinised mantle rocks reflect the cumulative sum of multiple alteration events, but to date, identifying distinct serpentinisation episodes has remained challenging due to limited knowledge of the spatial distribution of tracers of fluid-rock exchange. Here we present novel high spatial resolution (~10 μm) boron, nickel, calcium, and lithium concentration maps combined with *in situ* boron isotope analyses of strongly serpentinised mantle peridotites from the Troodos ophiolite, Cyprus. Our maps indicate strongly heterogeneous boron concentrations with high boron concentrations in early formed serpentine replacing olivine but much lower boron contents in mesh-textured serpentine and bastitic pyroxene. Late stage crosscutting serpentine veins have very low boron concentrations. In contrast, boron isotope measurements, made at coarser scales, are remarkably uniform (mean value $+11.9 \pm 3.2$ ‰, 1σ , $n = 49$). We interpret the high boron serpentine as reflecting the partial preservation of an early

pervasive serpentinisation episode by fluids with high boron concentrations sourced from the dehydration of the subducting Cyprus slab. Subsequent serpentine phases with moderate to low boron reflect progressive recrystallisation and leaching by low boron concentration meteoric waters.

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Introduction

Serpentinisation, the interaction of water with mantle rocks, is a crucial process in the Earth system, influencing the planetary water cycle. It plays a key role in plate tectonics (Guillot *et al.*, 2015), forms mountains through isostatic uplift (Evans *et al.*, 2021), and stores water, carbon, and fluid-mobile elements (Kodolányi *et al.*, 2012). Understanding where, when, and with what fluids mantle rock alteration occurs is vital for quantifying serpentinisation's role in planetary cycles. Existing evidence, such as crosscutting relationships and stable isotopic compositions, suggests a progressive series of reactions during multiple water-rock interaction episodes (Alt and Shanks, 2006). However, determining the conditions of distinct serpentinisation events is challenging due to the complex geological histories of altered mantle rocks, often overwritten by subsequent interactions (Kyser and Kerrich, 1991). Whole rock analyses reflect cumulative signatures, making geochemical and isotopic fingerprinting of discrete events difficult (Wenner and Taylor, 1973; Alt and Shanks, 2006) due to poorly constrained spatial and temporal distributions of elemental and isotopic changes resulting from serpentinisation events.

Boron, an abundant element in serpentine with concentrations reaching ~250 μg/g (Pabst *et al.*, 2011), far exceeds levels in the primitive mantle (<0.25 μg/g; Marschall *et al.*, 2017). This discrepancy makes boron and its isotopes valuable for discerning

serpentinisation processes and conditions (Boschi *et al.*, 2008; Vils *et al.* 2009; Martin *et al.*, 2016). Previous studies noted variations in boron concentrations and isotopic compositions, yet the lack of spatial context raises uncertainty about whether these variations indicate distinct serpentinisation events or the accumulation of successive episodes.

Our investigation focuses on the Troodos ophiolite's serpentinised mantle rocks, utilising high resolution (10 μm/pixel) 2D maps of boron content, calibrated against *in situ* and powder measurements. In conjunction with *in situ* boron isotope analyses, our results reveal contrasting boron signatures in the strongly altered Troodos mantle peridotites. This allows us to identify distinct serpentinisation events by contrasting fluid sources.

Geological Setting

The Troodos Massif in Cyprus houses an exceptionally well preserved ophiolite sequence, featuring an elliptical bullseye-patterned welt at its centre (Fig. 1a). This welt, with the highest elevations composed of mantle peridotites, is part of the Troodos Mantle Sequence, which is divided into two regions: the Olympus and Artemis domains (Wilson, 1959) that are interpreted as nested serpentinite diapirs with contrasting serpentinisation and deformation styles and intensities (Evans *et al.*, 2021).

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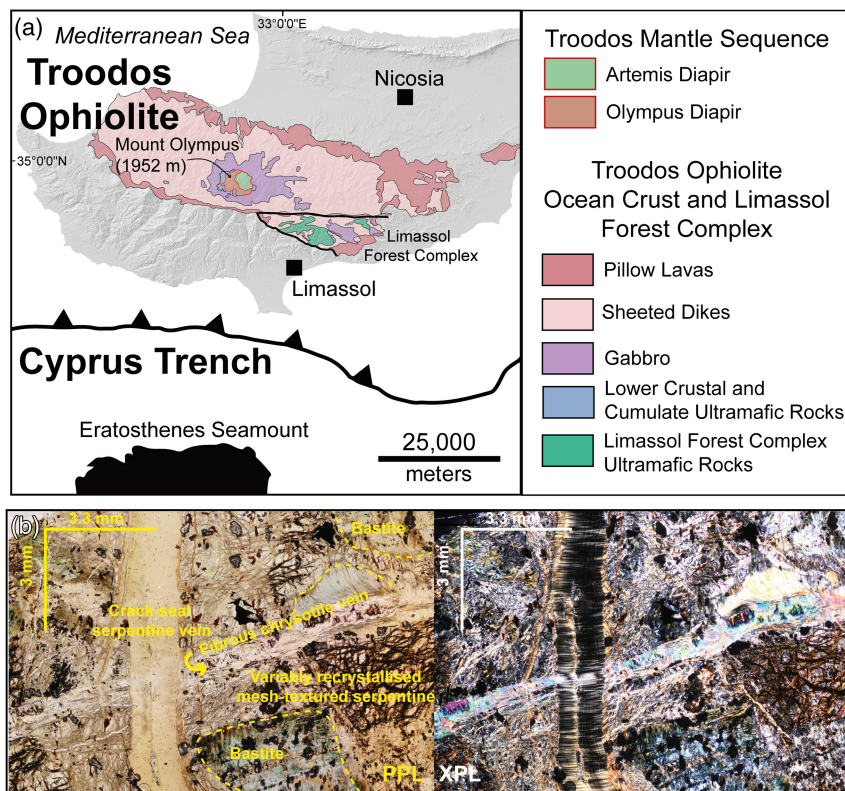


Figure 1 (a) Geological map of the Troodos ophiolite with the bullseye geometry of the Mantle Sequence highlighted (adapted from [Evans et al., 2021](#)). (b) Representative photomicrograph of completely serpentinised mantle peridotite located within the Artemis Diapir with variably recrystallised serpentine mesh texture, bastite, crack-seal serpentine vein and fibrous chrysotile vein. Scale bar is proportional to the mapped region of [Figure 2](#).

The Olympus Diapir consists of partially serpentinised tectonised harzburgites, while the Artemis Diapir is a sub-circular region with completely serpentinised peridotite blocks and clasts in a serpentinite breccia matrix ([Wilson, 1959](#); [Evans et al., 2021](#)). The Artemis rocks display variably recrystallised mesh-textured serpentine assemblages with distinctive features, including mutually crosscutting, fibrous asbestiform chrysotile and crack-seal serpentine vein sets ([Fig. 1b](#)). These serpentinite diapirs are proposed to originate from the tectonic juxtaposition of geochemically distinct mantle regions through serpentinite diapirism ([Batanova and Sobolev, 2000](#)).

The Troodos ophiolite originated at a Neo-Tethyan spreading ridge around 90–92 million years ago above a supra-subduction zone ([Moore et al., 1984](#)). However, its recent uplift and exposure, approximately 5.5 million years ago, is attributed to concentrated serpentinisation of the mantle wedge above a new north-dipping subduction zone. This zone consumes old oceanic crust, at least Mesozoic in age, on the leading edge of the Sinai plate (Cyprus slab) in the Cyprean trench to the south of the island. The initiation of subduction occurred in the early Miocene (~20 million years ago) ([Robertson, 1998](#); [Feld et al., 2017](#)). A significant negative Bouguer anomaly is centred on the Artemis Diapir, modelled as a vertical, cylindrical body of low density (~2,700 kg/m³) strongly serpentinised peridotite extending to a depth of approximately 11 km. This depth aligns with the inferred minimum depth of the top of the downgoing plate ([Gass and Masson-Smith, 1963](#); [Feld et al., 2017](#)). It's important to note that modern subduction differs from the supra-subduction zone setting that led to the formation of the Troodos ophiolite ([Moore et al., 1984](#); [Robertson, 1998](#)). Subduction beneath Cyprus stalled due to the collision of the

Eratosthenes plateau with the Cyprean trench less than 5 million years ago. This collision focused fluids released by the dehydration of the downgoing Cyprus slab beneath the Mount Olympus region, inducing serpentinisation hydration reactions and isostatic uplift ([Robertson, 1998](#); [Evans et al., 2021, 2024](#)).

The Troodos mantle peridotites have been uplifted and exposed since the Pleistocene ([Poole and Robertson, 1991](#)), a process enhanced by the incursion of meteoric waters. Oxygen and hydrogen isotope analyses of serpentinised Troodos rocks have yielded various interpretations, including exchange with Cretaceous seawater at the spreading ridge, exchange at 200 to 300 °C with waters sourced from the Cyprus slab, or exchange at less than 50 °C with meteoric water ([Magaritz and Taylor, 1974](#); [Sheppard, 1980](#); [Nuriel et al., 2009](#); [Evans et al., 2021, 2024](#)).

Analytical Methods

Boron concentrations were mapped in polished serpentinite thick sections using a 10 × 10 μm ablation beam from an Elemental Scientific Lasers NWR193 Excimer laser ablation (LA) system with a TwoVol2 ablation cell coupled to an Agilent 8900 Triple Quadrupole ICP-MS. Boron concentrations were calibrated against *in situ* and pressed powder pellet (PPP) boron concentrations determined at coarser resolutions (150 × 50 μm). Boron isotope analyses of polished thick sections and pressed powder pellets (PPP) were acquired using a Thermo Scientific Neptune Plus multi-collector inductively coupled plasma (MC-ICP) mass spectrometer. For more details on our analytical procedures refer to the [Supplementary Information](#).

Results

The *in situ* boron elemental 2D map of a fully serpentinised sample from the Artemis Diapir (Fig. 2a) indicates three distinct serpentine styles with varying boron concentrations: 1) high boron concentration serpentine, pseudomorphing original olivine grains, 2) moderate boron serpentine, coexisting with the B-rich style, and 3) discrete, irregular low boron concentration serpentine veins and mesh-textured background serpentine, which crosscuts types 1 and 2. The nickel content map (Fig. 2b) reflects the original primary texture before serpentinisation, with former olivine grains exhibiting high nickel contents and former pyroxene grains having relatively lower nickel contents. Calcium maps (Fig. 2c) highlight the contrast between mesh-textured serpentine and serpentine veins, with high calcium counts indicating calcium-rich inclusions. Lithium contents (Fig. 2d) show a distinct pattern from boron, with high lithium content occurring in serpentine pseudomorphing original pyroxene grains, while some late stage serpentine veins with mesh-textured serpentine generally have low lithium contents.

Samples from the Olympus and Artemis diapirs show similar boron concentrations and isotopic compositions (Fig. 3). In the Troodos Mantle Sequence, whole rock pressed powder pellet boron concentrations range widely (7 to 80 $\mu\text{g/g}$, mean $34 \pm 21 \mu\text{g/g}$, $n = 17$; Fig. 3a). *In situ* boron concentrations from polished thick sections exhibit a similar range (3 to 49 $\mu\text{g/g}$, mean $21 \pm 13 \mu\text{g/g}$, $n = 49$; Fig. 3b), with notable differences between serpentine types (Fig. 3b). Mesh-textured serpentine

has a higher mean boron concentration ($26 \pm 12 \mu\text{g/g}$, $n = 26$) compared to serpentine veins ($12 \pm 11 \mu\text{g/g}$, $n = 20$). Chrysotile veins in thick sections and pellets have boron concentrations ranging between 10 and 35 $\mu\text{g/g}$ (Fig. 3).

Boron isotopic compositions ($\delta^{11}\text{B}$) of serpentine veins and serpentinites in the Troodos Mantle Sequence range from +6.7 to +18.4 ‰ (mean $+11.9 \pm 3.2 \text{‰}$, $n = 49$; Fig. 3c). Olympus and Artemis Diapirs' serpentinites have indistinguishable boron isotopic compositions, suggesting alteration by a uniform external fluid despite their different mantle origins. The Troodos Mantle Sequence's boron isotopic compositions align with those from various serpentinisation settings, yielding a mean $\delta^{11}\text{B}$ value of $+17.3 \pm 10.2 \text{‰}$ ($n = 195$; Fig. 4). Published serpentinite boron concentrations vary widely (mean $\delta^{11}\text{B}$ of $35 \pm 32 \mu\text{g/g}$, $n = 195$; Fig. 4b).

Discussion

High resolution (10 μm) boron elemental mapping of serpentinised mantle rocks highlights a highly heterogeneous spatial distribution of boron (Fig. 2). We interpret the three texturally distinct serpentines resulting from at least two temporally distinct alteration events. The initial pervasive serpentinisation by a fluid with a high boron concentration formed B-rich serpentine through the pseudomorphic replacement of olivine and serpentine vein precipitation. A second pervasive alteration event by a low boron concentration fluid is recorded by the mesh-textured

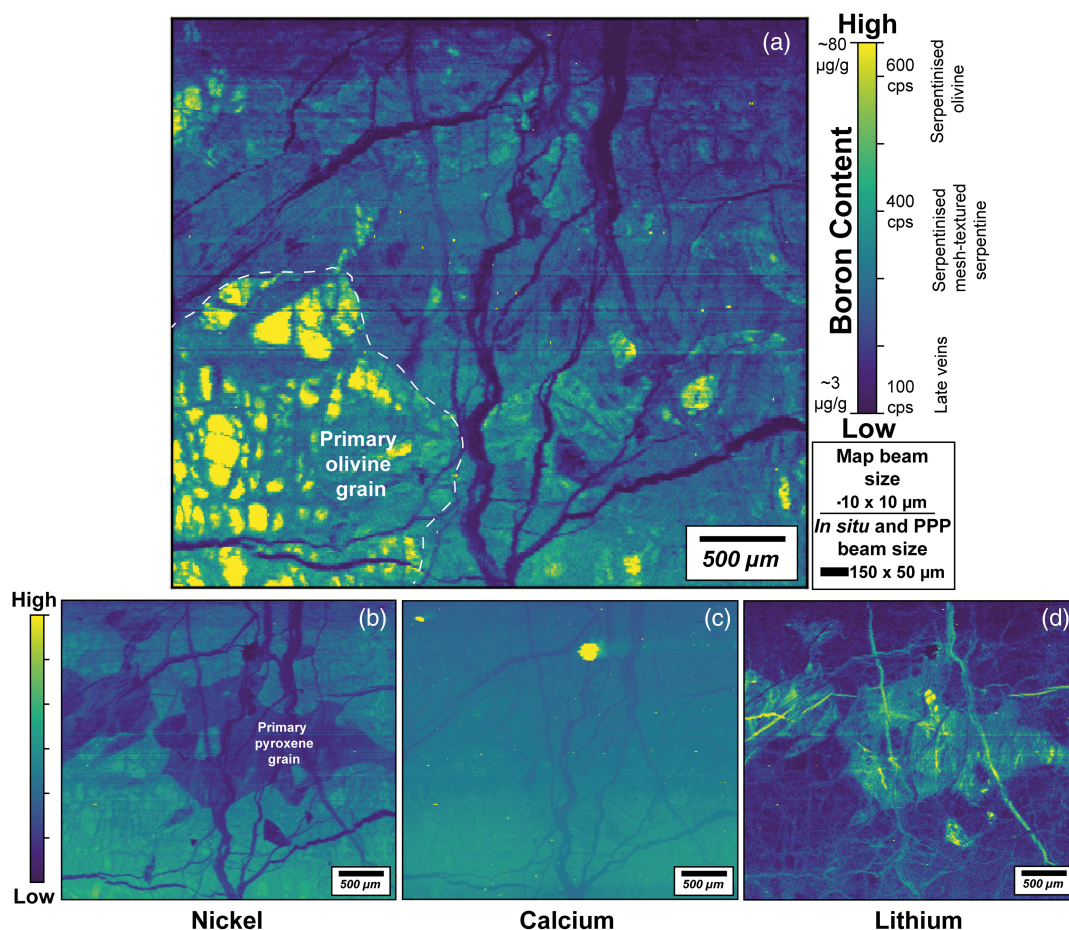


Figure 2 *In situ* (a) boron, (b) nickel, (c) calcium, and (d) lithium elemental count map of a completely serpentinised peridotite (sample AY2-2) from the Artemis Diapir. Data is plotted as counts per second and the relative differences are colour mapped accordingly using a linear colour map scale.

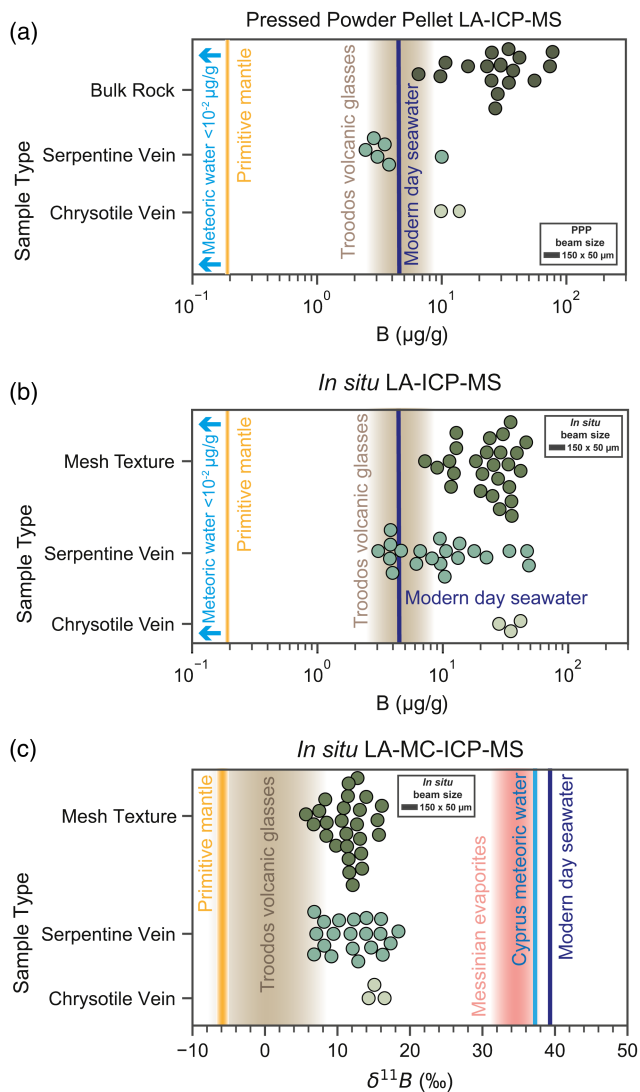


Figure 3 Caltch plots showing (a) B (µg/g) concentration data from pressed powder pellet LA-ICP-MS, (b) *In situ* LA-ICP-MS B (µg/g) concentration, (c) *in situ* LA-MC-ICP-MS δ¹¹B on samples from the Artemis and Olympus Diapir. Boundaries of B (µg/g) and δ¹¹B of Troodos for the reservoirs and reference materials shown are given in the [Supplementary Information](#). Shading shows ±2σ of the reported value. Analytical errors (2 s.e.) are generally smaller than symbols.

serpentine, with lower boron concentrations indicating the differential leaching of boron. A subsequent channelled fluid event precipitated low boron and calcium but high lithium concentration serpentine in discrete veins and further leached boron from the surrounding mesh-textured host (Fig. 2).

An interpretation of multiple temporally distinct alteration events with contrasting fluid sources yielding serpentine of progressively lower boron concentrations is in agreement with the outlined geological history of the Troodos Mantle Sequence and interpretations of previous stable oxygen and hydrogen isotope analyses (Evans *et al.*, 2021). The initial high boron (~80 µg/g) pervasive serpentinisation most likely results from fluid liberated by dehydration of ocean crust and sediments from the subducting Cyprus slab (*e.g.*, Robertson, 1998; Evans *et al.*, 2021, 2024), similar to high B concentration fluids from Mariana forearc serpentinite mud volcanoes that are interpreted to be upwelling slab-derived fluids (~40 µg/g; Benton *et al.*, 2001; Mottl *et al.*, 2004).

The boron isotopic compositions (mean +11.9 ± 3.2 ‰, 1σ, n = 49) of the Olympus and Artemis diapirs are similar to strongly serpentinised ultramafic clasts recovered from Mariana forearc serpentinite mud volcanoes (mean +14.5 ± 4.5 ‰, 1σ, n = 21; Benton *et al.*, 2001). Estimating the δ¹¹B of serpentinising fluids from rock analyses is complex, requiring assumptions about the isotope partitioning of boron fluid species (Spivack and Edmond, 1987; Benton *et al.*, 2001; Boschi *et al.*, 2008; Vils *et al.*, 2009). However, at typical serpentinising conditions in the mantle wedge above subduction zones (pH > 8, ~250 °C; following McCollom *et al.* (2020), experimental determination of pK_B yields values of <5 (following Dickson, 1990; Arcis *et al.*, 2017). Consequently, borate ion is the dominant fluid species resulting in minimal pH dependent fractionation (Spivack and Edmond, 1987; Benton *et al.*, 2001; Boschi *et al.*, 2008; Arcis *et al.*, 2017). The δ¹¹B of the Mariana forearc slab-derived fluid is estimated to be ~+13 ‰ (Benton *et al.*, 2001). We note that the depths of dehydration beneath the Mariana forearc (~15 to 29 km; (Mottl *et al.*, 2004) are similar to the depths of the Cyprus slab beneath the Troodos Mantle Sequence region (Feld *et al.*, 2017; Evans *et al.*, 2021). The range (+6.7 to +18.4 ‰) in δ¹¹B in Olympus and Artemis serpentinites most probably reflects the progressive evolution of a serpentinising fluid that can be modelled by Rayleigh fractionation (Eq. 1; Fig. 4a)

$$\delta^{11}B_{Fluid} = \left(\delta^{11}B_{Fluid(Initial)} + 1000 \right) \left[\frac{[B_{Fluid}]}{[B_{Fluid(Initial)}]} \right]^{(\alpha-1)} - 1000 \quad \text{Eq. 1}$$

Calculations using fractionation factors of α = 0.982 and 0.989 for 100 and 400 °C respectively (following Liu and Tossell, 2005; Boschi *et al.*, 2008) and α = 0.990 and 0.996 for 100 and 200 °C respectively (following Hansen *et al.*, 2017) indicate that as fluid boron is sequestered into serpentine the isotopic composition of the fluid evolves to higher δ¹¹B values, regardless of which fractionation factors are used (Liu and Tossell, 2005; Hansen *et al.*, 2017). Consequently, higher δ¹¹B serpentinites will precipitate further along the flow path (Spivack and Edmond, 1987; Vils *et al.*, 2009).

Hydrothermally altered seafloor lavas commonly have high boron concentrations up to 200 µg/g (Yamaoka *et al.*, 2015b; Fonseca *et al.*, 2017) and the downgoing ancient altered ocean crust of the Cyprus slab is likely a significant reservoir of boron. Consequently, we propose that dewatering and dehydration reactions during the subduction of altered ocean crust of the Cyprus slab liberated the high boron fluid responsible for the initial pervasive serpentinisation of the Troodos Mantle Sequence.

Alternative interpretations such as Cretaceous seawater-derived hydrothermal fluids penetrating through the Troodos ocean crust and into the Olympus and Artemis mantle domains do not match analyses of the Troodos ophiolite that show decreasing boron concentrations (from 207 to 0.3 µg/g) and lower δ¹¹B signatures (from +15.6 to -1.7 ‰) with depth in the ocean gabbro and strong channelling of hydrothermal alteration in the gabbros (Yamaoka *et al.*, 2015b). Assuming that Cretaceous seawater-derived hydrothermal fluids had similar boron isotopic compositions and concentrations to modern fluids (+13.5 to +36.1 ‰ and 4.5 to 16 µg/g respectively; Yamaoka *et al.*, 2015a), the ratio of hydrothermal fluid relative to rock required to form a high boron concentration (80 µg/g) serpentine is very high (from ~5 to ~18) and hence unlikely.

The low boron fluid end member attributed to the second and third Troodos serpentinisation events is consistent with alteration by meteoric waters, where boron derived from the initial pervasive serpentinisation is remobilised by high pH (>9) groundwaters (Evans *et al.*, 2024) leaving residual or

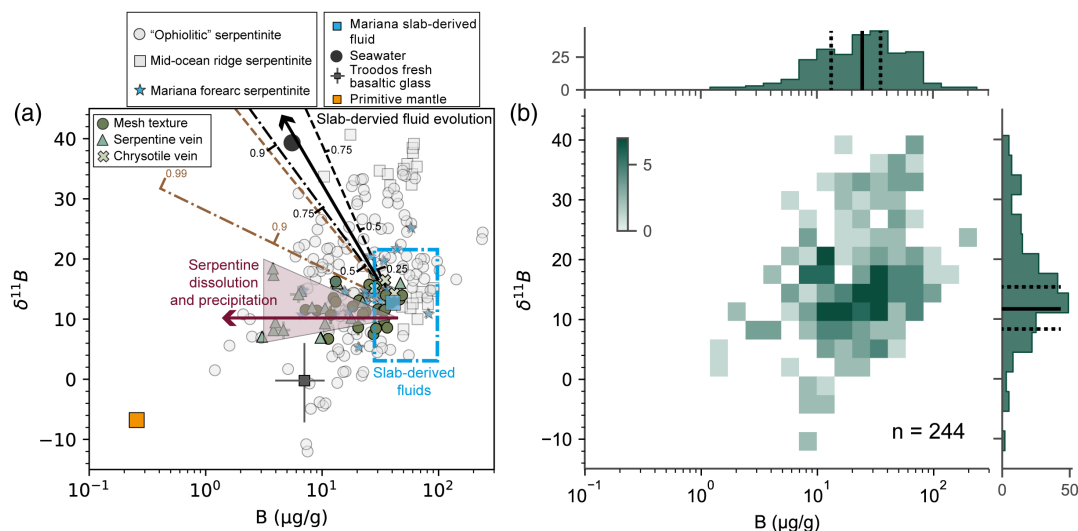


Figure 4 (a) $\delta^{11}\text{B}$ and B ($\mu\text{g/g}$) data from the Olympus and Artemis Diapirs of the Troodos Mantle Sequence compared with compiled published serpentinite measurements. (b) Density histogram plot of compiled and new $\delta^{11}\text{B}$ and B ($\mu\text{g/g}$) data. References for the compiled serpentinite values can be found in the [Supplementary Information](#). Black solid lines of the bivariate histograms refer to the mean values of this study with dashed black lines reflecting $\pm 1\sigma$. Analytical error bars (2 s.e.) are shown for data from this study, most symbols are larger than associated analytical error. Simple fluid evolution model calculations using a Rayleigh fractionation model (Eq. 1) are shown as a black dashed line at 100 °C and black dash-dotted line at 400 °C using serpentine–fluid fractionation equations of Liu and Tossell (2005) and the model equations of Boschi et al. (2008) to estimate a fractionation factor (a). Black dash lines reflect evolution extent as B is removed from the fluid during serpentinisation at 100 and 400 °C respectively. Additionally, calculations using the serpentine–water fractionation values of Hansen et al. (2017) are shown as a brown dashed line at 100 °C and a brown dash-dotted line at 200 °C respectively. Initial fluid $\delta^{11}\text{B}$ and concentration of the model is set as (+13‰ and 40 $\mu\text{g/g}$; Benton et al., 2001; Mottl et al., 2004). Blue box reflects assumed composition of slab-derived fluids at similar downgoing slab depths. Purple arrow and shading show vector of serpentine dissolution and precipitation. Isotopic fractionation between mineral and fluid phases likely results in scattering of the data. Black arrow shows slab-derived fluid evolution vector.

re-precipitated serpentinite with lower boron concentrations but with the boron isotope signature of the initial event (Fig. 3). This meteoric water alteration event is consistent with previous stable oxygen and hydrogen analyses (Magaritz and Taylor, 1974; Nuriel et al., 2009; Evans et al., 2021) and is favoured over alternative interpretations of an evolving dehydrating slab fluid source as proposed for the Mariana system (e.g., Kahl et al., 2015) or distinct fluid pulses with similar boron isotopic compositions but contrasting boron concentrations as suggested for the Mid-Atlantic Ridge Atlantis Massif (e.g., Boschi et al., 2008). These alternative interpretations are inconsistent with previous Troodos stable oxygen and hydrogen isotope compositions.

Novel high resolution elemental serpentinite mapping is a promising avenue in revealing the spatial distribution of boron and other elements (Ni, Ca, Li) in serpentinites. Elemental mapping leveraging a fine ablation beam size of $10 \times 10 \mu\text{m}$ illuminates the heterogeneously distributed contrasting boron signatures that would otherwise not be observed with a coarser ablation beam or bulk rock sampling. This knowledge yields deeper interpretation of determined boron concentrations of pressed powder pellets and *in situ* polished thick sections.

In addition to boron, nickel mapping in serpentinites appears to highlight the original primary texture of mantle peridotites. Calcium mapping shows differences between mesh-textured serpentinite and serpentinite veins. The distributions of lithium and boron differ as these elements show contrasting affinities for serpentinite that pseudomorphs olivine or pyroxene respectively. This is consistent with previous studies that demonstrate serpentinite Li content is dependent on protolith mineralogy and potentially previous melt–rock interaction events (e.g., Kodolányi et al., 2012). The high lithium abundance in some serpentinite veins within the element map (Fig. 2d) likely reflects precipitation from Li-enriched meteoric-derived hyperalkaline groundwaters (Evans et al., 2024).

Future studies to identify and determine the origin of fluid in serpentinised rocks should combine elemental mapping with *in situ* measurements of oxygen and hydrogen stable isotopes as well as novel isotopic tracers such as boron to identify distinctive geochemical and isotopic fingerprints of specific serpentinisation episodes.

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Additional Information

Supplementary Information accompanies this letter at <https://www.geochemicalperspectivesletters.org/article2407>.



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Imaging of boron in altered mantle rocks illuminates progressive serpentinisation episodes

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Supplementary Information

The Supplementary Information includes:

- Analytical Methods
- References for Figures 3 and 4
- Tables S-1 to S-4
- Figure S-1
- Supplementary Information References

Analytical Methods

All samples were prepared and analysed at the School of Ocean and Earth Science Geochemistry Research Facility, National Oceanography Centre Southampton (NOCS), University of Southampton. Samples were sawn to remove surficial weathering rinds and ground to remove contamination from the saw blade. Polished thick sections were prepared to a thickness of ~300 µm. Samples designated for grinding were then washed and ultra-sonicated in Milli-Q (18.2 MΩ H₂O) water and left to dry overnight in an oven at 65 °C. After drying, rock samples were crushed using a manual iron fly-press and wrapped in paper between plastic chopping boards to avoid trace metal contamination. Rock samples were then powdered using a chrome-steel jar mill in a Rocklabs grinder. Prior to analysis, powdered rock samples and standards UB-N and BCR-2 were pressed into pellets using a manual hydraulic press under a pressure equivalent to a mass of 10 T over a period of 30 minutes. An additional standard reference glass BCR-2g was mounted in epoxy resin and polished. Boron concentrations of pressed powder pellets (PPP) and in situ boron concentrations and boron isotopic compositions of thick sections were determined using an Elemental Scientific Lasers NWR193 Excimer laser ablation (LA) system with a TwoVol2 ablation cell (Bozeman, MT, USA). Prior to the acquisition of data, samples

and standards were pre-ablated at reduced power (20 %), lower repetition rate (10 Hz), and a faster raster ablation mode (200 $\mu\text{m/s}$) to remove any contaminants from the surface of the prepared polished thick section or pressed powder pellet.

Boron isotope analyses of polished thick sections were acquired using a Thermo Scientific (Thermo Fisher Scientific, Waltham, MA, USA) Neptune Plus multi-collector inductively coupled plasma (MC-ICP) mass spectrometer equipped with Faraday cup detectors, where ^{10}B and ^{11}B intensities were measured on the L3 and H3 Faraday cups, respectively. Tuning protocols for the LA-MC-ICP-MS method were optimised for sensitivity and stability, broadly following the procedure detailed in (Standish *et al.*, 2019). Typical operating parameters for the laser ablation system were a beam area of 50 x 150 μm , laser power at 50 % (yielding $\sim 6.7 \text{ J/cm}^2$), repetition rate of 50 Hz, raster ablation mode tracking at 10 $\mu\text{m/s}$. Acquisition of reference materials and samples typically integrate ~ 100 cycles, manifest as a ~ 1 mm track on the surface of the thick sections. Samples were run with a forward and backward ablation, resulting in a total ~ 2 mm ablated distance. Gas flow rates during analyses were typically: 750 mL/minute for He, 17 mL/minute for N, and 1.035 mL/minute for Ar. A 40 second on-peak gas blank was analysed before and after ablation, with 60 seconds allowed for sample wash-out prior to the latter. Dynamic blank corrections were applied cycle by cycle assuming a linear relationship between the preceding and succeeding blank measurements. Instrumental mass bias was corrected by sample-standard bracketing with USGS glass reference material BCR-2g and using the reference value $^{11}\text{B}/^{10}\text{B}$ of 4.035, $\delta^{11}\text{B}$ -3.83 ‰ (Kimura *et al.*, 2016). Typically, two analyses of BCR-2g would bracket an analysis of reference serpentinite UB-N pellet with <20 minutes ablation time between analyses of BCR-2g. Data were evaluated offline with cycles falling outside of 3σ of the mean removed. Integration time of the instrument was set to ~ 2 seconds per cycle. Precision and accuracy of the methodology was assessed using similarly analysed pressed powder pellets of reference materials UB-N and BCR-2. The mean internal precision (2 standard errors (SE)) of UB-N and BCR-2 pellets were ± 0.6 ‰ and ± 1.3 ‰ respectively. The mean $\delta^{11}\text{B}$ values of UB-N and BCR-2 were $+13.5 \pm 2.0$ ‰ (2σ , $n=20$) and -5.5 ± 2.2 ‰ (2σ , $n=12$), resulting in mean inaccuracies in $\Delta\delta^{11}\text{B}$, defined as $\delta^{11}\text{B}_{\text{Reported}} - \delta^{11}\text{B}_{\text{Laser}}$ following (Standish *et al.*, 2019) of 0.4 ‰ for UB-N and -0.4 ‰ for BCR-2 (**Table S-1**).

Boron concentration maps of polished thick sections were analysed with the laser ablation system coupled to an Agilent (Agilent Technologies Inc., CA, USA) 8900 Triple Quadrupole ICP-MS. Typical operating parameters for the laser ablation system were a beam area of 10 x 10 μm , repetition rate of 50 Hz, raster ablation mode tracking of 20 $\mu\text{m/s}$, and a laser power density of $\sim 6 \text{ J/cm}^2$. The Agilent ICP-MS acquisition time was ~ 0.375 seconds per cycle. Gas flow rates during analyses were typically: 800 mL/minute for He, 4 mL/minute for N, and 0.6 mL/minute for Ar. The area mapped was 3336 x 3000 μm , requiring ~ 18 hours of instrument acquisition time.

Spot boron concentrations of polished thick sections and pressed powder pellets were determined using the laser ablation system coupled to a Thermo Scientific Element 2 ICP-MS. Typical operating parameters for the laser ablation system were a beam area of 50 x 150 μm , repetition rate of 30 Hz, raster ablation mode tracking of 10 $\mu\text{m/s}$, and a laser power density of $\sim 6 \text{ J/cm}^2$. The integration time was 2 seconds per cycle. Gas flow rates during analyses were typically: 800 mL/minute for He, 4 mL/minute for N, and 0.6 mL/minute for Ar. Data was evaluated offline with both dynamic



blank and drift correction. The mean internal precision (2 SE) of the reference materials UB-N and BCR-2g for boron concentration determination were $\pm 4.4 \mu\text{g/g}$ and $\pm 0.4 \mu\text{g/g}$ respectively. The mean determined B concentrations of UB-N and BCR-2g were $136.4 \pm 8.4 \mu\text{g/g}$ (2σ , $n=80$) and $2.7 \pm 1.2 \mu\text{g/g}$ (2σ , $n=80$) respectively, resulting in mean inaccuracies (ΔB , defined as $\text{B}_{\text{Reported}} - \text{B}_{\text{Laser}}$) of $3.6 \mu\text{g/g}$ for UB-N and $5.3 \mu\text{g/g}$ for BCR-2g (**Table S-1**).

Analytical results are reported in Table S-1, Table S-2, and Table S-3.

References for Figures 3 and 4

On Figure 3 the boundaries of B ($\mu\text{g/g}$) and $\delta^{11}\text{B}$ for Troodos volcanic glasses are from Fonseca *et al.*, (2017), modern seawater Foster *et al.*, (2010) and Spivack and Edmond (1987), Messinian evaporites Paris *et al.*, (2010) and primitive mantle Chaussidon and Jambon (1994). Cyprus meteoric water concentration are from Rose-Koga *et al.* (2006), meteoric water boron isotope composition is estimated from the equations of Boronina *et al.*, (2005) and Rose-Koga *et al.* (2006).

On Figure 4 the compiled serpentinite values are from the: Mariana forearc (Benton *et al.*, 2001); Mid-ocean ridge serpentinite, Atlantis Massif (Boschi *et al.*, 2008), Mid-Atlantic Ridge ODP Leg 209 (Vils *et al.*, 2009), and dredged serpentinites (Spivack and Edmond, 1987); ‘Ophiolitic’ serpentinite, Cerro Del Almirez (Harvey *et al.*, 2014), ophiolitic serpentinites (Martin *et al.*, 2016), Corsica and Western Alps (Martin *et al.*, 2020), Californian serpentinites (Yamada *et al.*, 2019), and Erro Tobbio serpentinite (Scambelluri and Tonarini, 2012). Boundaries of B ($\mu\text{g/g}$) and $\delta^{11}\text{B}$ of Troodos volcanic glasses (Fonseca *et al.*, 2017), modern seawater (Foster *et al.*, 2010; Spivack and Edmond, 1987), and primitive mantle (Chaussidon and Jambon, 1994; Marschall *et al.*, 2017) are also shown.



Supplementary Tables

Table S-1 $\delta^{11}\text{B}$ Precision and accuracy table for LA-MC-ICP-MS and LA-ICP-MS

| | $\delta^{11}\text{B}$ (‰) Reference Value | $\delta^{11}\text{B}$ (‰) This study | $\Delta\delta^{11}\text{B}$ | B ($\mu\text{g/g}$) Reference Value | B ($\mu\text{g/g}$) This study | ΔB |
|----------------------------------|--|---|-----------------------------|---|--|---------------------|
| UB-N (pressed powder pellet) | +13.1 ‰ (Gangjian <i>et al.</i> , 2013) | +13.5 ± 2 ‰ (2 σ , n=20) | +0.4 ‰ | 140 $\mu\text{g/g}$ (Govindaraju, 1995) | 136.4 ± 8.4 $\mu\text{g/g}$ (2 σ , n=80) | 3.6 $\mu\text{g/g}$ |
| BCR-2 (pressed powder pellet) | -5.9 ‰ (Gangjian <i>et al.</i> , 2013) | -5.5 ± 2.2 ‰ (2 σ , n=12) | -0.4 ‰ | 4.3 ± 3.9 $\mu\text{g/g}$ (2 σ ; n=7; compiled (Jochum <i>et al.</i> , 2005) | - | - |
| BCR-2g (glass) | -3.8 ‰ (Kimura <i>et al.</i> , 2016) | - | - | 8 ± 8.1 $\mu\text{g/g}$ (2 σ ; n=34; compiled (Jochum <i>et al.</i> , 2005) | 2.7 ± 1.2 $\mu\text{g/g}$ (2 σ , n=80) | 5.3 $\mu\text{g/g}$ |



Table S-2 In situ polished thick section summary results

| | Mesh Texture | | Serpentine Vein | | Chrysotile Vein | | All in situ polished thick sections | | Whole Rock | Serpentine Vein | Chrysotile Vein | All Pressed Powder Pellets |
|-------------------------|--------------------|-------------|--------------------|----------|--------------------|----------|-------------------------------------|-------------|------------|-----------------|-----------------|----------------------------|
| | $\delta^{11}B$ (‰) | B (ug/g) | $\delta^{11}B$ (‰) | B (ug/g) | $\delta^{11}B$ (‰) | B (ug/g) | $\delta^{11}B$ (‰) | B (ug/g) | B (ug/g) | B (ug/g) | B (ug/g) | B (ug/g) |
| # of samples | 26 | 26 | 20 | 20 | 3 | 3 | 49 | 49 | 17 | 6 | 2 | 25 |
| Average | 11.5 | 26 | 12 | 12 | 15.3 | 35 | 11.9 | 21 | 34 | 6 | 12 | 25 |
| 1 σ (symmetric) | 2.7 | 12 | 3.9 | 11 | 1.1 | 7 | 3.2 | 13 | 21 | 3 | 3 | 21 |
| Median | 11.5 | 25 | 11.9 | 9 | 15.1 | 35 | 11.9 | 19 | 29 | 4 | 12 | 24 |
| 1 σ (asymmetric) | +2.3/- 2.9 | +11/- 13 | +4.1/- 3.7 | +8/-6 | +1/-0.5 | +5/-5 | +3.6/-3.6 | +16/- 12 | +21/-15 | +6/-1 | +1/-1 | +15/-17 |
| Min | 5.8 | 7 | 6.9 | 3 | 14.3 | 28 | 5.8 | 3 | 7 | 3 | 10 | 3 |
| Max | 16.2 | 49 | 18.4 | 47 | 16.5 | 42 | 18.4 | 49 | 80 | 10 | 14 | 80 |

Table S-3 (overleaf) In situ boron concentrations and boron isotopic compositions of polished thick sections.

| Table S-3 Sample Name | Sample Type | Sample Area | $^{11}\text{B}/^{10}\text{B}$ | $\pm 2\text{SE}$ | $\delta^{11}\text{B}$ | $\pm 2\text{SE}$ | B ($\mu\text{g}/\text{g}$) | $\pm 2\text{SE}$ |
|--------------------------|---------------------------|-------------|-------------------------------|------------------|-----------------------|------------------|---------------------------------|------------------|
| AA4-1 | Mesh texture | Artemis | 4.0776 | 0.0028 | 6.72 | 0.70 | 11.30 | 0.34 |
| AA4-3 | Mesh texture | Artemis | 4.0807 | 0.0032 | 7.49 | 0.79 | 27.60 | 0.67 |
| AA4-5 | Serpentine Vein | Artemis | 4.0835 | 0.0058 | 8.18 | 1.41 | 4.65 | 0.67 |
| AA4-7 | Serpentine Vein | Artemis | 4.0790 | 0.0038 | 7.08 | 0.94 | 3.05 | 0.09 |
| AA4-9 | Serpentine Vein | Artemis | 4.0833 | 0.0039 | 8.15 | 0.95 | 3.82 | 0.23 |
| AA4-11 | Serpentine Vein | Artemis | 4.0886 | 0.0036 | 9.46 | 0.89 | 3.97 | 0.07 |
| AH1-3 2 | Mesh texture | Olympus | 4.0851 | 0.0035 | 8.58 | 0.86 | 36.23 | 0.69 |
| AH1-3 4 | Mesh texture | Olympus | 4.0845 | 0.0043 | 8.43 | 1.04 | 29.00 | 1.37 |
| AH1-3 5 | Mesh texture | Olympus | 4.0853 | 0.0053 | 8.63 | 1.29 | 20.41 | 0.63 |
| AR3-1 conc line | Mesh texture | Artemis | 4.1068 | 0.0008 | 13.97 | 0.19 | 35.27 | 0.58 |
| AR3-1 serp andradite | Serpentine andradite Vein | Artemis | 4.1000 | 0.0007 | 12.27 | 0.18 | 33.96 | 0.35 |
| AR3-1 chrysotile | Chrysotile Vein | Artemis | 4.1170 | 0.0009 | 16.50 | 0.22 | 34.86 | 0.52 |
| AR3-1 recrystallised | Mesh texture | Artemis | 4.1034 | 0.0016 | 13.11 | 0.40 | 41.75 | 1.48 |
| AR3-1 lower chrysotile | Chrysotile Vein | Artemis | 4.1114 | 0.0015 | 15.09 | 0.37 | 28.07 | 1.70 |
| AR3-1 Ribbon | Mesh texture | Artemis | 4.1040 | 0.0040 | 13.27 | 0.99 | 46.24 | 1.81 |



| Table S-3 Sample Name | Sample Type | Sample Area | $^{11}\text{B}/^{10}\text{B}$ | $\pm 2\text{SE}$ | $\delta^{11}\text{B}$ | $\pm 2\text{SE}$ | B ($\mu\text{g}/\text{g}$) | $\pm 2\text{SE}$ |
|----------------------------------|---|-------------|-------------------------------|------------------|-----------------------|------------------|---------------------------------|------------------|
| AR3-1 Serpentine in centre | Serpentine Vein | Artemis | 4.1067 | 0.0054 | 13.95 | 1.32 | 48.65 | 0.47 |
| AH2-1 1 | Mesh texture | Olympus | 4.0936 | 0.0063 | 10.69 | 1.53 | 11.87 | 0.49 |
| AH2-1 6 | Black fine grained serpentine vein | Olympus | 4.1074 | 0.0058 | 14.11 | 1.40 | 6.28 | 0.98 |
| AH2-1 black vein | Black fine grained serpentine vein | Olympus | 4.0879 | 0.0019 | 9.28 | 0.47 | 10.58 | 0.44 |
| AH2-1 2 | Mesh texture | Olympus | 4.1049 | 0.0022 | 13.51 | 0.53 | 12.52 | 0.70 |
| AH2-1 3 | Mesh texture | Olympus | 4.1160 | 0.0021 | 16.24 | 0.52 | 13.08 | 0.60 |
| AH2-1 4 | Mesh texture | Olympus | 4.1024 | 0.0015 | 12.88 | 0.38 | 13.08 | 1.61 |
| BA1-X1 | Mesh texture | Artemis | 4.0957 | 0.0023 | 11.22 | 0.56 | 23.70 | 0.56 |
| BA1-X2 | Mesh texture | Artemis | 4.0943 | 0.0019 | 10.87 | 0.47 | 22.41 | 0.75 |
| BA1 X3 | Mesh texture | Artemis | 4.1131 | 0.0027 | 15.52 | 0.65 | 28.88 | 2.00 |
| BA1 X4 | Serpentine Vein | Artemis | 4.1160 | 0.0027 | 16.24 | 0.67 | 13.22 | 1.59 |
| BA1 X5 | Serpentine Vein | Artemis | 4.1204 | 0.0031 | 17.33 | 0.76 | 3.83 | 0.20 |
| BA1 X6 | Serpentine Vein | Artemis | 4.1248 | 0.0040 | 18.41 | 0.98 | 3.78 | 0.17 |
| BA1 Y1 | Mesh texture | Artemis | 4.1033 | 0.0046 | 13.09 | 1.12 | 20.66 | 1.92 |



| Table S-3 Sample Name | Sample Type | Sample Area | $^{11}\text{B}/^{10}\text{B}$ | $\pm 2\text{SE}$ | $\delta^{11}\text{B}$ | $\pm 2\text{SE}$ | B ($\mu\text{g/g}$) | $\pm 2\text{SE}$ |
|--------------------------|-----------------------|-------------|-------------------------------|------------------|-----------------------|------------------|--------------------------|------------------|
| BA1 Y2 | Serpentine Vein | Artemis | 4.1095 | 0.0042 | 14.64 | 1.01 | 6.61 | 0.50 |
| BA1 Y3 | Serpentine Vein | Artemis | 4.1147 | 0.0028 | 15.93 | 0.68 | 13.57 | 0.65 |
| BA1 Y4 | Black serpentine vein | Artemis | 4.1150 | 0.0030 | 16.01 | 0.73 | 46.98 | 1.92 |
| BA1 Y5 | Mesh texture | Artemis | 4.1139 | 0.0031 | 15.72 | 0.75 | 25.44 | 0.66 |
| BA1 Z1 | Serpentine Vein | Artemis | 4.0981 | 0.0025 | 11.80 | 0.61 | 10.60 | 0.91 |
| BA1 Z2 | Serpentine Vein | Artemis | 4.0987 | 0.0027 | 11.95 | 0.67 | 8.21 | 0.44 |
| BA1 Z3 | Serpentine Vein | Artemis | 4.1024 | 0.0030 | 12.87 | 0.72 | 22.27 | 0.94 |
| AX1 X1 mesh | Mesh texture | Artemis | 4.0993 | 0.0010 | 12.10 | 0.25 | 18.36 | 0.34 |
| AX1 X2 mesh | Mesh texture | Artemis | 4.0974 | 0.0009 | 11.63 | 0.22 | 30.18 | 0.57 |
| AX1 X3 recryz | Mesh texture | Artemis | 4.0961 | 0.0013 | 11.32 | 0.32 | 33.33 | 0.40 |
| AX1 X4 bastite | Mesh texture | Artemis | 4.0900 | 0.0020 | 9.80 | 0.49 | 33.92 | 0.63 |
| AX1 X5 vein edge | Mesh texture | Artemis | 4.0970 | 0.0011 | 11.54 | 0.26 | 7.20 | 0.40 |
| AX1 X6 polyg vein | Serpentine Vein | Artemis | 4.0917 | 0.0009 | 10.22 | 0.22 | 17.80 | 0.59 |
| AX1 X7 recryz vein ed | Mesh texture | Artemis | 4.0966 | 0.0012 | 11.44 | 0.30 | 9.02 | 0.22 |
| AX1 X8 recryz | Mesh texture | Artemis | 4.0970 | 0.0013 | 11.54 | 0.31 | 34.71 | 0.84 |
| AX1 X9 mesh | Mesh texture | Artemis | 4.1017 | 0.0013 | 12.69 | 0.31 | 38.97 | 0.49 |



| Table S-3 Sample Name | Sample Type | Sample Area | $^{11}\text{B}/^{10}\text{B}$ | $\pm 2\text{SE}$ | $\delta^{11}\text{B}$ | $\pm 2\text{SE}$ | B ($\mu\text{g}/\text{g}$) | $\pm 2\text{SE}$ |
|---------------------------------|--------------------|-------------|-------------------------------|------------------|-----------------------|------------------|---------------------------------|------------------|
| AX1 X10 chrysotile | Chrysotile Vein | Artemis | 4.1082 | 0.0010 | 14.30 | 0.25 | 41.68 | 0.76 |
| AJ1 Ant Back 1 | Antigorite Vein | Olympus | 4.0783 | 0.0006 | 6.91 | 0.14 | 9.80 | 0.18 |
| AJ1 Ant Back 2 | Antigorite Vein | Olympus | 4.0782 | 0.0006 | 6.89 | 0.15 | 9.67 | 0.13 |



Table S-4 Pressed Powder Pellets Results Table

| Sample | Location | Sample Type | B ($\mu\text{g/g}$) | $\pm 2\text{SE}$ |
|--------|----------|------------------------------|-----------------------|------------------|
| AG2 | Olympus | Olympus Bulk Rock | 58.18 | 2.25 |
| AH1 | Olympus | Olympus Bulk Rock | 36.23 | 0.69 |
| AH2 | Olympus | Olympus Bulk Rock | 11.24 | 2.12 |
| CX1 | Olympus | Olympus Bulk Rock | 6.83 | 0.48 |
| PP2 | Olympus | Olympus Bulk Rock | 26.29 | 0.52 |
| CJ1 | Olympus | Olympus Bulk Rock | 10.23 | 0.49 |
| AJ1 | Olympus | Serpentine (antigorite) vein | 9.80 | 0.18 |
| CO1B | Olympus | Serpentine vein | 2.50 | 0.55 |
| CO2 | Olympus | Serpentine vein | 2.91 | 0.59 |
| AA3 | Artemis | Serpentine vein | 3.05 | 0.09 |
| BA1A | Artemis | Serpentine vein | 3.78 | 0.17 |
| CU5 | Artemis | Serpentine vein | 3.49 | 0.36 |
| CR1 | Artemis | Chrysotile | 10.07 | 2.11 |
| CU3 | Artemis | Chrysotile vein | 13.87 | 0.86 |
| AA4-2 | Artemis | Artemis Bulk Rock (Halo) | 27.60 | 0.67 |
| A03C | Artemis | Artemis Bulk Rock (Halo) | 35.26 | 1.42 |
| BA1B | Artemis | Artemis Bulk Rock (Halo) | 28.88 | 2.00 |
| CU4 | Artemis | Artemis Bulk Rock | 25.89 | 2.32 |
| CU2 | Artemis | Artemis Bulk Rock | 43.11 | 2.82 |
| BA1C | Artemis | Artemis Bulk Rock | 23.70 | 0.56 |
| A02 | Artemis | Artemis Bulk Rock | 75.77 | 10.36 |
| AR1 | Artemis | Artemis Bulk Rock | 30.48 | 1.71 |
| CU1 | Artemis | Artemis Bulk Rock | 38.37 | 4.45 |
| AR2 | Artemis | Artemis Bulk Rock | 79.54 | 6.35 |
| PP1 | Artemis | Artemis Bulk Rock | 16.68 | 0.65 |



Supplementary Figures

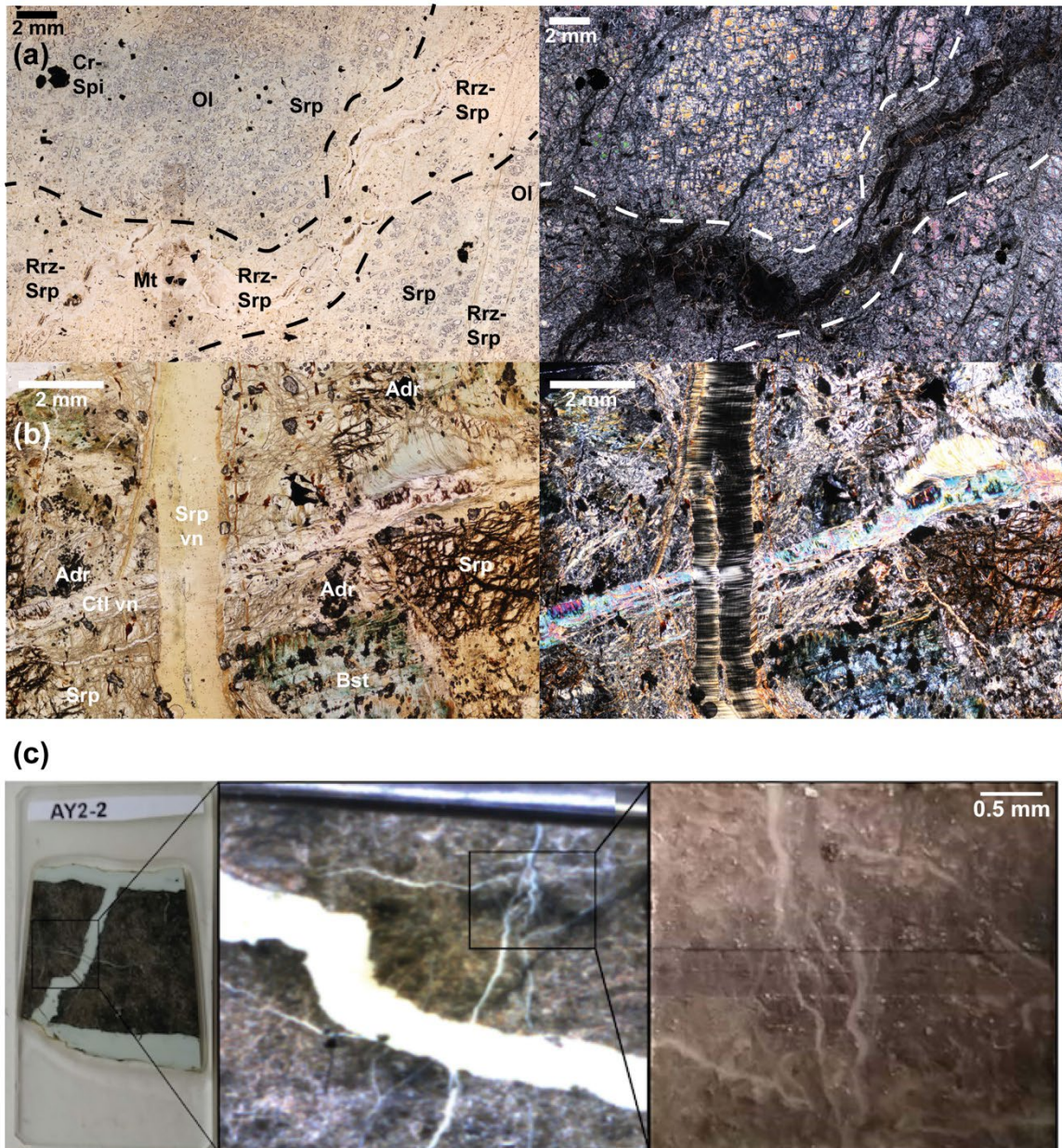


Figure S-1 (a) Representative sample of partially serpentinised mantle peridotite with olivine-serpentine mesh-texture and variably recrystallised irregular veins shown. Sample AH2 from the Olympus Diapir. (b) Representative sample of completely serpentinised mantle peridotite with a variably recrystallised mesh texture, crack-seal serpentine vein and fibrous chrysotile vein. Sample AX1 from the Artemis Diapir. Adr = Andradite, Srp vn = Serpentine vein, Ctl vn = Chrysotile vein, Bst = bastite (relict Opx), Ol = Olivine, Rrz-Srp = Recrystallised serpentine, Srp = Serpentine, Mt = Magnetite. (c) Image of thick section AY2-2 used for elemental mapping with magnified images of the mapped region.

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