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Isotopic variation in Semail Ophiolite lower crust reveals crustal-level melt aggregation

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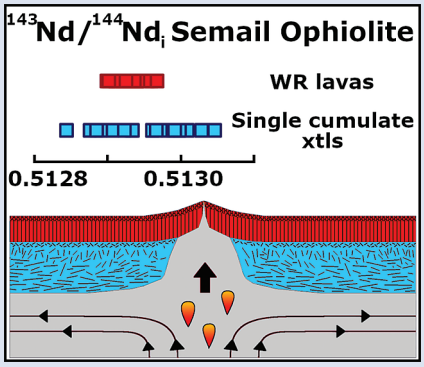
Abstract



The scale and magnitude of compositional heterogeneity in the mantle has important implications for the understanding of the evolution of Earth. Heterogeneity of the upper mantle is often evaluated based on mid-ocean ridge basalt compositions, despite their homogenisation prior to eruption. In this study we present Nd and Sr isotope data obtained by micro-drilling single plagioclase and clinopyroxene crystals in gabbroic cumulates of the Semail Ophiolite (Oman) and show that mantle source variability is better preserved in the lower crust than in the extrusive suite. Analysis of sub-nanogram quantities of Nd in plagioclase revealed a range in $^{143}Nd/^{144}Nd_i$ in the Wadi Abyad crustal section that is three times greater than recorded in the extrusive suite. The isotopic variability is preserved in plagioclase, whereas clinopyroxene is isotopically homogeneous. These data imply that the mantle is heterogeneous on the scale of melt extraction, and that a significant proportion of homogenisation of erupted melts occurs in the oceanic crust, not the mantle.

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Introduction

Planetary differentiation and plate tectonics have led to marked compositional heterogeneity in Earth's mantle. Quantification of the scale and magnitude of mantle heterogeneity has important implications for the understanding of these processes and is one of the major challenges in geochemistry. Because the mantle cannot be sampled directly, this problem remains unresolved. Mid-ocean ridge basalts (MORB) are the most voluminous magmatic rocks on Earth, continually resurfacing two-thirds of the planet on an approximately 200 Myr timescale. They form through decompression melting of the upper mantle and typically undergo partial crystallisation in crustal magma chambers before being erupted onto the seafloor (*e.g.*, Klein and Langmuir, 1987). Long-lived radiogenic isotopes are insensitive to crystallisation processes and are robust mantle source tracers. Many studies have therefore focused on the isotopic composition of MORB to assess along-ridge mantle heterogeneity (*e.g.*, Zindler *et al.*, 1979). Magmatic processes at mid-ocean ridges, however, act to homogenise discrete magma batches (*e.g.*, Rubin and Sinton, 2007), thus obscuring variability at the scale of melt extraction. Homogenisation of isotopic variability is particularly common at fast-spreading ridges, where higher melt production enhances magma mixing and reduces mantle-related variation in MORB compositions (Rubin and Sinton, 2007). Melt batch-scale heterogeneity is partially preserved in phenocrysts and their melt inclusions, which are believed to record unaggregated melt compositions (*e.g.*, Lange *et al.*, 2013). This suggests that lower crustal cumulates potentially record a greater isotopic variability than their extrusive counterparts.

Explicit consideration for the scale of sampling is essential for a full understanding of the lower oceanic crust due to the presence of significant compositional heterogeneity at scales ranging from millimetres to hundreds of metres (*e.g.*, Lissenberg *et al.*, 2013; Coogan, 2014). Whole rock geochemical analyses cannot resolve sample- or crystal-scale variations. Isotopic heterogeneity within individual minerals is to be expected if crystal growth spans replenishment and magma mixing episodes (Davidson and Tepley, 1997; Lange *et al.*, 2013). Furthermore, whole rock isotopic data may mask heterogeneity if overgrowth on cumulus minerals and/or intercumulus minerals crystallised from isotopically distinct migrating interstitial melts (Yang *et al.*, 2013).

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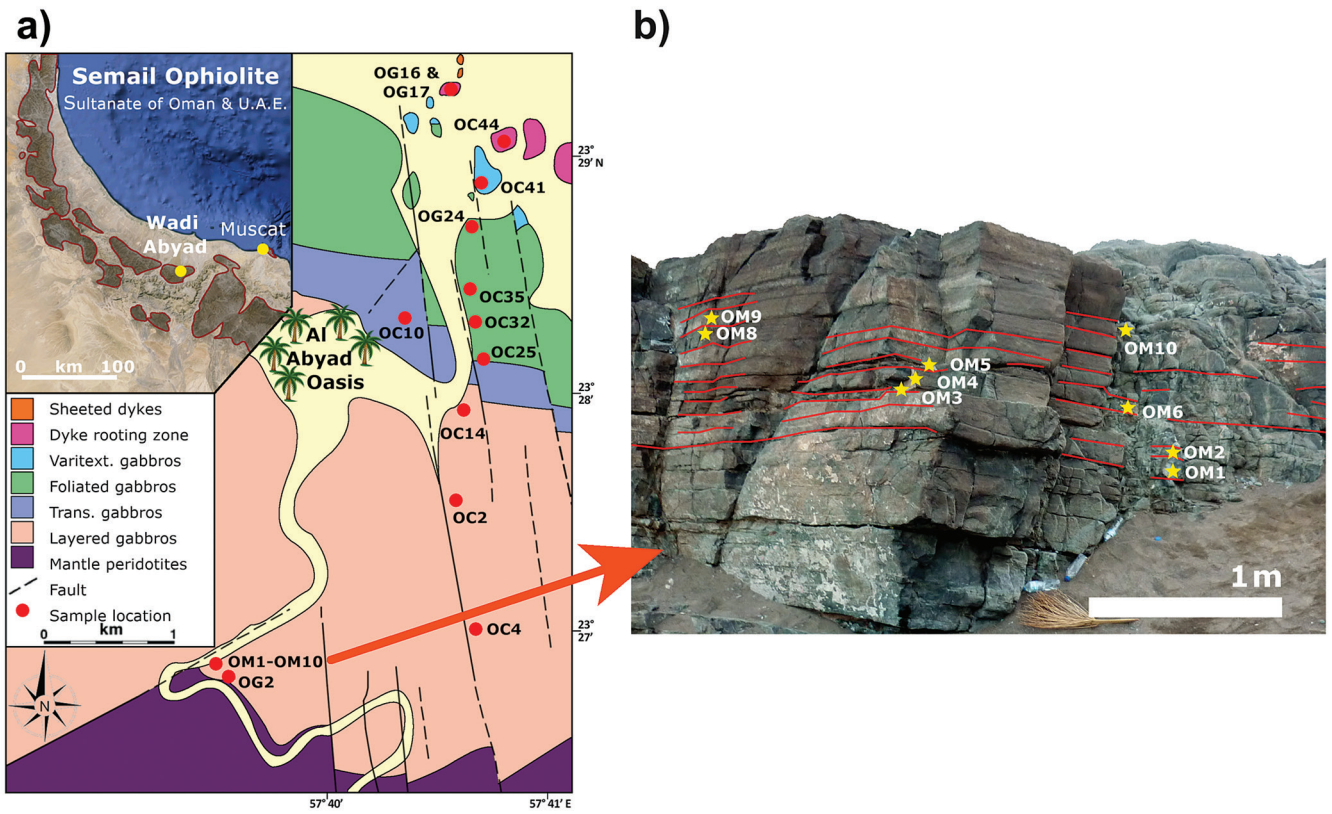

Figure 1 **(a)** Geological map of the lower crustal section at Wadi Abyad, in the Semail Ophiolite, Oman, with sample locations indicated (after MacLeod and Yaouancq, 2000; sample names were shortened by omitting the numerals '95' preceding the letters 'OC' and 'OG' and '14' preceding 'OM'). **(b)** An outcrop at the base of the layered gabbros was sampled at ~15 cm intervals (one sample per modal layer).

Due to its inaccessibility, oceanic crust is often studied using on-land analogues such as the 96 Ma Semail Ophiolite in Oman and the UAE (Nicolas, 1989; Rioux *et al.*, 2013). Although the Semail Ophiolite may have formed in a subduction-related setting (MacLeod *et al.*, 2013), its mode of formation and isotopic signature is comparable to MORB (Godard *et al.*, 2006) and therefore an ideal location for this study. A micro-sampling technique for plagioclase and clinopyroxene crystals allowed isotopic analyses of single mineral grains for which the textural context and geochemical composition were known and avoided averaging isotopic variations within a sample.

Geological Setting and Methods

The Semail Ophiolite lower crust at Wadi Abyad consists of a ~1700 m thick sequence of modally layered gabbros, overlain by non-layered, steeply-foliated gabbros (~650 m) and ~150 m of varitextured gabbros and a 'dyke rooting zone' transition into the base of a sheeted dyke complex (MacLeod and Yaouancq, 2000). Thirteen gabbros and olivine-gabbros spanning the entire lower crustal stratigraphy were sampled at an average interval of ~200 m, complemented with nine sampled modal layers (~15 cm thick) from a single outcrop near the base of the layered gabbros (Fig. 1). Major and trace element data collected from thick sections by EMP and LA-ICPMS (Supplementary Information, Table S-4e) were combined with EDS-SEM major element mapping of polished rock slabs to select domains most suitable for micro-sampling in the latter. Single crystal isotopic analyses (Nd and Sr in plagioclase; Nd in clinopyroxene) were performed on material obtained by micro-drilling individual crystals. The amount of Nd recovered ranged between ~1-5 ng in clinopyroxene and ~0.1-0.5 ng in plagioclase, while ~500 ng Sr was obtained from plagioclase. Plagioclase samples were spiked with a ^{149}Sm-^{150}Nd mixed-spike prior to digestion for Sm/Nd isotope dilution analysis. Chemical separation for Sr, Nd and Sm was performed using standard ion exchange techniques and samples were measured on a Thermo Scientific Triton *Plus* thermal ionisation mass spectrometer (TIMS), using 10^{13} Ω amplifiers for the measurement of Sm (50-250 pg) and Nd in the plagioclase samples. Isotope ratios were age-corrected to 96 Ma (subscript "i"; Rioux *et al.*, 2013). The reproducibility (2 SD) of ^{143}Nd/^{144}Nd for BCR-2 reference material is 161 ppm for aliquots of 0.15-0.34 ng Nd and 82 ppm for aliquots of 1.9-2.6 ng Nd. A detailed description of the methods and results for internal and external standards is provided in the Supplementary Information.

Results

Plagioclase and clinopyroxene crystals have ^{143}Nd/^{144}Nd$_i$ typical of MORB, with values ranging from 0.512844 to 0.513046 (Figs. 2 and 3, Supplementary Information, Table S-4a-d). Clinopyroxene samples reveal an almost uniform Nd isotope composition, with a range of 225 ppm and only two samples, from the foliated gabbros and dyke rooting zone, are outside error (by 10 ppm); the ppm notation here indicating the range relative to the average ^{143}Nd/^{144}Nd$_i$ of the dataset (0.512936).

Plagioclase samples display a greater range in Nd isotope composition, 395 ppm, with the lowest values outside error of the highest values by up to 88 ppm (Fig. 2). This range is well in excess of the variation found in the extrusive suite of the entire Semail Ophiolite (whole rock; 134 ppm). It is also significantly larger than existing whole rock and mineral separate data for the lower crust (123 ppm). It is, however, comparable to the range found in gabbroic cumulates in the Maqsad mantle harzburgites (whole rock; 414 ppm), which are interpreted to be individual melt batches within a fossil mantle diapir (Benoit *et al.*, 1996; Fig. 4, all data sources in figure caption).

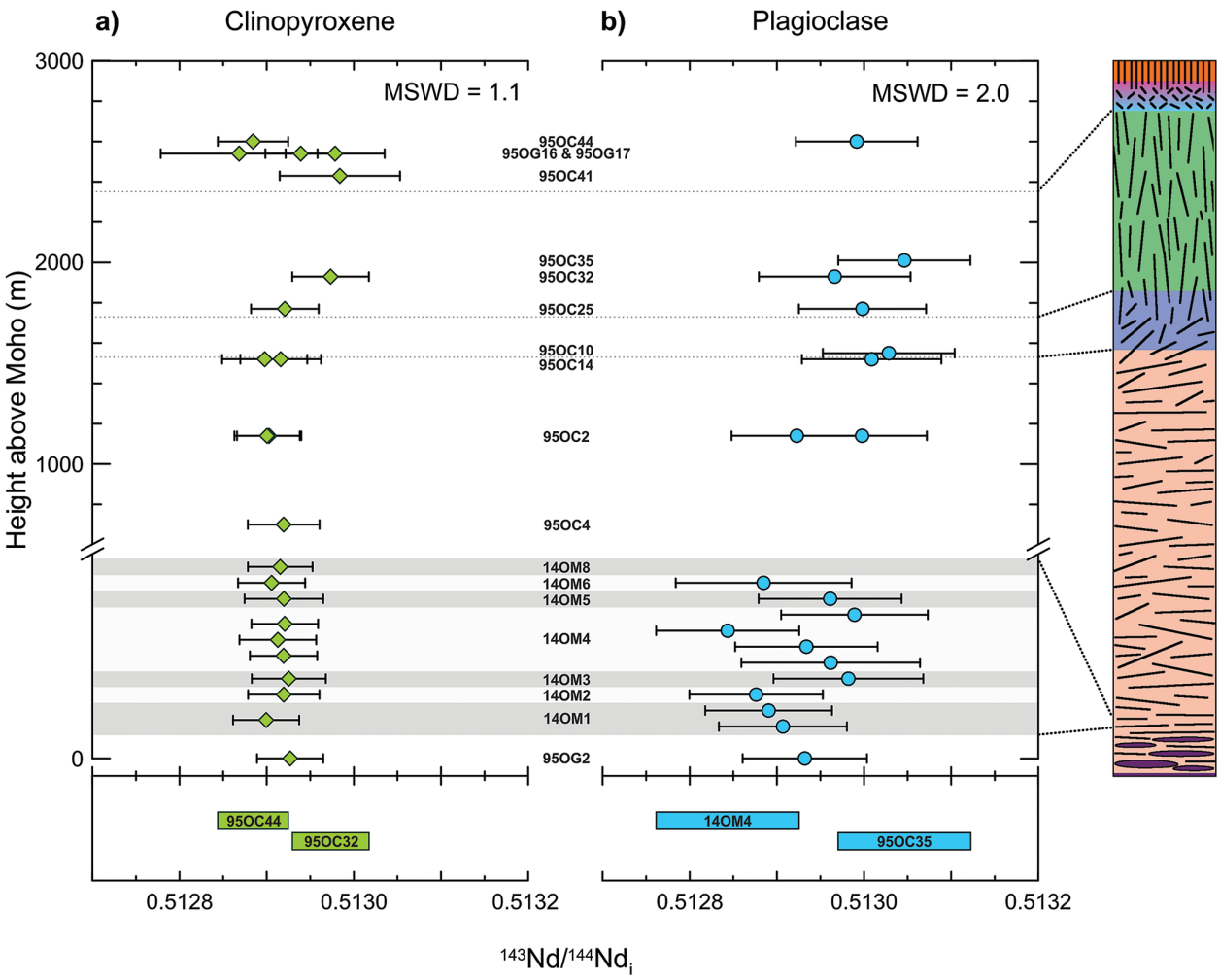

Figure 2 Age-corrected $^{143}Nd/^{144}Nd_i$ of **(a)** single clinopyroxene and **(b)** plagioclase crystals in the Abyad section. Clinopyroxene has a near uniform composition whereas plagioclase shows variability outside analytical uncertainty. Grey and white alternating bars at the base of the section indicate the individual modal layers of the sampled layered gabbro outcrop (not to scale). Coloured bars at the bottom of the figure represent the uncertainty for the two most extreme samples.

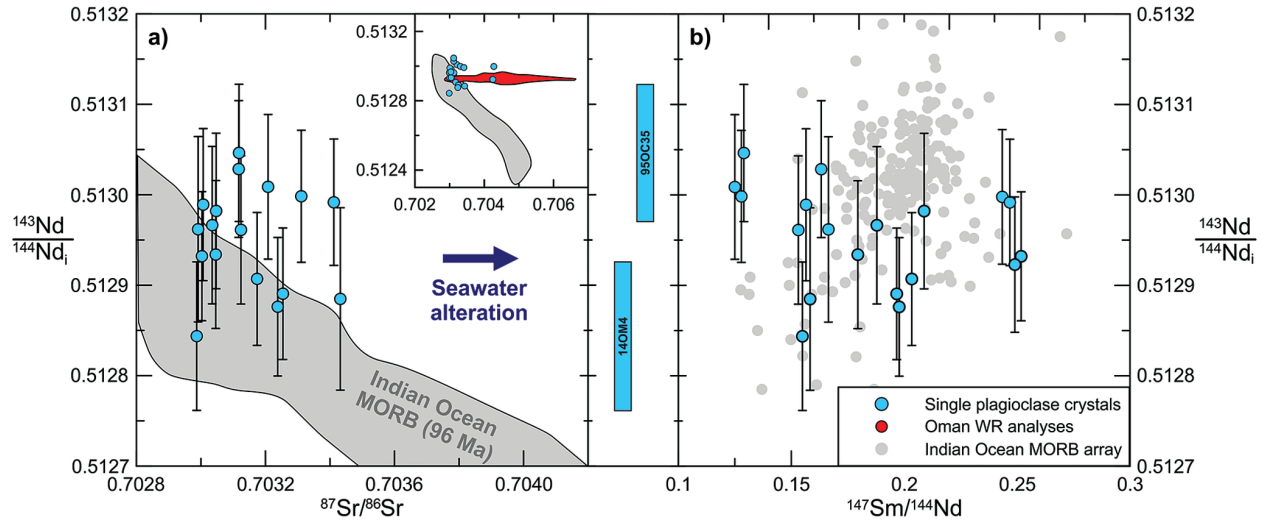

Figure 3 **(a)** Age-corrected $^{143}Nd/^{144}Nd_i$ and $^{87}Sr/^{86}Sr$ compositions of plagioclase samples. Indian Ocean MORB data from the PetDB database and age-corrected to 96 Ma using model Sm/Nd and Rb/Sr ratios for depleted MORB mantle (Workman and Hart, 2005). The ranges of whole rock age-corrected Sr and Nd isotope analyses of the plutonic and extrusive series of the Semail Ophiolite are shown in the inset (McCulloch *et al.*, 1980; Godard *et al.*, 2006; Rioux *et al.*, 2012, 2013; de Graaff, 2016). **(b)** Age-corrected $^{143}Nd/^{144}Nd_i$ and $^{147}Sm/^{144}Nd$ ratios show no correlation, whereas the present day Indian Ocean MORB array, shown for comparison, displays a broad positive correlation. Coloured bars in the centre panel represent the uncertainty in $^{143}Nd/^{144}Nd_i$ for the two most extreme samples.

The plagioclase Nd data (n = 19) yield a mean square weighted deviation (MSWD = 2.0) that is significantly greater than the critical value (1.7; following Wendt and Carl, 1991; see Supplementary Information), meaning that the probability that the data form a single, normally distributed population is <5 %. We therefore argue that the observed $^{143}Nd/^{144}Nd_i$ heterogeneity in plagioclase is statistically significant and represents a primary feature. The clinopyroxene data (n = 22) show an MSWD that is smaller than the critical MSWD (1.1 *versus* 1.6, respectively), implying that the clinopyroxene data constitute a statistically homogeneous population at the current level of precision.

There is no discernible stratigraphic trend in plagioclase isotopic composition and the samples record a relatively large range in $^{147}Sm/^{144}Nd$ ratios (0.13-0.25) that does not correlate with $^{143}Nd/^{144}Nd_i$ (Fig. 3). With the exception of two samples, Nd and Sr isotope ratios in plagioclase crystals are closer to the MORB array than whole rock isotopic analyses (red field in Fig. 3). It is difficult to interpret the variability of strontium isotopes in plagioclase purely in terms of mantle source as seawater alteration is known to elevate $^{87}Sr/^{86}Sr$ without affecting $^{143}Nd/^{144}Nd$ (McCulloch *et al.*, 1980). The limited range observed in the micro-drilled $^{87}Sr/^{86}Sr$ compositions potentially records less extensive seawater alteration but we cannot disentangle minor alteration from source heterogeneity.

Discussion

Our data represent the first micro-drilled Nd isotope mineral analyses for lower ocean crustal rocks and provide constraints on both ocean crustal and mantle processes. Compared to previous whole rock analyses of plutonic rocks, single plagioclase crystals record a larger Nd isotope variability and are thus much more suitable for studying the full extent of isotopic variability in the lower oceanic crust. The similarity of the observed Nd isotope range in plagioclase to the gabbros from the Maqsad diapir (Fig. 4), taken to represent individual melt batches in upwelling mantle (Benoit *et al.*, 1996), suggests that the lower crust preserves the heterogeneity of melt batches at a mineral scale. This implies that the upper mantle is compositionally heterogeneous at the scale of melt extraction (*e.g.*, Lange *et al.*, 2013), and that the melt extraction process did not fully homogenise melts. The isotopic variability in whole rock analyses of the overlying dykes and lavas, representing fully aggregated and erupted melts, is limited compared to plagioclase in the lower crust (Fig. 4) and indicates that the substantial mixing and homogenisation required to produce MORB takes place in the crust. Previous comparisons of upper crustal basalts to whole rock analyses of plutonic rocks led to a substantially different conclusion as illustrated in Figure 4 (*e.g.*, Coogan, 2014). The whole rock data alone suggest the upper

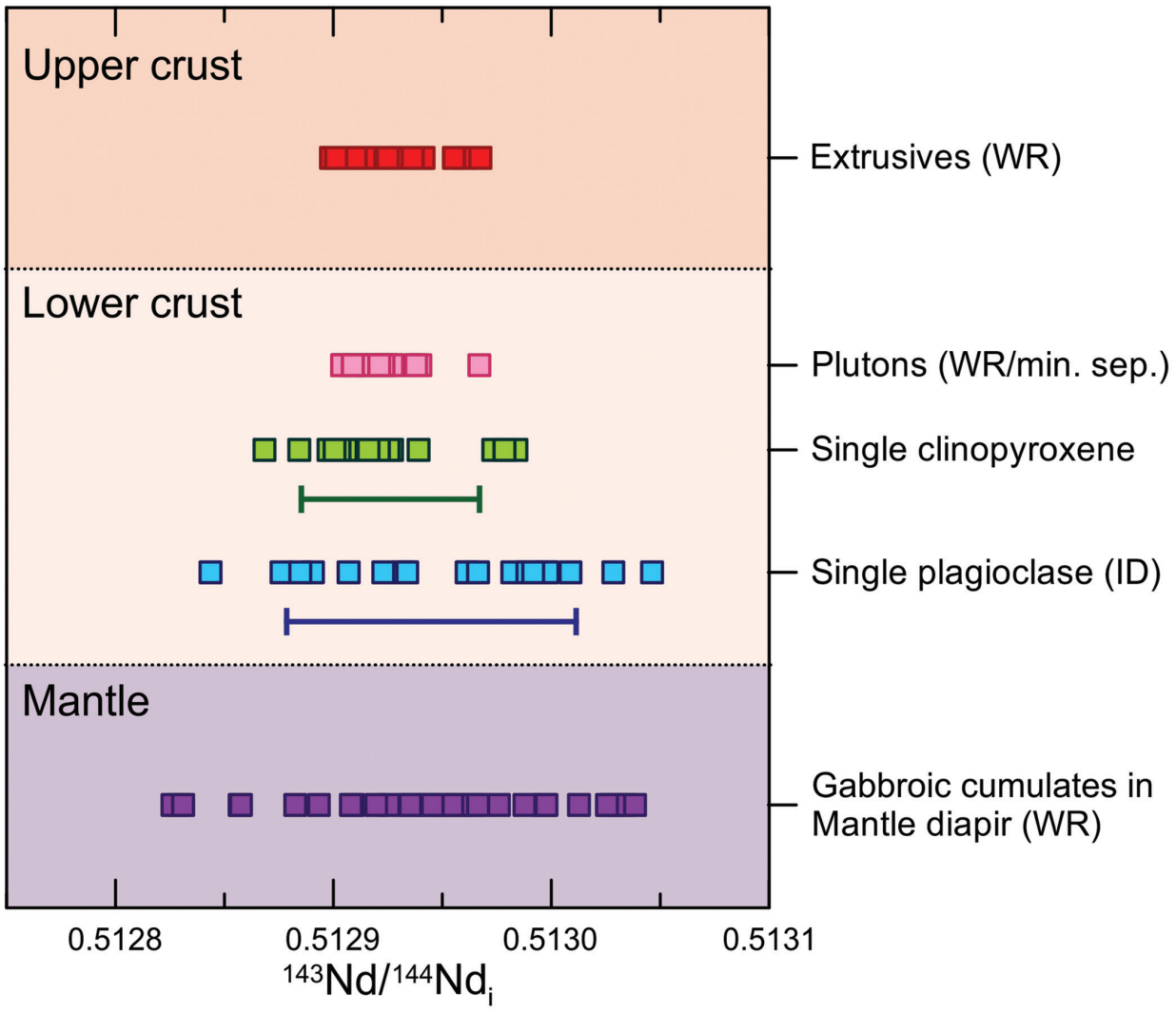

Figure 4 Comparison of the ranges in Nd isotopes found in this study with previous results of the Semail Ophiolite (McCulloch *et al.*, 1980; Benoit *et al.*, 1996; Godard *et al.*, 2006; Rioux *et al.*, 2012, 2013; de Graaff, 2016). Range for repeated analyses of BCR-2 reference material is indicated by the green (1.9-2.6 ng Nd) and blue (0.15-0.34 ng Nd) bars (displayed at an arbitrary position on the x-axis).

and lower crust formed from melts that were homogenised to similar extents. Our findings also contrast with Koga *et al*. (2001) who concluded that trace element variability in the Moho transition zone in the Semail Ophiolite is comparable to that in the upper crustal sheeted dyke complex and lavas and that mixing must have taken place predominantly in the mantle.

This study demonstrates that the approach of using mineral trace element ratios alone to study source compositions can be problematic. We find that plagioclase Sm/Nd does not correlate with $^{143}Nd/^{144}Nd_i$ (the time integrated Sm/Nd), whereas a positive correlation would be expected if Sm/Nd is inherited from a mantle source (Fig. 3). Several other processes can affect Sm/Nd, such as degree of mantle melting, fractional crystallisation, melt replenishment (O'Neill and Jenner, 2012), melt-rock reactions (Lissenberg and MacLeod, 2016) and variable partition coefficients due to changes in pressure, An %, pH_2O and melt composition (Bédard, 2006). Hence, it is difficult to interpret a source signature from plagioclase trace element compositions. Since Nd isotope ratios are not fractionated by these processes, they are uniquely suited for investigating sources of mantle derived melt.

The contrast in $^{143}Nd/^{144}Nd_i$ variability between clinopyroxene (homogeneous) and plagioclase (heterogeneous) can have multiple causes. The first is sub-solidus diffusion of REE. This unlikely to be responsible for homogenisation, however, since Nd diffusion coefficients of clinopyroxene are two orders of magnitude lower than plagioclase (van Orman *et al*., 2001; Cherniak, 2003) and modelling shows that diffusion would primarily affect plagioclase rather than clinopyroxene for conditions prevalent in the lower oceanic crust (Coogan and O'Hara, 2015).

The second is a crystallisation sequence of plagioclase before clinopyroxene during progressive magma mixing and associated homogenisation. Textures of Wadi Abyad gabbros are granular to intergranular, with an apparent olivine -> plagioclase -> clinopyroxene crystallisation sequence. The absence of troctolites in the section, however, may indicate a near simultaneous saturation of plagioclase and clinopyroxene (Thomas, 2003). Furthermore, moderate textural re-equilibration during slow cooling may obscure primary textures and make it difficult to distinguish co-crystallisation from sequential crystallisation (MacLeod and Yaouancq, 2000).

Finally, reactive porous flow of interstitial melts may be responsible for isotopic homogenisation in clinopyroxene. Migration of late stage melts by porous flow is pervasive throughout the entire lower crust (Lissenberg *et al*., 2013; Lissenberg and MacLeod, 2016). Clinopyroxene and plagioclase in gabbros at Wadi Abyad show features that have been ascribed to reactive porous flow, including an up-section increase in enrichment and fractionation of elements proportional to their incompatibility (*e.g.*, La/Nd and Ce/Y) and trace element disequilibrium between coexisting plagioclase and clinopyroxene (Lissenberg and MacLeod, 2016). If the migrating interstitial melts are isotopically homogeneous, as seems likely, melt-crystal reaction can conceivably homogenise isotopic variation in the lower crust. It is currently unclear, however, whether clinopyroxene is more sensitive to melt-rock reactions than plagioclase, as would be implied by the data: further work on this subject is required.

This work demonstrates that isotopic analysis of single grains in plutonic rocks has great potential to study the scale of mantle heterogeneity and the nature of crustal processes. By establishing that greater variability exists in the lower crust than in the erupted lavas of the Semail Ophiolite, we have shown that isotopic studies of whole rock MORB mask the full scale and extent of compositional heterogeneity of the mantle source. Micro-sampling successfully avoids interstitial grains and domains most affected by alteration (*i.e.* crystal rims) and, with the latest generation of sensitive amplifier technology, yields enough material to identify isotopic variability. Future research should employ these techniques to investigate further the presence of stratigraphic trends on various scales and isotopic disequilibrium between clinopyroxene and plagioclase. A better understanding of the variability of isotopic composition in the lower oceanic crust has implications for the location of melt emplacement and the location and extent of magma mixing at oceanic spreading centres, as well as the scale and magnitude of mantle heterogeneity.

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Additional Information

Supplementary Information accompanies this letter at http://www.geochemicalperspectivesletters.org/article1827.

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