

Neodymium isotope disequilibria in subducted sediments, and potential consequences for subduction-zone recycling

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ABSTRACT

Neodymium isotope geochemistry is pivotal to understanding magma genesis and crustal recycling processes at convergent plate margins. Here, we present Nd isotope compositions of rare earth element (REE)-rich minerals from blueschist- to eclogite-facies rocks that represent previously subducted continental sediments. We find Nd isotope heterogeneity in all of our samples. Apatite preserves diverse detrital isotopic compositions despite metamorphism up to at least 600 °C and 3.0 GPa, whereas lawsonite appears to be isotopically homogenous. Epidote and monazite are major REE hosts under eclogite-facies conditions; in our samples these phases also retain Nd isotope heterogeneity, even up to ultrahigh-pressure conditions of 700 °C and 4.0 GPa. These findings contest the assumption of Nd isotope equilibration during subduction-zone metamorphism, so caution is needed when using Nd isotopes to calculate subducted sediment contributions to arc magmas or for determining rates of crustal recycling through subduction zones.

INTRODUCTION

The Sm-Nd isotope system has been instrumental in advancing our understanding of convergent plate margin processes, including fingerprinting mantle and crustal contributions to arc magmas (e.g., Straub et al., 2010), deducing crustal recycling efficiencies (e.g., McCulloch and Perfit, 1981), and constraining the timing and duration of metamorphism in ancient convergence zones (e.g., Baxter and Scherer, 2013). Applications of Nd isotopes commonly include an implicit assumption that isotopic equilibrium is attained at the time of metamorphism or of partial melting and melt extraction. Nevertheless, there is evidence that isotopic heterogeneity can persist during metamorphism and partial melting of crustal rocks (e.g., Perini et al., 2009). Refractory rare earth element (REE) phases (e.g., apatite, monazite) included in, and shielded by, metamorphic minerals are recognized to disturb Sm-Nd isochron dating of metamorphism (Baxter and Scherer, 2013), and observations of Nd isotopic heterogeneity during anatexis have been attributed to variable dissolution of REE-rich accessory minerals during melting (Zeng et al., 2005; Perini et al., 2009). However, so far only Fisher et al. (2017) has directly measured mineral-scale Nd isotopic heterogeneity in anatectic granites.

Neodymium isotope disequilibria in metamorphic rocks is commonly regarded to be of pre-metamorphic origin, with sluggish reaction of Nd-rich minerals and slow Nd diffusion rates aiding retention of isotopic heterogeneity

through metamorphism (Zeng et al., 2005; Perini et al., 2009). Clastic sedimentary rocks in particular may contain detrital minerals of diverse origin and isotopic composition. If these detrital signatures survive metamorphism and partial-melting processes, then they may impart significant isotopic heterogeneity to mineral assemblages of metamorphic rocks or to melts produced by anatexis. Hammerli et al. (2014) showed that diverse Nd isotope signatures of detrital apatite in metasedimentary rocks can be retained under regional low-pressure metamorphism to temperatures above 500 °C, even under fluid-saturated conditions. However, the extent to which mineral-scale isotopic heterogeneity can be preserved under other conditions of metamorphism remains largely unknown.

Subduction of continental sediments is interpreted to play a crucial role in the generation of arc magmas and the chemical refertilization of the mantle (Plank and Langmuir, 1993; Spandler and Pirard, 2013). Neodymium isotopes are widely used to track the influence of sediment subduction (e.g., McCulloch and Perfit, 1981), yet little is known of how the Nd isotope system responds to progressive subduction-zone metamorphism. Here, we present Sm-Nd isotope compositions of REE-rich minerals from blueschist- to eclogite-facies rocks that collectively represent progressive subduction of sediments from shallow forearc depths (~20 km) down to subarc (~120 km) conditions (Fig. 1). We show that mineral-scale Nd isotopic heterogeneity may persist to great depths in subduction zones,

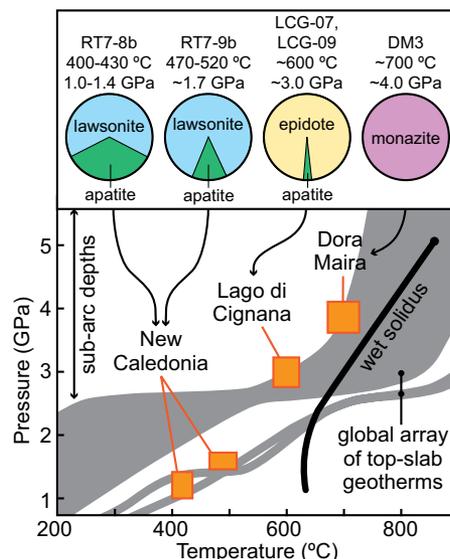


Figure 1. Top panel: Distribution of bulk-rock Nd between rare earth element (REE) minerals in studied samples. Bottom panel: Pressure-temperature conditions of peak metamorphism of studied samples compared with global array of top-slab geotherms (gray) and wet pelite solidus (black) (adapted from Spandler and Pirard, 2013). See Appendices DR1 and DR2 (see footnote 1) for estimates of metamorphic conditions for samples and Nd contents of minerals, respectively.

which may complicate the application of Nd isotopes to tracking element transfer and crustal recycling rate at convergent plate margins.

SAMPLES

During subduction, crustal rocks undergo progressive metamorphism from blueschist-facies conditions up to ultrahigh-pressure (UHP) eclogite-facies conditions, as well as significant devolatilization (Fig. 1). To understand Nd isotope evolution through this process, we examined five metamorphic rock samples, representative of subducted continental sediment, from three well-studied high-pressure (HP)–UHP metamorphic belts. Descriptions of the geology of these samples and the metamorphic belts are presented in Appendix DR1 in the GSA Data

Repository¹, so only a brief geological outline is presented here. Bulk-rock major and trace element and Sm-Nd isotope compositions of the samples are presented in Appendix DR2.

The lowest-grade samples come from the Diahot blueschist belt of New Caledonia (South Pacific), which represents a clastic metasedimentary sequence that was subducted to forearc depths (~15–50 km) in the mid- to late Eocene (Pirard and Spandler, 2017). Sample RT7-8b is a fine-grained, crenulated, quartz-mica-chlorite schist with blueschist-facies indicator minerals lawsonite and glaucophane. Apatite, titanite, and zircon are accessory phases. Peak metamorphic conditions are ~400 °C and between 1.0 and 1.4 GPa (Fig. 1; Appendix DR1). Sample RT7-9b is a carbonaceous schist consisting mostly of quartz, muscovite, and glaucophane, with minor lawsonite, titanite, apatite, graphite, and trace zircon. Estimated peak metamorphic conditions for this sample are between 470 and 520 °C and ~1.7 GPa (Fig. 1; Appendix DR1). Both samples are of pelitic composition (Appendix DR2) with original sedimentary detritus derived from diverse sources, as revealed by the large range of ages and Hf isotope compositions of their detrital zircons (Pirard and Spandler, 2017). The major Nd-hosting minerals in both cases are lawsonite and apatite, although apatite only hosts ~14% of Nd in sample RT7-9b (Fig. 1).

Ophiolitic rocks exposed at Lago di Cignana, Italian western Alps, reached UHP conditions of ~3.0 GPa and ~600 °C (Groppo et al., 2009). The sequence includes metasedimentary rocks derived from diverse continental sources based on radiogenic isotope data (Mahlen et al., 2005) and detrital zircons of variable age (Rubatto et al., 1998). Two samples are examined here. Sample LCG-07 is an eclogitic gneiss comprising quartz, omphacite, garnet, Na-Ca amphibole, white mica, and minor apatite, REE-rich epidote, rutile, and calcite. This assemblage is partly overprinted by retrograde actinolite, albite, REE-poor epidote, and titanite (Appendix DR1). Sample LCG-09 is a quartz-garnet-carbonate-white mica schist, with minor titanite, apatite, and epidote. The primary host of Nd in these rocks is REE-rich epidote, with minor amounts in apatite (Fig. 1; Appendix DR2). Both phases belong to the peak metamorphic mineral assemblage.

The final sample, DM3, is from the UHP Brossasco-Isasca unit of the Dora Maira massif, Italian western Alps (Compagnoni et al., 1994). Sample DM3 is a coarse-grained quartz-phengite-garnet schist with minor kyanite and

accessory rutile, zircon, and monazite. This pelitic metasedimentary rock reached peak metamorphic conditions of ~4.0 GPa and ~700 °C, based on textural relicts of coesite in garnet porphyroblasts and on rutile and phengite compositions (Appendix DR1). Neodymium is almost entirely hosted by monazite that occurs as clusters of fine (<100 μm) grains within phengite (Appendix DR1).

Nd ISOTOPE COMPOSITION OF REE-RICH MINERALS

The Nd isotope composition of major light REE (LREE)-rich mineral phases in each sample were determined by laser ablation–multicollector–inductively coupled plasma–mass

spectrometry (LA-MC-ICP-MS). The analytical methods and results are presented in Appendix DR3 and in Figures 2 and 3. Initial ϵ_{Nd} values are calculated to the time of peak metamorphism.

Lawsonite from blueschist samples RT7-8b and RT7-9b has a fairly uniform isotopic composition of $\epsilon_{Nd} \sim 1$, which is similar to the bulk-rock isotope composition of RT7-8b (Figs. 2A and 2B). By contrast, ϵ_{Nd} of apatite from these samples spans a large range, from +6 to as unradiogenic as –21.

Apatite from eclogitic sample LCG-07 also spans a large range of ϵ_{Nd} from ~0 to –12 (Fig. 2C). ϵ_{Nd} of REE-rich epidote in this sample varies from ~–10 to >0. Few analyses overlap with the bulk-rock ϵ_{Nd} value of –5.4. Apatite and

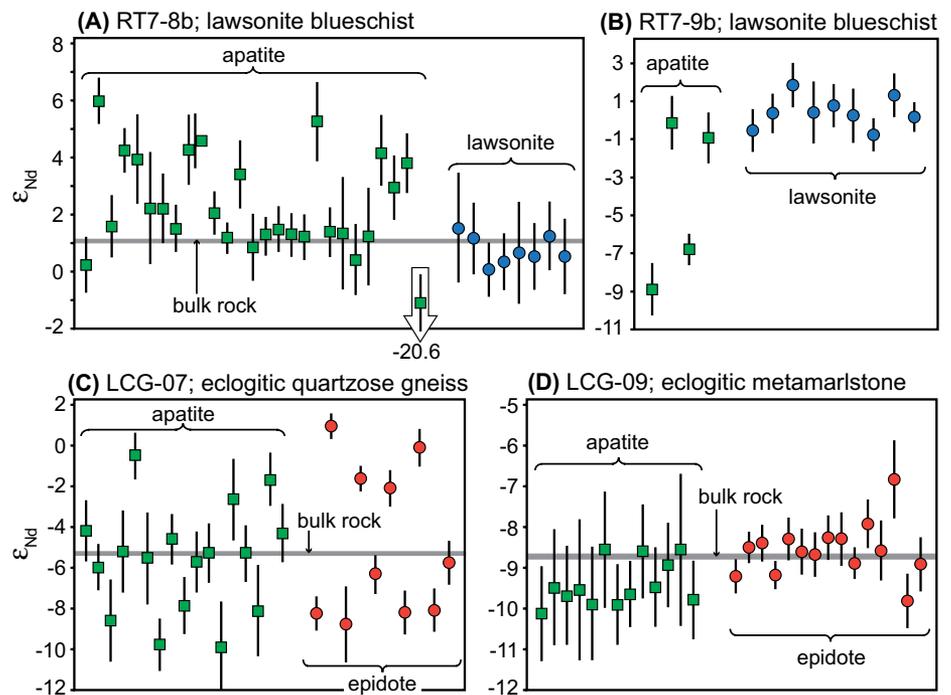


Figure 2. Rare earth element (REE) mineral and bulk-rock ϵ_{Nd} ($t = 42$ Ma) values for blueschist- and eclogite-facies samples from New Caledonia (South Pacific) (A, B) and Lago di Cignana (Italy) (C, D). Note that no bulk-rock isotopic value is available for sample RT7-9b.

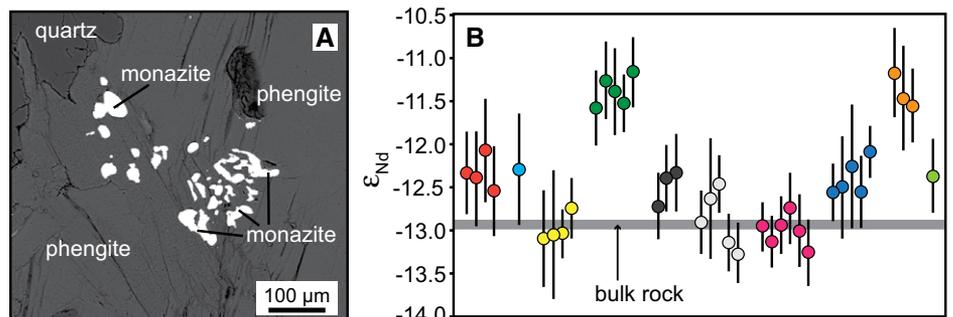


Figure 3. A: Backscatter electron image of monazite grain cluster within phengite from sample DM3. B: Monazite and bulk-rock ϵ_{Nd} ($t = 35$ Ma) for sample DM3. Each point represents a single spot analysis, and each color group corresponds to an individual grain cluster. Note that grains within each cluster have similar ϵ_{Nd} values, whereas there is significant ϵ_{Nd} variation between clusters.

¹GSA Data Repository item 2018303, geological setting and description of samples, analytical techniques, bulk-rock geochemistry and Sm-Nd isotope compositions, and mineral Sm-Nd isotope compositions, is available online at <http://www.geosociety.org/datarepository/2018/> or on request from editing@geosociety.org.

epidote in metamarlstone sample LCG-09 are much more homogenous, with most analyses within uncertainty of the bulk-rock ϵ_{Nd} value of -8.7 (Fig. 2D). A couple of epidote grains, however, have ϵ_{Nd} values outside the range of values for the bulk rock and most other epidotes.

Monazite in sample DM3 occurs as clusters of fine grains included in phengite (Fig. 3A). ϵ_{Nd} values of monazite within each cluster are very similar; however, the values of individual grain clusters vary from -13.0 ± 0.34 (2 standard deviations, SD) up to -11.4 ± 0.36 (2 SD) (Fig. 3B). Clusters with different isotopic composition can be as little as 10 mm apart in the sample.

DISCUSSION

Preservation of Isotopic Heterogeneity during Metamorphism

It is expected that most of the LREEs in crustal rocks are redistributed from lawsonite (\pm apatite) to epidote or monazite during transition from low-grade blueschist-facies conditions to eclogite-facies conditions (Spandler et al., 2003). The samples examined here span this metamorphic transition (Fig. 1) and hence are used to assess Sm-Nd isotope equilibration in sedimentary protoliths during HP and UHP metamorphism. We find statistically significant Nd isotopic heterogeneity between LREE-rich minerals in all of our samples (Figs. 2 and 3). The heterogeneity is not attributed to post-metamorphic processes, as metamorphism is relatively young (Eocene) in all cases, and there is little evidence of retrograde alteration of LREE-rich minerals in the samples (Appendix DR1). There is also no indication that bulk REE contents of the samples were significantly modified by fluids during metamorphism (Appendix DR1; Spandler et al., 2003; Mahlen et al., 2005). Instead, the isotopic heterogeneity is considered primarily to be of pre-metamorphic origin. Apatite in the blueschists and in eclogite sample LCG-07 shows the greatest isotopic diversity, which we consider to be most likely due to preservation of the detrital (i.e., pre-subduction) isotopic signature of individual grains. Although we do not have direct age control on individual REE minerals, a detrital origin would be consistent with textural features of apatite grains in the samples (Appendix DR1) and U-Pb and Lu-Hf isotopic variability of zircons from these rocks (Rubatto et al., 1998; Pirard and Spandler, 2017). The range of isotopic values likely reflects derivation of the original sediments from ancient continental materials as well as from relatively juvenile volcanism (Mahlen et al., 2005; Pirard and Spandler, 2017). Hammerli et al. (2014) also interpreted Nd isotope heterogeneity of apatite from low-pressure metasedimentary rocks of the Mount Lofty Ranges, South Australia, to be due to preservation of detrital Nd isotope signatures up to metamorphic temperatures of 550°C . Our

results would suggest that detrital signatures in apatite can survive to higher temperatures—at least 600°C —under HP to UHP metamorphic conditions. Taken together, the results testify to the resistance of detrital apatite to re-equilibration during regional metamorphism.

In contrast to apatite, lawsonite in our samples appears to be largely isotopically homogenous and similar in isotopic composition to the bulk rock (Fig. 2). This is perhaps unsurprising, as lawsonite is entirely a metamorphic phase. On the other hand, we observe significant isotopic variation in epidote, particularly in sample LCG-07. Unlike lawsonite, epidote is formed in many geological environments, so the observed isotopic heterogeneity of epidote may reflect preservation of detrital REE-rich epidote grains that formed cores for subsequent overgrowth of metamorphic REE-poor epidote, a premise consistent with the common occurrence of detrital epidote in clastic sedimentary rocks (Spiegel et al., 2002). Alternatively, if epidote is of metamorphic origin (Janots et al., 2008), it may have inherited the variable Nd isotope composition of detrital apatite, which would require chemical equilibration during epidote growth to be limited to the sub-grain ($<100\ \mu\text{m}$) scale. Romer and Xiao (2005) proposed a similar process to explain grain-scale isotopic heterogeneity in epidote from the Sulu UHP belt (eastern China).

Our results indicate that there was a greater degree of Nd isotope homogenization between REE phases during metamorphism of metamarlstone sample LCG-09 (Fig. 2D) compared to LCG-07. The relative homogeneity of LCG-09 may reflect original sediment accumulation from restricted source regions with limited isotopic variability, or that LCG-09 may have undergone more extensive recrystallization (and hence, Nd isotope homogenization) due to the high reactivity of C-O-H fluids produced locally during prograde metamorphism (Frezza et al., 2011).

The monazite clusters in metapelite sample DM3 are contained within unaltered phengite of UHP composition (Appendix DR1), and so were not affected by retrograde metamorphism. We also discount a detrital origin, as detrital monazite in pelitic sediments would not be expected to survive eclogite-facies metamorphism (Janots et al., 2008). We therefore interpret the isotopic variation between monazite grain clusters (Fig. 3B) to be inherited from precursor (possibly detrital) apatite or REE-rich epidote, which reacted to form monazite during prograde metamorphism. This hypothesis is consistent with the suggestion of Luais et al. (2001) that Nd isotope disequilibria in eclogite-facies samples from the western Alps are due to inclusion of old, detrital REE minerals in garnet.

Overall, the results demonstrate that isotopic disequilibrium in REE minerals can be retained at the sub-centimeter scale even to metamorphic conditions of $4.0\ \text{GPa}$ and 700°C . This

scale of heterogeneity requires limited isotopic exchange between REE minerals during metamorphism and fluid release, which is consistent with the relatively refractory nature of many REE-rich minerals and the slow rates of Nd diffusion under most metamorphic conditions (Zeng et al., 2005; Perini et al., 2009).

More broadly, our results provide empirical evidence that the Nd isotope disequilibria observed in garnet-based Sm-Nd isotope geochronology (e.g., Baxter and Scherer, 2013) and in some cases of crustal melting (Zeng et al., 2005; Perini et al., 2009; Fisher et al., 2017) can be due to isotopic heterogeneity among metamorphic mineral assemblages. We note that metasedimentary rocks of siliciclastic origin have the greatest potential to preserve isotopic heterogeneity because they derive from detrital mineral populations with inherent isotopic variability.

Implications for Crustal Recycling in Subduction Zones

Element transfer from the subducting slab to the mantle wedge is generally regarded to be due to upward flow of hydrous fluids and/or melts produced as a consequence of metamorphic dehydration of the downgoing slab (Spandler and Pirard, 2013). Sedimentary components of the slab are thought to supply many key trace elements to arc magmas, including the LREEs that are used to calculate slab temperatures (Plank et al., 2009) and sediment recycling efficiencies via Nd isotopes (e.g., McCulloch and Perfit, 1981). Recent studies demonstrate that slab fluids can transport significant quantities of LREEs (e.g., Tsay et al., 2017), although fluid migration in the slab is likely to be highly channelized (i.e., non-pervasive), even at the micrometer scale (e.g., Plümper et al., 2017). Our results show that Nd isotope heterogeneity of LREE-rich minerals in subducted sediments can survive major metamorphic dehydration reactions and can persist to sub-arc conditions (Fig. 1). Therefore, it is conceivable that fractionation of the bulk-rock Nd isotope composition will take place during fluid release from the slab if fluid reaction favors an isotopically distinct population of REE minerals. This may occur via preferential dissolution of one LREE-rich mineral over another (e.g., apatite versus monazite; see Zeng et al., 2005) or if isotopically distinct LREE phases are isolated from fluid reactions due to inclusion in minerals such as garnet (e.g., Luais et al., 2001). These situations are more likely to occur in relatively cold subduction zones, where low temperatures favor persistence of isotopic heterogeneity in the slab and limit fluid-rock equilibration.

If fluids liberated from the slab can have different Nd isotopic compositions than their source rocks, then the Nd isotope composition of arc magmas may not be the robust measure of slab contributions to the magma source as is

currently regarded. To demonstrate this point, we examine the simple Nd isotope mixing model used by McCulloch and Perfit (1981) to calculate that Aleutian arc lavas contain between 2% and 8% of a slab sediment component. If we assume that the Nd isotope composition of the slab fluid was fractionated by 5 ϵ units (i.e., much less than the isotopic variability observed in sample LCG-07; Fig. 2) to more radiogenic values compared to the source, the apparent slab sediment component doubles to between 3% and 16%. These calculations are purely for demonstration purposes and are not proposed to be directly relevant to Aleutian lavas; nevertheless, Nd isotope fractionation during slab processing may explain more subtle variability in the Nd isotope composition of arc lavas (or decoupling of Nd isotopes from other slab tracers) observed from within individual arc segments, or even from individual volcanoes (e.g., Straub et al., 2010).

Undoubtedly, Nd isotopes remain a very powerful tracer of source compositions, and the potential complications outlined here may be assessed through collective analysis of multiple isotopic and trace element data sets. For this purpose, interpreting Nd isotopes in tandem with an isotope tracer that readily reacts during metamorphism, such as Li, may be particularly beneficial.

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