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Nitrogen (N) and its isotopes in serpentinized forearc wedges and implications for N cycling across subduction zones

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ABSTRACT

We investigated nitrogen (N) sources and incorporation in the shallow forearc of an active subduction zone by studying samples from two serpentinite seamounts (South Chamorro and Conical; Ocean Drilling Program Legs 195 and 125) along the Mariana convergent margin. We report N concentrations and $\delta^{15}N$ values for serpentinized peridotites, serpentinite muds, and metabasaltic clasts. All are enriched in N relative to likely ultramafic and mafic protoliths. A modest positive correlation between N and $\delta^{15}N$ for the serpentinized peridotites could reflect extents of N infiltration by N-bearing fluids. Mixing calculations identify the source of the fluids as dehydrating metasediments and/or altered oceanic crust from the subducting Pacific slab. Such addition is consistent with that of other fluid mobile elements (B, Li, As, and Cs), but their concentrations show little correlation with N. The general lack of correlation in concentrations of N and major and trace elements complicates the identification of mineral hosts and N residency (e.g., loose adsorption on serpentine minerals). The enrichment of N in these samples indicates that serpentinized mantle wedges could be a globally significant N reservoir. If subducted, this reservoir could provide significant amounts of isotopically heavy N to sub-arc depths and beyond.

INTRODUCTION

Forearc mantle wedges can be pervasively hydrated by infiltrating hydrous fluids released by dehydration of subducting slabs (Peacock and Hyndman, 1999; Wang et al., 2009). Large ion lithophile elements (LILEs) and other fluid mobile elements (FMEs), with isotopic signatures distinctive of subducting slabs, are conveyed into these ultramafic rocks, yielding a reservoir with significance to models of global geochemical evolution (see Savov et al., 2007; Pagé et al., 2018).

Nitrogen is highly enriched in seafloor sediments (e.g., Ocean Drilling Program [ODP] Leg 129 drilling at the Mariana margin; N mean [1σ] = 117 ± 189 ppm; $\delta^{15}N_{air}$ = +2.8‰ ± 2.6‰; Sadofsky and Bebout, 2004) relative to the depleted mantle (N = 0.04–2 ppm; $\delta^{15}N$ = −5‰ ± 2‰; Marty and Dauphas, 2003) and other slab lithologies (N in altered oceanic crust [AOC] from Leg 129 mean [1σ] = 6 ± 5 ppm; $\delta^{15}N$ = −3.5‰ ± 3.8‰; N in serpentinized slab mantle mean [1σ] = 8 ± 4 ppm; $\delta^{15}N$ = +1.3‰ ± 2.7‰; Li et al., 2007; Li et al., 2023). Despite containing less N than sediments, AOC and serpentinized slab mantle are significant N reservoirs due to their volume (Li et al., 2007; Li et al., 2023). The extent of N release from slabs and sediments at forearc or greater depths is likely related to the thermal structure of the subduction zone (see Bebout et al., 2016).

In sediments, N primarily resides as NH_4^+ in the crystal lattice of K-bearing minerals such as clays and micas via K-NH_4^+ substitution (e.g., Honma and Itihara, 1981). In AOC, NH_4^+ may substitute for Na^+ and/or Ca^{2+} in amphiboles, pyroxenes, or feldspars (Honma and Itihara, 1981; Li et al., 2007; Watenphul et al., 2010; Busigny et al., 2018). The N host in serpentinites is less clear than in sediments, with studies of oceanic modern seafloor and low- to high-grade serpentinites suggesting various possibilities for N residency without a K-rich phase. These include sealed voids, fluid inclusions, clinopyroxene, intergranular N_2, or NH_4^+ fixed in serpentine minerals via adsorption (Philippot et al., 2007; Halama et al., 2014; Pagé et al., 2018; Cannaò et al., 2020; Li et al., 2023).

Li et al. (2023) proposed that oceanic serpentinites could be a critical N reservoir, suggesting substantial N retention to great depths in subduction zones based on the overlap in N concentrations with high-grade serpentinites (cf. Halama et al., 2014). If deeply subducted (e.g., by subduction erosion), serpentinized ultramafic rocks in forearc wedges could convey N with $\delta^{15}N$ higher than "mantle values" ($\delta^{15}N$ ∼−5‰) to sub-arc depths or beyond, depending on the degree of retention (Halama et al., 2014; Pagé et al., 2018; Cannaò et al., 2020).

In this study, we examined whether forearc serpentinites are enriched in N via interaction with slab-derived fluids, relative to their protoliths, by analyzing serpentinized peridotites and serpentinite muds from South Chamorro and Conical Seamounts, located between the Mariana trench and its volcanic arc front (Fig. 1; see Savov et al., 2005). Also, we report N concentrations and $\delta^{15}N$ for four metabasaltic clasts from these seamounts, affording a first direct insight into AOC metamorphosed and brought via mud volcanism to the ocean floor in an active subduction zone. Their compositions can be compared with those of metabasalts in exhumed high- and ultrahigh-pressure metamorphic terranes (see the compilation in Mallik et al., 2023).

DESCRIPTION OF SAMPLES AND ANALYTICAL RESULTS

Serpentinized peridotites (*n* = 28), serpentinite muds (*n* = 4), and metabasaltic clasts (*n* = 4) were recovered from ODP Legs 195 (Holes 1200A, E, D, and F) and 125 (Holes 779A and 780C) at South Chamorro and Conical Seamounts below the seawater infiltration zone (Fig. 1; Savov et al., 2005). These sites are ∼1000 km apart and at depths of 18–19 km

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above the subducting Pacific slab, 80 km west of the trench axis (Fryer et al., 2018).

Protoliths for the serpentinized peridotites were highly melt-depleted mantle wedge harzburgites and dunites, now extensively serpentinized with common brucite, lizardite, chrysotile, and rare antigorite (indicating low-temperature alteration <250 °C; Savov et al., 2005, 2007). Loss on ignition (LOI) values are typically ~13% without elevated CaO and lowered MgO, consistent with the presence of brucite rather than Ca carbonate. The muds consist primarily of serpentine ± brucite, chlorite, amphiboles, phengite, spinel, carbonates, pyroxene, talc, and epidote and reflect mechanical mixture of serpentinites and as much as 5% metabasaltic clasts (Savov et al., 2007). Savov et al. (2005) suggested that the pumpellyite to greenschist- to blueschist-grade metamorphic clasts (metamorphic ages near 50 Ma; Maekawa et al., 2001; Tamblyn et al., 2019) are metabasalts and metagabbros from the Pacific subducting slab (see the Supplemental Material[1] Fig. S1). Detailed geological and petrologic information for the samples is provided by Savov et al. (2005, 2007, and references therein).

Nitrogen concentrations and isotope compositions were analyzed at Lehigh University (Bethlehem, Pennsylvania, USA) using the carrier gas methods described in Supplemental Material Table S1. Serpentinized peridotites and muds contain 0.8–68 ppm N (mean = 24 ± 20, 1σ), with $\delta^{15}N_{air}$ ranging from −5.2‰ to +5.1‰ (mean = −0.2 ± 2.7‰, 1σ). The metabasaltic clasts contain 6–29 ppm N (mean = 12 ± 11, 1σ) with $\delta^{15}N$ ranging from −3.5‰ to +11.6‰ (mean = +4.6‰ ± 6.7‰, 1σ). Notably, one metabasaltic clast exhibits the highest $\delta^{15}N$ reported thus far for AOC (sample 2H-2W, $\delta^{15}N$ = +11.6‰; see Table S1; Fig. 2). Most serpentinized ultramafic samples analyzed in this study have N concentrations higher than that of the depleted mantle (N ~1.5 ppm; Fig. 2). Samples from Conical Seamount have higher N concentrations than those from South Chamorro Seamount but similar $\delta^{15}N$.

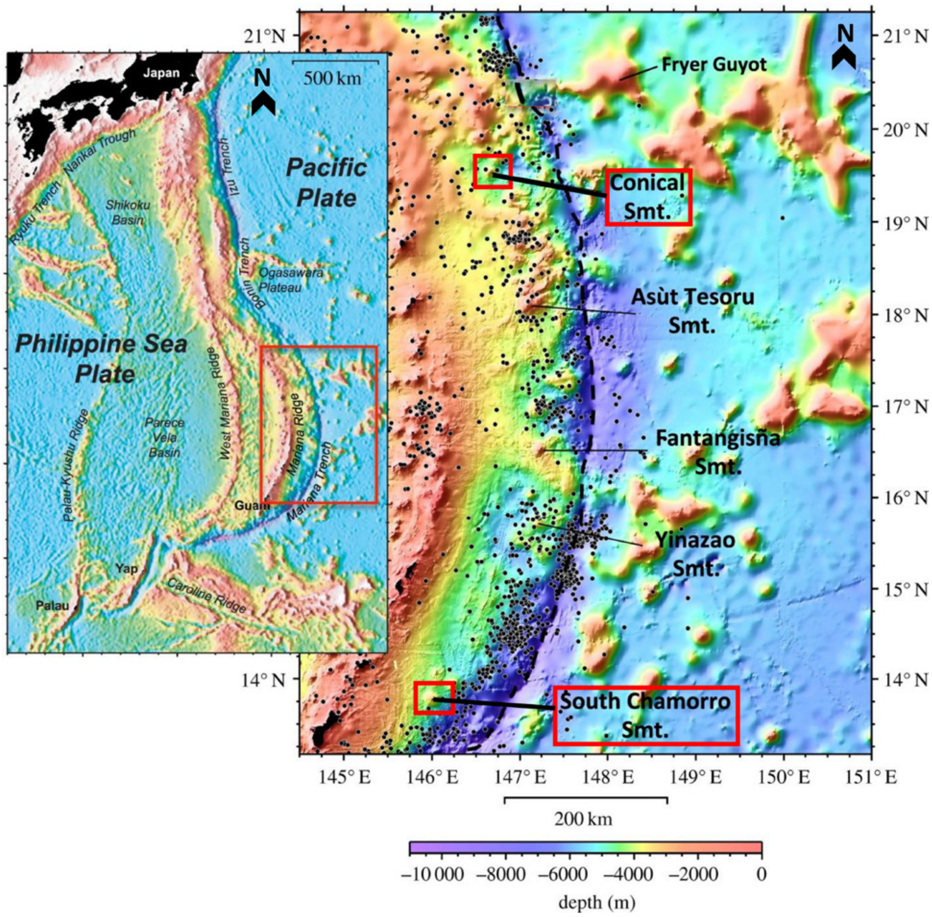

Figure 1. Bathymetric regional (inset left) and local map (right) of Mariana forearc region, highlighting study locations: South Chamorro Seamount (Ocean Drilling Program [ODP] Leg 195, Site 1200) and Conical Seamount (ODP Leg 125, Sites 778–780). Black dots are earthquake positions (0–50 km depths; 1900–2017; NEIC database). Modified after Fryer et al. (2020).

DISCUSSION

Fluid Sources and Nitrogen Enrichment

Geochemical features of serpentinized peridotites, such as major and trace element concentrations, enrichments in FMEs, and pore water compositions from the seamounts, suggest infiltration by N-bearing slab-derived fluids (Mottl, 1992; Savov et al., 2005, 2007). Relative to unsubducted protoliths, N is dramatically enriched in serpentinized peridotites, particularly in Conical Seamount samples, by more than an order of magnitude (see Fig. 3); these samples also show extreme enrichment of FMEs such as B, Li, As, Sb, and Cs (Savov et al., 2007). No correlation was found between N and LOI, suggesting that N enrichments are not related to extents of hydration and/or alteration (Philippot et al., 2007; Halama et al., 2014). However, all samples have abundant hydrous mineralogy and high LOI, indicating the possibility that infiltrating H_2O-rich slab-derived fluids varied in their N concentrations, perhaps related to differing sources (i.e., differing sediment-AOC proportions) or modification of fluid compositions along complex flow pathways.

Additions of N from metasedimentary sources were inferred for low- to high-grade-metamorphosed serpentinites from Monte Nero (Italy), Erro-Tobbio (Italy), and Cerro de Almirez (Spain), serpentinized metaperidotites from Gagnone (Swiss Alps), and wedge serpentinites from Himalayas subducted to >100 km depths (see Fig. 2; Halama et al., 2014; Pagé et al., 2018; Cannaò et al., 2020). For the Mariana serpentinites studied here, calculated mixing of N with an initial depleted mantle average composition (N ~1.5 ppm; $\delta^{15}N$ = −5‰) indicates N addition via fluids from dehydrating metasediments and/or AOC (Figs. 2 and 3; Supplemental Material Text S3). The $\delta^{15}N$ range of the N-enriched Mariana samples could reflect mixing of N from the two sources (sediment and AOC), presumably in fluids but possibly in a hybridized source (e.g., a mélange zone), differing extents of interaction with fluids derived from devolatilizing sources, and/or fluid-rock isotopic fractionation during serpentinization reactions. A seawater source for the N addition in the samples studied here can be ruled out via several lines of evidence, including shallow seawater penetration (<5 meters below sea floor [mbsf]) based on pore water geochemistry, low $^{87}Sr/^{86}Sr$ of the serpentinites (<0.705) compared to seawater (0.709), absence of aragonite crystal precipitation in the serpentinite muds below 5 mbsf (produced via interaction of high-alkalinity and low-Ca deep fluids with low-alkalinity

[1]Supplemental Material. Photomicrographs of sample 1H-5W (18-20) from South Chamorro Site 1200F (Fig. S1), analytical techniques (Fig. S2), mixing model (Fig. S3), calculation of global N reservoir in serpentinized forearc wedges (Fig. S4), and whole-rock N concentration and isotopic data (Table S1). Please visit https://doi.org/10.1130/GEOL.S.27305844 to access the supplemental material; contact editing@geosociety.org with any questions.

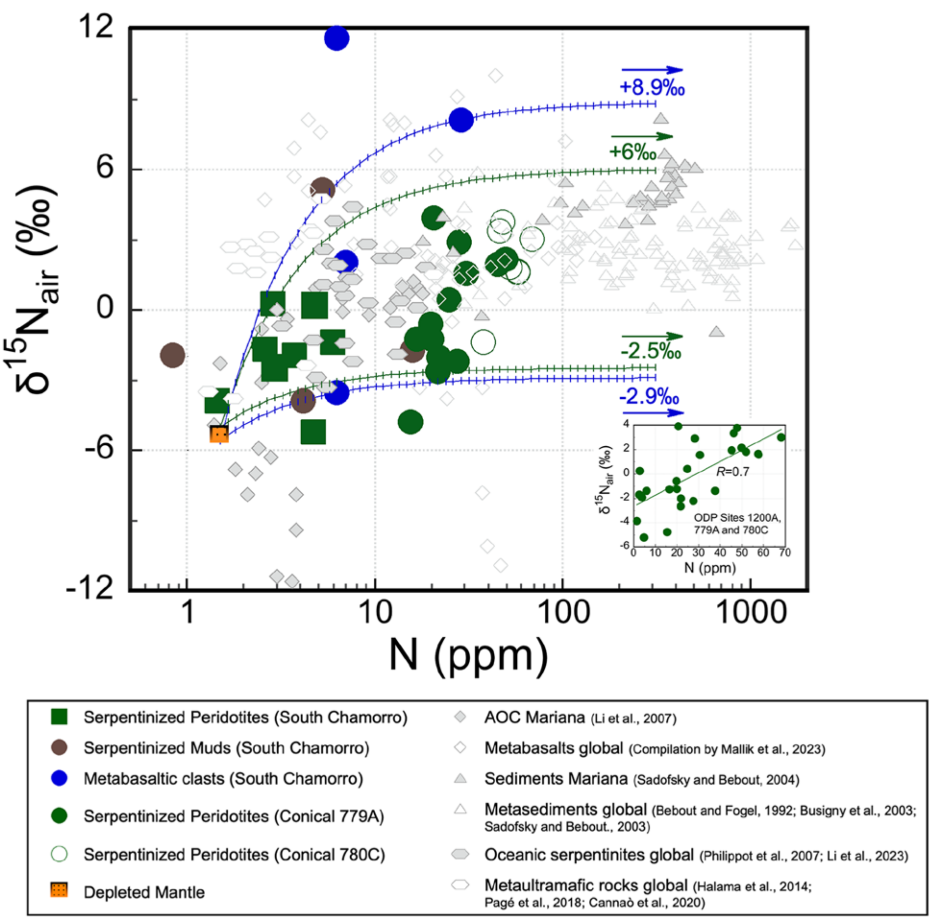

Figure 2. Nitrogen (N) concentrations versus δ^{15}N for samples analyzed in this study. Blue and green lines show results of the mixing model of N addition to initially N-depleted mantle (see text and Supplemental Material Text S3 [text footnote 1]). Lines are the upper and lower limit of the calculated N and δ^{15}N for the metabasaltic clasts (blue) and the serpentinized peridotites (green). Vertical ticks mark 10% mixing increments extending toward the arrows. Relevant lithologies are plotted in gray for comparison (see legend for details and sources). Inset plot shows N and δ^{15}N correlation for serpentinized peridotites analyzed here. ODP—Ocean Drilling Program; AOC—altered oceanic crust.

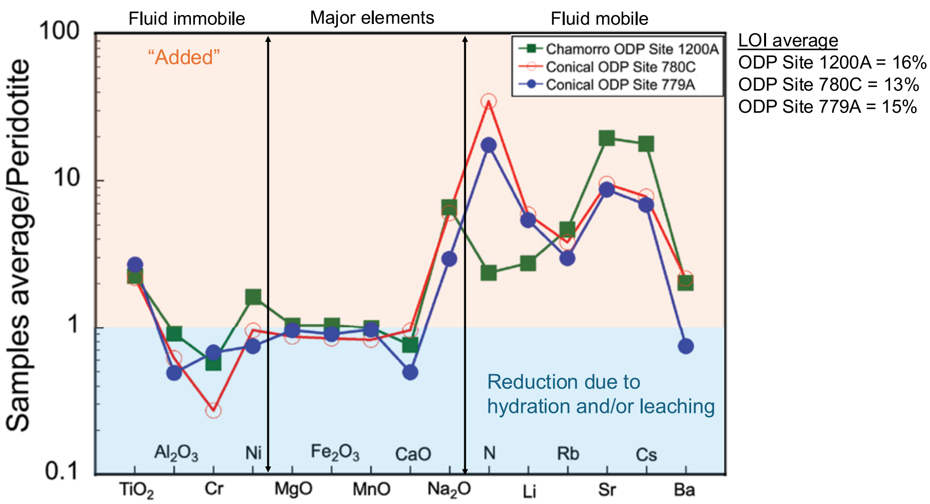

Figure 3. Diagram comparing averaged concentrations of selected fluid immobile, major, and fluid mobile elements of interest for serpentinized peridotites normalized to concentrations for non-serpentinized peridotite (Ohara and Ishii, 1998; Rizeli et al., 2016). Elements in upper orange field are "added" during hydration, and in lower blue field, reduced and/or leached. ODP—Ocean Drilling Program; LOI—loss on ignition.

and high-Ca seawater), and B and Li systematics pointing to non-seawater-related processes (Mottl, 1992; Savov et al., 2005, 2007). For comparison, unsubducted oceanic serpentinites also show no correlation between N and δ^{15}N, reflecting interaction with seawater at various depths (Li et al., 2023).

Negligible K_2O concentrations in the serpentinites and muds studied here and lack of correlation with N argue against K-bearing minerals as N hosts, unlike in metasedimentary rocks (e.g., Busigny and Bebout, 2013). More broadly, the lack of correlations between N and major or trace elements complicates identifying N hosts. This has been observed for other serpentinites including those from subduction-zone metamorphic terranes (e.g., Halama et al., 2014), leading previous workers to propose serpentine minerals as N hosts via adsorption rather than cation substitutions in their crystal lattices (see the high N contents in serpentine veins in Li et al., 2023). Lafay et al. (2016) documented Cs^+ adsorption onto serpentine minerals, and Cs^+ and NH_4^+ have similar ionic radii and show general similarity in geochemical behavior. Yu et al. (2023) documented NH_4^+ adsorption onto chlorite, a phase present in the Mariana mud samples. Harris et al. (2022), in an ion microprobe study, found significant N concentrations (as high as 83 ppm) in chlorite and suggested NH_4^+ incorporation during crystal growth into vacant interlayer sites rather than adsorption onto crystal edges and surfaces. Adsorption or structural incorporation of NH_4^+ into silicates with little or no K^+ (e.g., some clays, chlorite, and possibly serpentine and brucite) and its release or retention during prograde metamorphism could significantly influence N cycling to greater depths in subduction zones (cf. Halama et al., 2014; Li et al., 2023).

Variations in N Enrichment at South Chamorro and Conical Seamounts

The N enrichment in serpentinites at South Chamorro and Conical Seamounts differs considerably, with Conical showing higher concentrations than South Chamorro (Figs. 2 and 3). This could reflect differences in pore water ammonia concentration (slightly lower at South Chamorro; Mottl, 1992), contrasts between summit and flank holes (related to fluid flow paths), and depth to the slab (slightly lower at South Chamorro than at Conical), thus differing degrees of slab metamorphism. Mineralogy and trace element compositions of samples from South Chamorro and Conical Seamounts are indistinguishable (Savov et al., 2005), with no evidence suggesting differences in slab and sediment compositions or proportions below the two. One likely explanation is heterogeneity due to differing fluid flow paths and related degrees of fluid-rock interaction at greater depths.

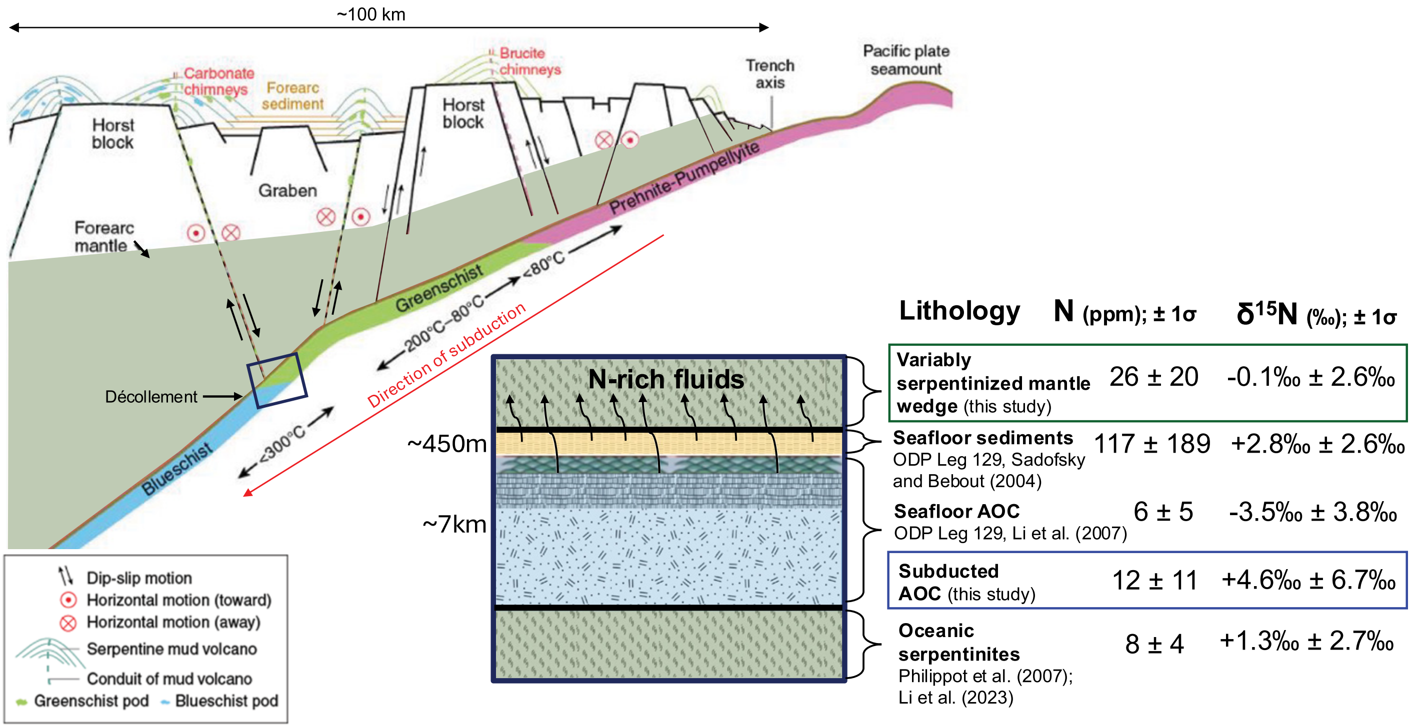

Figure 4. Schematic representation of Mariana forearc and nitrogen (N) systematics highlighting N and $\delta^{15}N$ of serpentinized peridotite and subducted altered oceanic crust (AOC) samples analyzed in this study. Data for relevant lithologies shown for comparison. Depth from seamounts to square marked in slab is ~19 km, corresponding to depth to slab for South Chamorro and Conical Seamounts. Modified from Fryer et al. (2018). ODP—Ocean Drilling Program.

Metabasaltic Clasts and Their Nitrogen Signature

The metabasaltic clasts contain a wide range of N and $\delta^{15}N$ as do metabasaltic rocks from exposed metamorphic suites (see the compilation in Mallik et al., 2023), overlapping with the Mariana AOC N concentrations drilled on Leg 129 outboard of the Mariana trench (Li et al., 2007; see Figs. 2 and 4). The N concentrations and $\delta^{15}N$ values higher than those of the incoming AOC section at ODP Site 801 (Figs. 2 and 4) perhaps reflect the presence of K-bearing high-pressure phases such as phengite (Supplemental Material Figure S1), interaction with fluids from sedimentary sources along the subduction interface during metamorphism, and superimposed fractionation due to devolatilization. It is important to note that these clasts appear to reflect somewhat warmer, high-pressure–high-temperature metamorphism ~50 m.y. ago at initial stages of subduction in the Izu-Bonin-Mariana arc-trench system (Tamblyn et al., 2019) and thus are not directly representative of modern pressure-temperature or N retention in the Mariana slab or at the subduction interface.

Forearc Serpentinite: A Potentially Important Nitrogen Reservoir

Pagé et al. (2018) and Cannaò et al. (2020) suggested that the anomalously high-N rocks in the Himalayas and Gagnone metamorphic suites (mean N ppm = 33 ± 18 ppm, 1σ) could reflect enrichment in ancient forearc wedges via infiltrating slab-derived fluids. Our results suggest that pockets in the mantle wedge with high N are indeed possible, particularly evident in the summit Hole 780C at Conical Seamount (mean N = 35 ± 16 ppm, 1σ). Our N isotope data confirm that this N is most likely sourced from metasedimentary dehydration. Metaultramafic rocks studied by Halama et al. (2014; potentially hydrated in the forearc mantle wedge) have N and $\delta^{15}N$ (1–21 ppm and −3.8‰ to +4.7‰) overlapping with the values in our data set (Fig. 2), consistent with N retention in deeply subducted serpentinites produced in forearc mantle wedges.

Given its volumetric significance, the serpentinized forearc wedge is a potentially important, previously unrecognized N reservoir. Seismic data indicate a serpentinized region of ~10–25 km thick across the Mariana forearc (Tibi et al., 2008). Hyndman and Peacock (2003) proposed that over several tens of millions of years, the Mariana forearc mantle wedge could be entirely serpentinized, given the large amount of slab-derived fluid generated. Using our mean N concentration for serpentinized peridotites, a conservative estimate of serpentinization of 10% for global forearc wedges (may commonly be ~20%; Hyndman and Peacock, 2003), and assuming a global volume of serpentinized forearc for each trench equivalent to Mariana, we calculate that forearcs globally could contain between 1.0×10^{14} to 6.0×10^{14} kg of N (see Supplemental Material Text S4). This reservoir is much smaller than those summarized by Bebout et al. (2016) for continental crust (1.1×10^{18} kg), the atmosphere (3.9×10^{21} kg), and the upper mantle (1.7×10^{17} kg) but comparable to and even larger than important surface reservoirs such as the ocean, soils, biomass, marine biota, and terrestrial vegetation (7.7×10^{14} kg combined).

Some authors have suggested that deep subduction of serpentinized forearc wedges plays a role in arc magma generation (see discussion in Savov et al., 2007). The possible deep subduction of forearc serpentinites containing N with elevated $\delta^{15}N$ (mean = −0.1‰ ± 2.6‰, 1σ; Fig. 4), relative to unmodified mantle (−5‰ ± 2‰), should be considered in attempts to balance subduction inputs and outputs in arc volcanic gases (Mitchell et al., 2010). If retained in forearc wedges and not more deeply subducted, these serpentinites could act as an important transient reservoir of N over geologic time scales. Later subduction of these serpentinites, perhaps related to changing convergent margin dynamics, could result in contribution of this isotopically heavy N to greater depths in the mantle.

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