



On the (mis)behavior of water in the mantle: Controls on nominally anhydrous mineral water content in mantle peridotites



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ABSTRACT

In magmatic settings, water behaves as an incompatible species and should be depleted during melting and enriched during metasomatism. Previous studies have identified correlations between nominally anhydrous mineral (NAM) water content ($[H_2O]$) and indices of metasomatism or melt extraction, seemingly confirming this behavior in the mantle. However in detail, these correlations are ambiguous and do not reflect robust controls on NAM $[H_2O]$. We measured orthopyroxene (opx) and clinopyroxene (cpx) $[H_2O]$ in variably hydrated and metasomatized peridotite xenoliths from the Navajo volcanic field (NVF) that sample the Colorado Plateau subcontinental lithospheric mantle (SCLM), an endmember of SCLM hydration and metasomatism. These xenoliths span a wide range of pyroxene $[H_2O]$ (opx from 50 to 588 ppm wt. H_2O ; cpx from 38 to 581 ppm wt. H_2O), but NAM $[H_2O]$ does not correlate with either indices of melt depletion or metasomatism.

Growth of hydrous minerals suggests higher water activity than in anhydrous peridotites, and therefore hydrous-mineral-bearing xenoliths and anhydrous xenoliths should have different NAM $[H_2O]$ and water activities. However, when the two groups are compared no significant differences can be found in either NAM $[H_2O]$ or water activity. We propose that the high diffusivity of hydrogen in the mantle allows for equilibration of water activity in the mantle over sub-kilometer length scales over geologic time. Such diffusive equilibration reduces water activity variability and results in the blurring and destruction of correlations between NAM $[H_2O]$ and indices of metasomatism or melt extraction. As a result of diffusive equilibration of water, there is a large difference in the variability of concentration between NAM $[H_2O]$ (spanning ~ 2 orders of magnitude) and similarly incompatible elements such as Ce in the same peridotites (spanning ~ 4 orders of magnitude). This difference in behavior explains why H_2O/Ce ratios in mantle peridotites are highly variable relative to those of basalts.

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1. Introduction

Mantle water concentration has a fundamental influence on mantle processes. For example, the strength of olivine decreases dramatically as water content increases, decreasing the viscosity of the mantle (e.g., Hirth and Kohlstedt, 1996; Mei and Kohlstedt, 2000). Addition of water to subcontinental lithospheric mantle (SCLM) via metasomatism from subduction-related melts or fluids can decrease lithosphere viscosity, which may significantly affect intracrustal deformation, rheology, strain localization, and can lower the solidus (e.g., Hirth and Kohlstedt, 1996; Humphreys et al., 2003; Dixon et al., 2004; Li et al., 2008; Behr and Smith, 2016). Over geologic time this may result in lithosphere deformation and possibly delamination (Lee et al., 2011). Delaminated SCLM may

be incorporated into the convecting mantle and contribute mantle geochemical variability (McKenzie and O'Nions, 1983).

Hydrogen in the mantle may be stored either in the structures of hydrous minerals (e.g. amphibole) or as defects within the crystal lattices of nominally anhydrous minerals (NAMs: e.g., olivine, pyroxene, garnet). In NAMs, H enters the crystal lattice as protons attached to structural oxygens at atomic point defects (Beran and Zeman, 1969). The concentration of structural H is calculated in its oxide form, as parts per million H_2O , and referred to broadly as water content or $[H_2O]$ (ppm H_2O by weight). Water in the mantle is thought to behave as an incompatible species, with similar incompatibility to La and Ce (Michael, 1995; Aubaud et al., 2004); therefore, mantle minerals are expected to lose water during melting. In contrast, interaction of the lithospheric mantle with water-rich slab-derived fluids and/or melts can result in metasomatism (here defined as cryptic chemical alteration commonly characterized by light rare earth element [LREE] and large ion lithophile element enrichment) and/or modal hydra-

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tion (here defined as the growth of new hydrous mineral phases, such as amphibole, chlorite, and antigorite). Mantle metasomatism and modal hydration result in compositional changes (e.g., increase in La/Sm or in modal hydrous minerals) that are expected to correlate with water enrichment in mantle NAMs.

In reality, correlations between mantle NAM [H₂O] and indices of either melt extraction or metasomatism are weak or absent in many mantle xenolith suites. A few studies have reported correlations between indices of melt depletion and mantle NAM [H₂O] (e.g., Hao et al., 2014; Warren and Hauri, 2014). For example, NAM [H₂O] correlates with whole rock Yb concentrations (an indicator of melt depletion) in variably metasomatized and melt depleted peridotite xenoliths from the Chinese Cathaysia block (Hao et al., 2014). Other studies note relationships between NAM [H₂O] and atypical indices of metasomatism (e.g., Ti in cpx, bulk Na content, Fe³⁺ in garnet), but not with more emblematic indicators of mantle metasomatism (e.g., La/Sm in cpx, whole rock La/Yb, growth of K-bearing or OH-bearing minerals) (e.g. Peslier et al., 2012; Doucet et al., 2014). In contrast, other studies do not find any correlations between NAM [H₂O] and indices of melting or metasomatism. For example, Denis et al. (2015) studied variably melt depleted and metasomatized xenoliths from Massif Central (France) and found no relationships between cpx, opx, or calculated whole rock [H₂O] and indices of metasomatism, melt depletion, or modal hydration. These conflicting observations raise fundamental questions as to the behavior of hydrogen in the mantle and the controls on [H₂O] in NAMs. In this study, we examine a set of metasomatized and modally hydrated peridotite xenoliths from the Colorado Plateau.

Farallon flat slab subduction beneath the North American Cordillera lasted from ~80 to 25 Ma, releasing fluids and/or melts that hydrated and metasomatized the SCLM (Humphreys et al., 2003). Dixon et al. (2004) predicted that Farallon-derived fluids should have increased NAM [H₂O] in the Colorado Plateau mantle lithosphere. Previous NAM [H₂O] studies of Navajo Volcanic Field (NVF) peridotite xenoliths report some of the highest [H₂O] observed in mantle peridotites, with [H₂O] up to 402 ppm in opx and 957 ppm in cpx (the highest ever measured in natural peridotite samples) (Skogby et al., 1990; Li et al., 2008). Despite NVF SCLM, being long recognized as an extreme endmember of mantle hydration and metasomatism, no previous study has examined the relationship between NAM [H₂O], metasomatism, and modal hydration in the NVF xenoliths. The variable modal hydration and metasomatism observed in NVF xenoliths provide a natural laboratory to study the co-variance of these two processes and NAM [H₂O] in the supra-subduction zone mantle. Here, we show that: (1) correlations between bulk [H₂O] and indices of melt extraction are related to changes in cpx modal abundance rather than changes in NAM [H₂O]; (2) NAM [H₂O] and indices of metasomatism do not display convincing consistent relationships; and (3) rapid hydrogen diffusion in the mantle equilibrates water activity in peridotites with different melt extraction and metasomatic histories, and decouples NAM [H₂O] from indices of metasomatism or melt extraction.

2. Geological setting and sample description

The Colorado Plateau, located in the southwestern United States, is composed of two crustal provinces: the Yavapai province (2.0–1.8 Ga) and the Mazatzal province (1.8–1.6 Ga). These two provinces represent juvenile arcs that were accreted onto the Laurentian margin (Bennett and DePaolo, 1987). Re–Os t_{RD} model ages of peridotite xenoliths from localities within the Colorado Plateau show that the SCLM is similar in age to the overlying crust, suggesting simultaneous formation of crust and lithospheric mantle (Byerly and Lassiter, 2012; Marshall et al., 2017b).

Beginning at ~80 Ma, Farallon plate subduction on the west coast of North America decreased in subduction angle until the slab flattened beneath the SCLM, beginning a period of so called “flat-slab” subduction (Bird, 1988; Humphreys et al., 2003). This flat slab extended eastward from the west coast by more than 700 km into the interior of North America, resulting in compressional deformation in the Cordilleran crust (Laramide orogeny) and hydration of the SCLM. Previous work has documented enrichments in fluid-mobile elements (e.g., Cs, Pb, U, Li, Ba, Rb, Sr), high Sr/Nd ratios, and LREE enrichments in peridotite xenoliths from the southwest United States (e.g., Lee, 2005; Marshall et al., 2017a, 2017b). In the NVF xenoliths, these trace element enrichments are correlated with depletion in ¹⁸O resulting from metasomatism by an ¹⁸O-depleted aqueous fluid and/or melt likely derived from the subducting Farallon slab (Marshall et al., 2017a). Slab rollback occurred ~40 Ma exposing the lithospheric mantle to hotter asthenosphere, possibly triggering post-Laramide magmatism (Humphreys et al., 2003; Lee, 2005).

The formation of the NVF (~30 to 24 Ma) in the central Colorado Plateau coincided with the removal of the Farallon slab from beneath the SCLM. The NVF is a >30,000 km² volcanic field located in the central Colorado Plateau composed mostly (~50) of minette and a few (~8) unusual diatreme-forming intrusions called serpentinized ultramafic microbreccia diatremes (Roden, 1981). The two sample localities of this study, Moses Rock (1 sample) and Green Knobs (16 samples), are two diatremes located in the northern and southern portions of the NVF, respectively (Supplementary Fig. 1).

Peridotite xenoliths from the serpentinized ultramafic microbreccia diatremes are dominantly spinel lherzolites and harzburgites, with some uncommon garnet peridotites or peridotites with garnet rims around spinel (for petrographic descriptions see appendices of Marshall et al., 2017a and Marshall et al., 2017b). Navajo Volcanic Field xenoliths contain mantle-derived hydrous minerals such as antigorite serpentine, chlorite, and amphibole, which grew within the SCLM at ~600 °C (these are referred to here as “primary” hydrous minerals). Many NVF peridotite xenoliths contain primary hydrous minerals (up to 63% hydrous minerals by mode), which replace the anhydrous peridotitic mineralogy (see Smith and Levy, 1976; Smith, 1979, 2010). Although some garnet remains in samples with garnet rims around spinel, garnet in the garnet peridotites has been fully replaced by chlorite. The prior presence of garnet has been inferred in some samples by identification of chlorite pseudomorphs after garnet and heavy REE (HREE) depletion in cpx (Roden et al., 1990). Modal hydration varies widely in mineralogy, texture, and modal abundance, likely reflecting differences in P, T, and water availability (Smith, 1979).

This study divides NVF xenoliths into two groups based on thin section observations: anhydrous peridotites and hydrous-mineral-bearing peridotites. Hydrous-mineral-bearing peridotites contain some perceptible amount of primary (mantle-derived) hydrous minerals, whereas anhydrous peridotites contain no primary hydrous phases. Secondary fine-grained serpentine grows along cracks and grain boundaries in many xenoliths and is associated with late, low-temperature (<300 °C) serpentinization within the diatreme following eruption (Smith, 1979). This later secondary, fine-grained, grain-boundary serpentine is texturally distinct from the earlier mantle-derived primary hydrous minerals that are grain-penetrating and coarse-grained. Presence or absence of secondary serpentine does not affect classification into the anhydrous or hydrous-mineral-bearing groups.

The NVF xenoliths also have variable trace element compositions, ranging from incompatible-element-depleted lherzolites to incompatible-element-enriched harzburgites. Marshall et al. (2017b) categorized peridotite samples into three different groups based on their primitive-mantle-normalized REE patterns: Group D

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