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Metasomatism in the sub-continental lithospheric mantle beneath the south French Massif Central: Constraints from trace elements, Li and H in peridotite minerals

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ABSTRACT

Mantle metasomatism by percolating melts/fluids can significantly modify the geochemical and mineralogical compositions of the sub-continental lithospheric mantle (SCLM). We present a detailed study of water contents and Li concentrations and isotopic compositions in mantle minerals from a suite of peridotite xenoliths entrained by a Cenozoic Strombolian volcano in the southern French Massif Central (FMC). Wide ranging clinopyroxene trace element distributions (e.g., $(\text{La}/\text{Yb})_N$ from 0.25 to 22.21; Ti/Eu ratios from 453 to 4892) suggest that the SCLM has undergone metasomatism by carbonatitic melts/fluids or melts/fluids related to subducted materials. Two amphibole-bearing samples exhibit depletion of light rare earth elements (LREE; $(\text{La}/\text{Yb})_N = 0.26$ and 0.30, respectively) in amphiboles, similar to that in co-existing clinopyroxenes; these samples indicate that amphiboles grew during a separate modally metasomatic event predating the cryptic metasomatism accounting for LREE enrichment and negative HFSE anomalies in other samples. Mineral Li concentrations are similar to those in the normal mantle, with inter-mineral Li partitioning nearly equilibrated and intragranular Li distributions nearly homogeneous. However, negative $\delta^7\text{Li}$ values of pyroxenes in some samples (as low as -8.8‰ in clinopyroxene of sample MC38) can be attributed to diffusive exchange with a small-volume melt of moderate Li concentration and light Li isotopic composition, originally associated with a recycled component. Preservation of the currently observed large inter-mineral Li isotopic variations indicates that melt percolation occurred shortly before entrainment of the peridotite xenoliths by the host magma. Mineral water contents vary from 41 to 428 ppm in clinopyroxenes and from 28 to 152 ppm in orthopyroxenes, and their roughly negative co-variation with co-existing olivine Fo contents imply that partial melting was the main control over mineral water content variations in most samples. Varied water contents in LREE-enriched metasomatized samples indicate the involvement of metasomatic agents of different origins. The aqueous agent responsible for generation of amphiboles in two samples did not produce a notable increase in the water contents of coexisting nominally anhydrous minerals.

1. Introduction

Peridotite xenoliths, commonly entrained by alkali basalts or kimberlites, provide direct information on the sub-continental lithospheric mantle (SCLM; e.g., Pearson et al., 2014). Generally, the SCLM has been depleted by various degrees of partial melting, and has experienced modal or cryptic metasomatism (e.g., O'Reilly and Griffin, 2013) producing significant mineralogical and geochemical variations (e.g., Frey and Green, 1974; Frey and Prinz, 1978; O'Reilly and Griffin, 1988; Rudnick et al., 1993, 2004; Coltorti et al., 1999; Ionov et al., 2002).

Hydrogen can be incorporated into the crystal lattices of mantle nominally anhydrous minerals (NAMs; e.g., Bell and Rossman, 1992;

Ingrin and Skogby, 2000; Hirschmann, 2006; Demouchy and Bolfan-Casanova, 2016). As a trace element in NAMs, hydrogen behaves incompatibly during partial melting and fractional crystallization, and is often compared to Ce (Michael, 1995). Since water is a common component of metasomatic agents at mantle depths, hydrogen in mantle NAMs may represent an effective tracer of mantle metasomatism (Peslier et al., 2012; Doucet et al., 2014; Demouchy et al., 2015; Denis et al., 2015).

Li incorporates into silicate mantle minerals by substituting for Mg and Fe (Seitz and Woodland, 2000; Zhang and Wright, 2012) with intermediate incompatibility similar to Yb during partial melting processes (Ryan and Langmuir, 1987; Brenan et al., 1998; Ottolini et al.,

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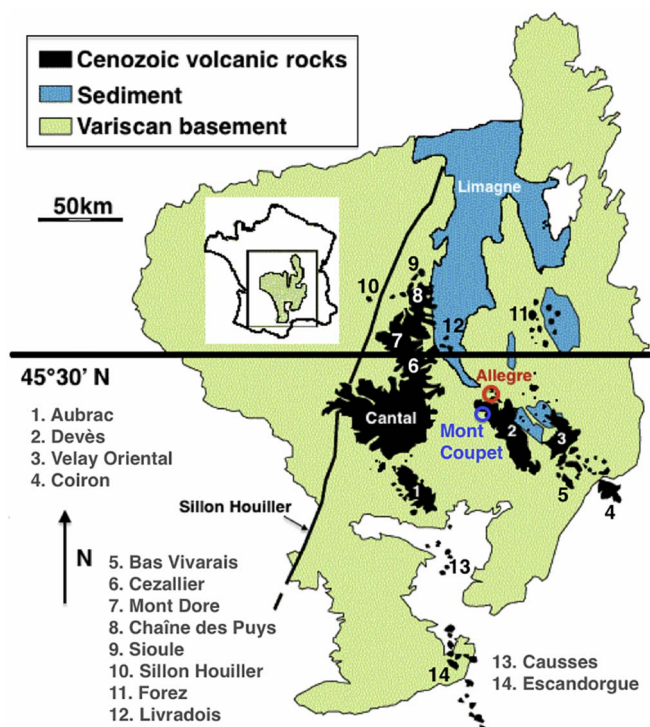


Fig. 1. Simplified geological map of the FMC, and the spatial distribution of Cenozoic volcanoes. The sampling locality of Mont Coupet, (Dèves, region 2), is marked by a blue circle; a photo of the Razas Grand quarry (the exact sampling location) is shown in the lower right corner. A nearby locality (Allègre), from which peridotite xenoliths have also been studied (Gu et al., 2016), is marked by a red circle for comparison. The boundary between the northern and southern domains (horizontal black line; Lenoir et al., 2000) is also drawn (see text). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

2009). The large mass difference between ^6Li and ^7Li (~16%) results in significant isotopic fractionations during various geochemical processes, producing large Li isotopic variations (see review in Tomascak et al., 2016). Melt/fluid-rock interactions strongly affect both the Li contents and isotopic compositions of mantle minerals (e.g., Seitz and Woodland, 2000; Seitz et al., 2004; Woodland et al., 2004; Tang et al., 2007, 2011; Aulbach and Rudnick, 2009; Zhang et al., 2010; Su et al., 2012, 2014; Gu et al., 2016).

The basement of the French Massif Central (FMC) formed during the Variscan orogeny (e.g., Matte, 1986). Based on petrological and geochemical observations of large quantities of peridotite xenoliths, the FMC SCLM consists of two distinct domains to the north and south of a boundary at ~45°30'N (Fig. 1; Lenoir et al., 2000). The more refractory northern domain is characterized by Sr–Nd isotopic compositions that overlap the compositional field of the European Asthenospheric Reservoir, whereas those of the southern domain are more similar to MORB (Granet et al., 1995; Lenoir et al., 2000; Downes et al., 2003). Significant negative high field strength element (HFSE) anomalies have been reported in xenoliths from the northern domain, but are less common in the south, suggesting metasomatism by carbonatitic melts/fluids and silicate melts in the northern and southern domains, respectively (Lenoir et al., 2000; Downes et al., 2003; Wittig et al., 2007). Other studies have attributed metasomatism of peridotites and pyroxenites to melts/fluids related to recycled materials during Variscan subduction (Downes and Dupuy, 1987; Deloule et al., 1991; Touron et al., 2008; Yoshikawa et al., 2010; France et al., 2015; Gu et al., 2016).

In this study, we present major and trace element compositions, water contents, and Li concentrations and isotopic compositions of minerals in peridotite xenoliths transported by Cenozoic volcanism in the FMC to investigate the metasomatic history of the SCLM beneath

the FMC. We determine the behavior of H and Li during SCLM metasomatism.

2. Geological background

Extensive magmatism has occurred across the FMC since the early Cenozoic in three magmatic phases: pre-rift volcanism due to lithospheric bending, rift-related volcanism due to crustal thinning, and a later (15 Ma ~ 6.9 ka ago) magmatic event resulting from asthenospheric upwelling (Michon and Merle, 2001). Many mantle xenoliths have been transported to the surface by these volcanoes (Lenoir et al., 2000). Low-velocity seismic anomalies observed beneath the FMC have been attributed to either a mantle plume (Granet et al., 1995; Goes et al., 1999) or a shallow thermal anomaly related to the Alpine Orogeny (Fichtner and Villaseñor, 2015).

We sampled xenoliths from the Razas Grand quarry in Mont Coupet (Devès, southern FMC domain; Fig. 1), which samples a volcano characterized by Strombolian eruptions 2 Ma ago.

3. Analytical methods

3.1. Sample preparation

We sampled more than twenty volcanic bomb-hosted peridotite xenoliths of 3–12 cm in diameter from the Razas Grand quarry. The outer basaltic shells were sawed off and the cores were cut into billets for polishing. Detailed petrographic observations were performed on thin sections under microscope. According to modal compositions, textures, and the colour of spinel (Spl), eight representative fresh samples, free of visible intrusive veins or any weathering features, were selected for electron microprobe analysis. A subset of six samples, covering the total major element compositional range, was subsequently used for trace element, Li content and isotopic composition, and water content analyses.

3.2. Electron microprobe analysis

Mineral major element compositions were measured on thin sections using a Cameca SX100 electron microprobe at the Service CAMPARIS, Pierre and Marie Curie University (France). Analyses were performed at an accelerating voltage of 15 kV and a beam current of 10 nA with a focused beam. Na and K were analyzed first with counting times of 10 s on peak and 5 s for backgrounds to minimize errors due to element migration. For other elements, counting times were 20 s on peak and 10 s for backgrounds. A series of natural and synthetic minerals was used as standards. At least three grains of each mineral phase were analyzed in a single thin section, and at least three points were analyzed on the core and rims of each crystal.

3.3. Laser ablation inductively coupled plasma mass spectrometry (LA-ICP-MS)

Clinopyroxene (Cpx) and amphibole (Amp) trace element compositions were determined via LA-ICP-MS at GeoRessources Laboratory, University of Lorraine (France). Double-polished thick sections (~0.15 mm thickness) or thin sections were ablated in-situ using a nanosecond excimer laser (GEOLAS Pro; 193 nm wavelength). The spot size was 44 μm , the ablation frequency 5 Hz, and the energy density 10 J/cm² per pulse. Ablation products were transported in a helium flow, mixed with argon gas, and then analyzed with an Agilent 7500 ICP-MS. A complete analysis comprised 30 s for background acquisition and 50 s for sample acquisition. Raw ion intensities were recorded as a function of time. SiO₂ contents obtained from electron microprobe analyses were used as an internal standard; NIST 612 and 614, analyzed at the beginning and end of the analytical session, were used as external standards. The concentrations of some minor elements in olivine (Ol),

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