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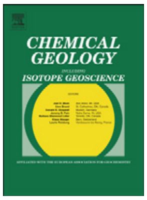

Distinguishing silicate and carbonatite mantle metasomatism by using lithium and its isotopes

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ABSTRACT

To investigate the effects of silicate and carbonatite metasomatism on mantle heterogeneity, we report lithium (Li) concentrations and isotopic compositions for olivine (Ol), orthopyroxene (Opx) and clinopyroxene (Cpx) from two suites of mantle xenoliths (Hannuoba, the North China Craton, and Haoti, the Western Qinling Orogen). The Hannuoba xenoliths range from lherzolite to pyroxenite and were affected by silicate metasomatism, whereas the Haoti xenoliths vary from harzburgite to wehrlite and were affected by carbonatite metasomatism. Lithium concentrations and isotopic compositions display a dichotomy between Hannuoba and Haoti xenoliths, and the overall variation exceeds what was previously reported. The minerals from Haoti xenoliths are more enriched in Li (Ol: 1.23–13.2 ppm; Opx: 3.00–82.8 ppm; Cpx: 1.39–112 ppm) than those from Hannuoba samples (Ol: 1.34–5.52 ppm; Opx: 0.23–16.1 ppm; Cpx: 1.18–79.8 ppm). Lithium isotopic compositions of these samples are highly variable in both suites of samples. δ^7Li ranges from +3.0‰ to +41.9‰ in Ol, from −21.0‰ to +20.2‰ in Opx and from −17.4‰ to +18.9‰ in Cpx for Hannuoba samples. Haoti minerals display a similar degree of variation with δ^7Li ranging from −29.1‰ to +19.9‰ in Ol, −16.9‰ to +18.0‰ in Opx and −45.1‰ to +19.6‰ in Cpx. On average, Li isotopic compositions of minerals from Hannuoba xenoliths follow the sequence of $\delta^7\mathrm{Li}_{\mathrm{Ol}} > \delta^7\mathrm{Li}_{\mathrm{Opx}} > \delta^7\mathrm{Li}_{\mathrm{Cpx}}$, whereas those from Haoti xenoliths are characterized by the opposite sequence of $\delta^7\mathrm{Li}_{\mathrm{Cpx}} > \delta^7\mathrm{Li}_{\mathrm{Opx}} > \delta^7\mathrm{Li}_{\mathrm{Ol}}$; in particular there is considerable difference in δ^7Li values of Ol. The Li elemental and isotopic data suggest that mantle metasomatism by distinct agents is an important process for generating the large heterogeneity of Li abundances and isotopic distribution in the lithospheric mantle. The distinct geochemical characteristics of Li isotopes in silicate and carbonatite metasomatism are closely related to the preferential incorporation of Li into minerals from distinct melts. These findings further demonstrate that the Li isotopic systematics may in turn help to discriminate between silicate and carbonatite metasomatism.
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1. Introduction

Metasomatism is an important and prevalent mechanism controlling the physical and chemical properties of the mantle through infiltration and percolation of silicate or carbonatite melts and aqueous fluids within the upper mantle (Roden and Murthy, 1985; Dautria et al., 1992). Different consequences can be expected, due to the significant disparity in the physical and chemical properties between silicate and carbonatite metasomatic agents. For example, carbonatite melts have much lower viscosity and density, and greater tendency toward wetting grain boundaries than the silicate melts (Genge et al., 1995; Dobson et al., 1996; Gasparik and Litvin, 2002), hence carbonatite metasomatism can elevate the electrical conductivity of the mantle by 2–3 orders of magnitude compared to silicate metasomatism (Gaillard et al., 2008). Considering their significant roles in the evolutionary history of the mantle and the genesis of basaltic and carbonatitic magmas, it is therefore important to establish geochemical proxies that will aid in deciphering the metasomatic history of mantle samples (Dautria et al., 1992; Laurora et al., 2001; Ying et al., 2004; Halama et al., 2009; Zhang et al., 2009).

Silicate metasomatism transforms lherzolite to websterite and orthopyroxenite through olivine (Ol)-consumption and orthopyroxene (Opx)-formation, whereas carbonatite metasomatism would consume Opx to form clinopyroxene (Cpx) and produce a rock series from harzburgite and/or lherzolite to Cpx-rich lherzolite and wehrlite (e.g., Yaxley et al., 1991; Ionov et al., 1996; Laurora et al., 2001). Silicon-

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rich glass inclusion/film is usually present in silicate metasomatized mantle xenoliths, whereas carbonate and CO_2-rich fluid inclusions are common phases in carbonatite metasomatized samples (Yaxley et al., 1991; Frezzotti et al., 2002; Su et al., 2012a). Some geochemical indicators such as Ca/Al, La/Yb, Zr/Hf, Nb/Ta and Ti/Eu ratios as well as trace element patterns have also been developed to distinguish between silicate and carbonatite metasomatism (e.g., Yaxley et al., 1991; Green et al., 1992; Veksler et al., 1998; Gorring and Kay, 2000). However, these indicators in many cases can be explained by both processes (e.g., Klemme et al., 1995; Sweeney et al., 1995; Laurora et al., 2001), hence additional indicators are needed to specifically discern between silicate metasomatism and carbonatite metasomatism.

Lithium and its isotopes may be such a tracer because of their unique distribution between mantle minerals. Compared to pristine peridotites (Li = 1–1.8 ppm in Ol and 0.50–1.3 ppm in pyroxenes; Seitz et al., 2004; Woodland et al., 2004; Jeffcoate et al., 2007), minerals in metasomatized peridotites have much higher Li concentrations and define larger ranges (Li = 0.6–5 ppm in Ol and 0.6–9.5 ppm in pyroxenes; Woodland et al., 2004; Rudnick and Ionov, 2007; Tang et al., 2010; Zhang et al., 2010). In particular, it has been demonstrated that Li is preferentially incorporated into Ol relative to Cpx and Opx during carbonatite metasomatism, whereas the opposite is true in silicate metasomatism (Seitz and Woodland, 2000; Woodland et al., 2004; Zhang et al., 2010; Tang et al., 2011). Lithium isotopes also display a dichotomy between equilibrated peridotites and metasomatized peridotites. Although a normal mantle is estimated to have a δ^7Li of +1‰ to +6‰, as sampled by fresh mid-ocean ridge basalts (Jeffcoate et al., 2007; Tomascak et al., 2008), peridotite xenoliths worldwide display a much larger range with Ol varying from −3‰ to +15‰ (average value: +4.7‰), Opx from −26‰ to +13‰ (average value: +1.1‰) and Cpx from −20‰ to +13‰ (average value: −1.1‰) (Su et al., 2012b and references therein). The large Li isotopic variations in these peridotite xenoliths are attributed to metasomatic processes (e.g., Nishio et al., 2004; Wagner and Deloule, 2007; Aulbach and Rudnick, 2009; Mallmann et al., 2009; Tang et al., 2012). Thus, combined studies of Li concentration and isotopic compositions in mantle-derived xenoliths may help to discriminate between silicate and carbonatite metasomatic agents. The actual mechanism of Li transfer from melt to mantle mineral remains unclear, although the importance of diffusion has been addressed in some studies (Rudnick and Ionov, 2007; Wunder et al., 2007). Also, it is unknown how Li isotopic variations depend on the nature of metasomatic agents because this has yet to be investigated and is in part due to the lack of Li isotopic data for carbonatite metasomatized samples.

Here, we report in situ Li concentration and isotopic data for two suites of mantle-derived xenoliths from Hannuoba, the North China Craton, and Haoti, the Western Qinling Orogen. These two suites of samples have experienced typical silicate and carbonatite metasomatism, respectively, and thus are ideal for exploring the potential of using Li abundances and isotopes to distinguish between silicate and carbonatite metasomatism.

2. Samples

2.1. Silicate-metasomatized xenoliths from Hannuoba

The Hannuoba basalt field is situated at the northern margin of the North China Craton and has been dated at 22 Ma to 14 Ma (Liu et al., 1990). It is well known for its abundant occurrence of diverse mantle xenoliths, varying in composition from peridotite (dunite, harzburgite, lherzolite and wehrlite) to garnet- and/or spinel-bearing pyroxenite (websterite, orthopyroxenite and clinopyroxenite) (Fig. 1a; Chen et al., 2001; Liu et al., 2005; Zhang et al., 2009; Zheng et al., 2009). These xenoliths have been investigated extensively and were interpreted as products of variable degrees of interaction between peridotites and silicate melts (not host basaltic melts) at a depth of 45–65 km with temperature ranging from 800 °C to 1100 °C. The evidence is as follows:

1) Composite xenoliths (e.g., peridotite with pyroxenite veins) display gradual modal variation in mineralogical composition and reaction textures. Orthopyroxenite, a Si-rich rock type, is abundant (Fig. 1a; Liu et al., 2003, 2005; Zhang et al., 2009).
2) The formation of Opx and the zoning texture in Ol indicate mineral transformations during interaction process, whereas the absence of hydrous minerals and/or secondary Cpx in peridotite and pyroxenite supports silicate melts to be the metasomatic agent (Choi et al., 2008).
3) Systematic trace element and Sr–Nd–Pb–Hf–Os–Li isotopic compositions of whole rocks and mineral separates indicate metasomatic overprinting (Song and Frey, 1989; Liu et al., 2005; Tang et al., 2007; Choi et al., 2008; Zhang et al., 2009; Tang et al., 2012).
4) Si-rich glass melt inclusions are abundantly present in Ol and Cpx in the Hannuoba peridotites (Liu et al., 2003; Du and Fan, 2011).
5) Zircon U–Pb dating and sulfide Re–Os dating of the Hannuoba xenoliths show that the inferred metasomatic agents correspond to regional geological events such as Triassic collision, Late Jurassic–Early Cretaceous post-collisional extension, and Tertiary magmatism (e.g., Liu et al., 2004; Zheng et al., 2009).

The xenoliths in this study include two lherzolites, ten websterites, three orthopyroxenites and three clinopyroxenites, covering all types of mantle-derived lithologies in Hannuoba. Their petrological and mineralogical characteristics are similar to those described in previous studies (e.g., Song and Frey, 1989; Chen et al., 2001; Zhang et al., 2013).

2.2. Carbonatite-metasomatized xenoliths from Haoti

The Qinling–Dabie Orogenic Belt is tectonically sutured between the North China Craton and Yangtze Craton. It was formed during the closure of the Paleo-Tethyan Ocean and the collision between the North China and Yangtze Cratons from Paleozoic to Mesozoic. Cenozoic kamafugite and carbonatite association dated at 23 Ma to 7 Ma is distributed in the Western Qinling (Yu et al., 2005). Mantle xenoliths collected in the Haoti cinder cones are mainly spinel and garnet facies lherzolites (Fig. 1b; Su et al., 2010a, 2011, 2012a,b,c). Recent studies suggest that the lithospheric mantle beneath the Western Qinling is compositionally stratified with a gradual decrease in fertility with depth, a feature that probably resulted from varying degrees of carbonatite metasomatism (Su et al., 2010a, 2011). Carbonatite metasomatic signatures recorded in these xenoliths are listed as follows:

1) Carbonate veins and discrete grains are commonly observed (Fig. 1b; Su et al., 2009, 2010a, 2012a).
2) Clinopyroxene has high CaO and Na_2O contents, has high light rare earth elements (LREE) relative to heavy rare earth elements (HREE), has Ba, Th, U, Pb and Sr enrichments and negative Ti, Hf and Y anomalies, and is in particular characterized by high Ti/Eu ratio (Su et al., 2010a,b, 2011). Additionally, the spongy textures are commonly present in the Cpx of the Haoti xenoliths and have been interpreted to result from decompressional melting (Su et al., 2011).
3) Whole-rock compositions of the Haoti peridotites display LREE-enrichment, positive Sr and Ba anomalies, carbonatite-like trace element patterns, and Sr–Nd–Pb isotopic mixing trend between DM (depleted mantle) and EMII (enriched mantle II) end members, which are consistent with the geochemical features resulting from carbonatite metasomatism (Su et al., 2010a, 2012a).
4) Previous Li isotopic studies indicate that the constituent minerals of the Haoti peridotites have extremely high Li contents and distinct δ^7Li values, which might be linked to carbonatite metasomatism (Su et al., 2012b).

The lherzolite xenoliths investigated here include two garnet-facies, six spinel-facies and one garnet-spinel coexisting samples, which were

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