



Iron isotope tracing of mantle heterogeneity within the source regions of oceanic basalts



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ABSTRACT

Mineralogical variations in the Earth's mantle and the relative proportions of peridotitic versus enriched and potentially crustally-derived pyroxenitic domains within the mantle have important implications for mantle dynamics, magma generation, and the recycling of surface material back into the mantle. Here we present iron (Fe) stable isotope data ($\delta^{57}\text{Fe}$, deviation in $^{57}\text{Fe}/^{54}\text{Fe}$ from the IRMM-014 standard in parts per thousand) for peridotite and garnet–pyroxenite xenoliths from Oahu, Hawaii and explore Fe isotopes as tracer of both peridotitic and pyroxenitic components in the source regions of oceanic basalts. The pyroxenites have $\delta^{57}\text{Fe}$ values that are heavy (0.10 to 0.27‰) relative to values for mid-ocean ridge and ocean island basalts (MORB; OIB; $\delta^{57}\text{Fe} \sim 0.16\text{‰}$) and the primitive mantle (PM; $\delta^{57}\text{Fe} \sim 0.04\text{‰}$). Pyroxenite $\delta^{57}\text{Fe}$ values are positively correlated with bulk pyroxenite titanium and heavy rare earth element (REE) abundances, which can be interpreted in terms of stable isotope fractionation during magmatic differentiation and pyroxene cumulate formation. In contrast, the peridotites have light $\delta^{57}\text{Fe}$ values (−0.34 to 0.14‰) that correlate negatively with degree of melt depletion and radiogenic hafnium isotopes, with the most depleted samples possessing the most radiogenic Hf isotope compositions and lightest $\delta^{57}\text{Fe}$ values. While these correlations are broadly consistent with a scenario of Fe isotope fractionation during partial melting, where isotopically heavy Fe is extracted into the melt phase, leaving behind low- $\delta^{57}\text{Fe}$ peridotite residues, the extent of isotopic variation is far greater than predicted by partial melting models. One possibility is derivation of the samples from a heterogeneous source containing both light- $\delta^{57}\text{Fe}$ (relative to PM) and heavy- $\delta^{57}\text{Fe}$ components. While pyroxenite is a viable explanation for the heavy- $\delta^{57}\text{Fe}$ component, the origin of the depleted light- $\delta^{57}\text{Fe}$ component is more difficult to explain, as melting models predict that even large (>30%) degrees of melt extraction do not generate strongly fractionated residues. Multiple phases of melt extraction or other processes, such as metasomatism, melt percolation or the assimilation of xenocrystic olivine with light $\delta^{57}\text{Fe}$ values may need to be invoked to explain these light $\delta^{57}\text{Fe}$ values; a caveat to this is that these processes must either preserve, or generate correlations between $\delta^{57}\text{Fe}$ and Hf isotopes. Published variations in $\delta^{57}\text{Fe}$ in mantle melting products, such as MORB and OIB, are also greater than predicted by melting models assuming derivation from $\delta^{57}\text{Fe}$ -homogeneous mantle. For example, OIB from the Society and Cook-Austral islands, which have radiogenic Pb and Sr isotope compositions indicative of recycled components such as subduction modified, low-Pb oceanic crust and terrigenous sediments have heavy mean $\delta^{57}\text{Fe}$ values ($\sim 0.21\text{‰}$) significantly distinct to those of other OIB and MORB, which could be explained by the presence of heavy- $\delta^{57}\text{Fe}$ pyroxenite cumulate or pyroxenitic melt components, whereas large degree partial melts, such as komatiites and boninites, display light Fe-isotopic compositions which may reflect sampling of refractory, light- $\delta^{57}\text{Fe}$ mantle components. Iron stable isotopes may therefore provide a powerful new means of fingerprinting mineralogical variations within the Earth's mantle and identifying the mineralogy of depleted and enriched components within the source regions of volcanic rocks.

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1. Introduction

Mineralogical variation in the Earth's upper mantle and the potential existence of enriched (pyroxenitic or eclogitic, pyroxene-dominated) and depleted (peridotitic, olivine-dominated) mantle

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components has been the subject of debate for several decades (Allègre and Turcotte, 1986; Hauri, 1996; Hofmann and White, 1982; Workman et al., 2004). Numerous studies on erupted melts have used major and trace elements and short- or long-lived radiogenic nuclides to constrain mantle compositional and/or mineralogical variability (Elliott et al., 2007; Hauri, 1996; Humayun et al., 2004; Jackson and Dasgupta, 2008; Prytulak and Elliott, 2007; Sigmarrsson et al., 1998; Sobolev et al., 2005; Stracke et al., 1999; Vlastelic et al., 1999). However, while pyroxenitic or eclogitic components are often invoked to account for the enriched isotopic signatures of oceanic basalts (Allègre and Turcotte, 1986; Hauri et al., 1996; Hirschmann and Stolper, 1996; Lassiter and Hauri, 1998; Lundstrom et al., 1999; Niu et al., 1999; Prinzhofer et al., 1989; Zindler et al., 1979; Zindler et al., 1984), their roles in generating mantle chemical heterogeneity remain controversial, as few independent tracers of source mineralogy exist, such that resolving compositional (trace element or radiogenic isotope) enrichment from mineralogical enrichment is challenging. For example, radiogenic isotope systems such as Sr, Nd, Hf and Pb can fingerprint crustally-derived components, but cannot distinguish whether these components remain present as distinct lithological units (e.g. as pyroxenite or eclogite), or whether they are completely homogenized into the mantle by means of convective stirring (Gurenko et al., 2009; Jackson and Dasgupta, 2008) such that only their geochemical signals remain.

Pyroxenite source components can be generated from both crustal and mantle-sourced protoliths. 'Crustal' pyroxenite components may be directly derived from recycled oceanic crust as eclogite (Pertermann and Hirschmann, 2003) or may form by i) partial melting of subducted eclogite and the reaction of these melts with mantle peridotite to form garnet pyroxenite (Hauri, 1996; Huang and Frey, 2005; Sobolev et al., 2005); this scenario has been invoked to explain the high SiO₂ and Ni contents and high Fe/Mn ratios of some Hawaiian basalts (Sobolev et al., 2007; Sobolev et al., 2005), or ii) the extraction of silica-rich fluids or melts from oceanic crust during subduction (Kogiso et al., 2003). In contrast, 'mantle' pyroxenite components are considered to form as high-pressure cumulates of low-degree mantle melts that infiltrate and crystallize near the base of the oceanic mantle lithosphere (Niu and O'Hara, 2003; Pilet et al., 2008) or through interaction and melt-rock reaction between magmas and surrounding peridotite wall-rock (Downes, 2007). For clarity, we use the term "pyroxenite" to refer to source mineralogy (olivine-free, pyroxene and garnet-bearing) rather than source origin unless this is referred to specifically.

Depleted peridotitic components have also been invoked within the source regions of OIB and MORB. Several studies have suggested that the Hawaiian plume mantle source contains long-term depleted and compositionally variable peridotitic components (Bizimis et al., 2013, 2005; Pietruszka and Garcia, 1999; Ren et al., 2006; Stracke et al., 1999) and depleted components have also been invoked in the source of Reunion OIB (Vlastelic et al., 2006) and MORB. Evidence for the latter is provided by correlated Hf–Nd isotopes in MORB (Salters et al., 2011) and the radiogenic Hf isotope compositions of Gakkel Ridge abyssal peridotites (Stracke et al., 2011), while mantle peridotites found at mid-oceanic spreading centers and as xenoliths in OIB provide some of the strongest evidence for the presence of ancient depleted peridotites in the convecting mantle (Bizimis et al., 2007; Burton et al., 2012; Liu et al., 2008; Stracke et al., 2011). However, identifying the presence of depleted components in mantle source regions is not without difficulty. A major challenge is the low incompatible element concentrations of refractory mantle peridotites, which means that they have little influence on the incompatible trace element and radiogenic isotope budget of erupted melts (Burton et al., 2012). Another challenge relates to the ease

with which the incompatible trace element and radiogenic isotope signature of mantle rocks can be overprinted by metasomatic processes with little or no change in mineralogy (Niu and O'Hara, 2003).

New tracers of both depleted and enriched mantle mineralogical components are thus required to compliment the extensive and rapidly growing evidence for mantle heterogeneity based on trace elements and radiogenic isotopes. Given the major advances that have been made in "heavy" (high atomic weight) metal stable isotope analyses over the last ten years, it is now timely to explore the use of these stable isotope systems as tracers of mantle heterogeneity. In this study, we explore the use of Fe stable isotopes as a tracer of mantle source mineralogy using peridotite and pyroxenite xenoliths from Hawaii as a case study.

1.1. Iron isotopes as a tracer of mantle mineralogical variations

Iron is a major cation in the Earth's mantle, with a bulk partition coefficient close to 1 (Herzberg, 2004; Keshav et al., 2004; Kogiso and Hirschmann, 2006; Pearce and Parkinson, 1993; Pertermann and Hirschmann, 2003; Sobolev et al., 2005; Weyer and Ionov, 2007). Consequently, Fe concentrations vary little with degree of melting (at constant pressure) in primary MORB melts (Klein and Langmuir, 1987) and peridotites (Ionov and Hofmann, 2007), at least for degrees of melting appropriate for present day MORB and OIB (generally <15–20%). Theoretical (Polyakov and Mineev, 2000) and empirical observations from equilibrated peridotites and pyroxenites (Weyer and Ionov, 2007; Williams et al., 2005) indicate that isotopically heavy Fe (high $\delta^{57}\text{Fe}$; parts per thousand deviation in $^{57}\text{Fe}/^{54}\text{Fe}$ from the IRMM-14 iron standard) will be concentrated in both low- and high-Ca pyroxenes relative to olivine (by ca. 0.15 to 0.20‰; Weyer and Ionov, 2007; Williams et al., 2005) due to differences in bonding environment. As melting preferentially consumes pyroxene over olivine, peridotitic residues, melts and cumulates derived from melts of peridotitic and pyroxenitic source regions should inherit distinct Fe isotope signatures reflecting both the degree of melt extraction and the nature of the source mineralogy. Furthermore, as the Fe contents of melts derived from peridotitic and pyroxenitic mineralogies (e.g., Sobolev et al., 2005 their Table 1) are approximately similar, neither lithology should disproportionately contribute to the Fe budget of erupted melts and the Fe isotope compositions of primitive lavas should therefore primarily reflect the mineralogies of their respective source regions.

While mineral-specific Fe-isotope partitioning effects have been recognized in a number of earlier studies (Beard and Johnson, 2004; Teng et al., 2008; Weyer et al., 2005; Weyer and Ionov, 2007; Williams et al., 2004, 2009, 2005) the effects of variable source mineralogy have not yet been extensively explored in either models attempting to simulate Fe isotope fractionation during partial melting or in calculations of the Fe isotope composition of the Earth's mantle. Existing estimates of the Fe isotope composition of the Earth's mantle or the bulk silicate Earth (BSE) are constrained by sampling to the upper mantle and are generally based on suites of comparatively primitive basalts or their melting residues. In an early study (Weyer et al., 2005), it was observed that unmetasomatized peridotites from a variety of tectonic settings displayed near-chondritic $\delta^{57}\text{Fe}$ values whereas oceanic basalts displayed heavier $\delta^{57}\text{Fe}$ values (mean ~ 0.16 ‰), similar to those of high-Mg lunar basalts but heavier than the chondritic $\delta^{57}\text{Fe}$ values displayed by SNC meteorites and eucrites (Poitrasson et al., 2004; Schoenberg and von Blanckenburg, 2006; Weyer et al., 2005). Weyer et al. (2005) concluded that the mean peridotite $\delta^{57}\text{Fe}$ provided the best estimate of the BSE, noting that relative differences in planetary mantle $\delta^{57}\text{Fe}$ could only be estimated through comparisons of erupted basalts and their meteoritic

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