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Appendix: Treatment of geochemical data

The geochemical database of major elements and isotopes from global OIBs from Jackson and Dasgupta (2008) is used to provide a global reference for the major element variability in OIBs (Supplementary Table 1). This high quality, filtered data set provides coupled measurements of radiogenic isotopes and major elements on the freshest, least evolved OIB samples, which facilitates estimating primary magma compositions and relating these melt compositions to mantle source variability inferred from their isotopic variability.

The major element compositions associated with the highest $^3He/^4He$ lavas globally were obtained for lavas from four active hotspots (Hawaii, Iceland, Galapagos and Samoa) and a flood basalt associated with the proto-Iceland plume (Baffin Island and West Greenland (BIWG)). At Hawaii, Galapagos and Samoa, the volcanoes with the highest $^3He/^4He$ are selected for comparison with the global OIB database, as it is critical to target and evaluate the major element composition of lavas originating from the high-$^3He/^4He$ mantle reservoir. At Hawaii, Galapagos and Samoa, the volcanoes with the highest $^3He/^4He$ (Loihi, Fernandina and Ofu, respectively) are relatively young (<1 m.y. old; McDougall 2010; Garcia et al. 2006; Geist et al. 2006; Kurz et al. 2009; Koppers et al. 2011) and are located at or near the active end of the respective "hotspot" track. Paired $^3He/^4He$ and major element and heavy radiogenic isotopic (i.e., $^{87}Sr/^{86}Sr$, $^{143}Nd/^{144}Nd$, $^{206}Pb/^{204}Pb$) data are not available on most lavas from these three volcanoes, but lavas from each volcano are found to exhibit elevated $^3He/^4He$. Therefore, to augment the geochemical data set for this analysis, both lavas with helium isotopic measurements and lavas that lack $^3He/^4He$ data from Loihi, Fernandina, and Ofu are used for evaluating the major element composition of the high-$^3He/^4He$ reservoir from each respective ocean island. The geochemical data for Loihi, Fernandina, and Ofu that were used in this study are from Georoc database (http://georoc.mpch-mainz.gwdg.de/georoc) and shown in Supplementary Table 1[1]. We emphasize that the $^3He/^4He$ of most lavas examined here are not known, as $^3He/^4He$ measurements are relatively rare while major element data required for our analysis are common in the literature.

Unlike Hawaii, Galapagos and Samoa, which have individual volcanic centers that are associated with elevated $^3He/^4He$, lavas with the highest $^3He/^4He$ (>30 Ra) in Iceland are located in the late Tertiary basalts in the northwest portion of the island (Hilton et al. 1999), and are not associated with specific volcanic centers. The Icelandic tertiary basalts remain poorly characterized for helium isotopic compositions (Breddam and Kurz 2001; Graham 2002; Harlou 2007); only a single lava with $^3He/^4He$ >30 Ra has been identified in outside of the Tertiary basalts of northwest Iceland (Macpherson et al. 2005). Therefore, instead of taking the approach of using specific volcanic centers as representative of the high-$^3He/^4He$ component in Iceland-which is the approach taken for characterizing melts of the high-$^3He/^4He$ mantle at Hawaii, Galapagos and Samoa-we limit our analysis in Iceland to samples from the Tertiary province with $^3He/^4He$ >30 Ra; the database for Iceland Tertiary lavas with paired major element analyses and $^3He/^4He$ >30 Ra is limited to only 3 samples (Hilton et al. 1999; Harlou 2007; Jackson et al. 2008), and they provide a preliminary characterization of the high-$^3He/^4He$ component beneath Iceland. The highest $^3He/^4He$ lavas globally (up to 50 Ra) are reported from the flood basalt province in Baffin Island and West Greenland (BIWG), which was erupted at 62 Ma and is associated with the proto-Iceland plume (e.g., Francis 1985; Robillard et al. 1992; Graham et al. 1998; Stuart et al. 2003; Kent et al. 2004; Starkey et al. 2009; Larsen and Pedersen 2009). Owing to their age, the $^3He/^4He$ ratios in the BIWG samples have experienced variable ingrowth of ^4He since eruption, and some of the BIWG lavas have relatively low $^3He/^4He$. Jackson et al. (2010) found a relationship between $^3He/^4He$ and helium concentrations that supports this hypothesis, and they showed that post-eruptive ingrowth of radiogenic helium is likely responsible for the relatively low $^3He/^4He$ observed in some of the BIWG samples, as these samples are helium-poor. Owing to eruption through continental crust, many BIWG lavas have experienced crustal assimilation, which perturbs their isotopic compositions away from mantle compositions (e.g., Larsen and Pedersen 2009). However, Larsen and Pedersen (2009) used major and trace element proxies for assimilation to identify lavas that have experienced the least assimilation, and they found that such lavas tend to plot in a tight cluster in Pb isotopic space. A subset of geochemically well-characterized lavas from the BIWG have also been characterized for their Pb-isotopic compositions, which is necessary for evaluating a role for crustal assimilation, and this subset of lavas is argued to have experienced minimal assimilation (Graham et al. 1998; Jackson et al. 2010) and is shown in all relevant figures. Additionally, a larger geochemical data set for BIWG lavas characterized for $^3He/^4He$ is available on lavas that have not been characterized for the Pb-isotopic compositions (Stuart et al. 2003; Starkey et al. 2009), and these data are shown separately in the relevant figures.

To estimate primary melt compositions, low-MgO lavas (<8 wt% for Loihi, Fernandina and BIWG, and <10 wt% for Ofu) were excluded, as such lavas can be affected by clinopyroxene and plagioclase fractionation. The major element compositions of the remaining lavas are affected primarily by fractionation or accumulation of olivine (as evidenced by horizontal data trends in MgO vs. CaO/Al_2O_3 plot) (Fig. 2). The lavas were renormalized to 100 wt% with all iron reported as FeO_{total}. Lavas were corrected for olivine fractionation/addition in 0.05 wt% increments until they were in equilibrium with mantle olivine of forsterite content 90 (i.e., Fo_{90}). Equilibrium olivines were generated allowing olivine-melt K_d to vary with the composition based on the parameterization of Tamura et al. (2000) presented in Lee et al. (2009) and assuming molar $Fe^{3+}/Fe_T = 0.1$.

For comparison we also included 43 MORB glass samples with the highest MgO concentrations, between 8.8 to 10.45 wt% MgO, which include 17 glass samples from Siqueiros (Perfit et al. 1996) and 26 glass samples with the highest MgO from Melson et al. (2002). These MORB samples are sufficiently primitive that they are affected only by olivine fractionation. The MORB samples are also olivine fractionation corrected to be in equilibrium with Fo_{90} olivine and the corrected major element compositions of the Siqueiros lavas are similar to the 26 glass samples with the highest MgO from Melson et al. (2002).