We have measured Ni, Ca, and Mn in olivine phenocrysts from volcanoes in the Galápagos Archipelago to infer the mantle source lithologies. Results show that peridotite is the dominant source lithology for Fernandina, Floreana, Genovesa, Wolf Island, and Darwin Island. These volcanoes largely characterize the PLUME, WD, FLO, and DUM Nd, Sr, and Pb isotopic endmembers of Harpp and White (2001). Volcan Wolf, Alcedo, Marchena, and Cerro Azul, also produced from the melting of peridotite sources, have isotopic compositions that can be defined by mixing of the four isotopic endmembers. Our analysis suggests that peridotite was present in the sources of the volcanoes covered in this study and therefore is the dominant source lithology of the Galápagos plume. Pyroxenite melting is generally focused in two isotopically distinct domains: Roca Redonda, Volcan Ecuador, and Sierra Negra in the enriched western part of the archipelago and Santiago, Santa Cruz, and Santa Fe in the depleted east. One implication of this finding is that the Western and Eastern Pyroxenite Domains represent two separate bodies of recycled crust within the Galápagos mantle plume. Furthermore, both isotopically enriched and depleted domains of the archipelago were generated from mixtures of peridotite and pyroxenite. This suggests that there is no relationship between the source lithology of the Galápagos plume and its isotopic characteristics. The identification of peridotite-source melting in volcanoes with isotopic characteristics that have been attributed to recycled crust points to the importance of mixing in OIB genesis, consistent with studies in the Canary Islands.
1. Introduction

Petrological modeling has shown that lavas with 65–95 Ma ages from the Caribbean Large Igneous Province (CLIP) were from a hotter source that melted more extensively than the 0–15 Ma lavas from the Galápagos Archipelago and the Carnegie and Cocos hot spot tracks [Herzberg and Gazel, 2009]. This provides petrological evidence for secular cooling of the Galápagos Plume. In general, mantle plumes for LIPs with Paleocene-Permain ages were hotter and melted more extensively than plumes of more modern oceanic islands [Herzberg and Gazel, 2009]. The high melt fractions, high mantle potential temperatures, and vast areas of magmatism associated with the largest LIPs are all consistent with formation in mantle plume heads [Richards et al., 1989; Coffin and Eldholm, 1994; Herzberg and Gazel, 2009]. In many cases, these events were so large that they had a profound impact on the hydrosphere, atmosphere, and biosphere [e.g., Saunders, 2005; Kerr, 2005; Wignall, 2005].

A major unresolved question emerges from these recent developments: Why were mantle plumes that formed old LIPs hotter than those that produce modern ocean island basalts (OIB)? One possibly has to do with variable amounts of recycled crust in the source of intraplate magmas has been inferred from radiogenic isotopes (e.g., Pb, Sr, Nd, Hf) [Zindler and Hart, 1986]. However, isotopic data cannot be used to determine whether the crust is still present as a lithological unit in the source [Sobolev et al., 2007; Herzberg, 2011b], or if it has been mixed and stirred back into peridotite [Gurenko et al., 2009; Day et al., 2010; Herzberg, 2011b]. Furthermore, it is not always clear whether the isotopic characteristics of recycled crust can be distinguished from those of metasomatized peridotite [e.g., Harpp and White, 2001; Pilet et al., 2008; Niu and O’Hara, 2003] or even deep melting in the lower mantle [Collerson et al., 2010].

This paper is the first of two contributions that are designed to constrain the amount of recycled crust in the Galápagos plume, and how it might have changed from the Cretaceous to the present. In this first report, we make use of high-precision Ni, Mn, and Ca contents of olivine phenocrysts in lavas from the 0–3 Ma Galápagos Archipelago to infer the presence and proportion of recycled crust in the mantle plume [Sobolev et al., 2007, 2005; Herzberg, 2011b]. Relative to a peridotite source, olivines with elevated Ni and Fe/Mn and lower Ca and Mn have been attributed to recycled crust in the source [Sobolev et al., 2007; Herzberg, 2011b]. Correlations between source lithology and He, Sr, Nd, and Pb isotopes are examined in order to test mixing models often invoked to explain isotopic variations. In a follow up to this study, we will report estimates of the amount of recycled crust in the ancestral Galápagos plume that melted to produce olivine-phyric basalts and picrites found throughout the CLIP and accreted Galápagos oceanic islands/tracks in Costa Rica and Panama.

2. Isotopic Background

Isotopic variations within the Galápagos have been used to divide the archipelago into four domains (Figures 1 and 2) [Hoernle et al., 2000]. The Eastern Isotopic Domain [Hoernle et al., 2000] is composed mostly of magmas with a depleted isotopic signature similar to that of mid-ocean ridge basalts. This observation has been interpreted as the entrainment of asthenosphere into the ascending plume [Geist et al., 1988; Harpp and White, 2001; White and Hofmann, 1978; White et al., 1993]. Unlike the entrainment
model, Hoernle’s model calls for a depleted component which forms as a result of progressive depletion of plume material that continuously melts as plume material migrates toward the Galápagos spreading center [Hoernle et al., 2000]. The depleted component in the plume, perhaps most prevalently in the magmas of Genovesa, has been termed DUM or depleted upper mantle by Harpp and White [2001].

More enriched magmas, erupted along the southern and western peripheries of the archipelago, define the Northern, Central, and Southern Isotopic Domains [Hoernle et al., 2000]. The island of Fernandina and its submarine flows have the highest $^3\text{He}/^4\text{He}$ ratios of any volcano in the Galápagos [Geist et al., 2006; Kurz et al., 2009; Kurz and Geist, 1999]. High primitive helium and neon isotope ratios indicate that a component contributing to the magmatism of Fernandina consists of primitive, relatively undegassed mantle [Kurz et al., 2009; Kurz and Geist, 1999; Harpp and White, 2001]. The source of the enrichment in $^3\text{He}$ and a moderate enrichment in Sr, Nd, and Pb isotopes is termed PLUME [Harpp and White, 2001].

A localized, highly enriched component found at the southern edge of the archipelago has the most radiogenic $^{87}\text{Sr}/^{86}\text{Sr}$ and $^{206}\text{Pb}/^{204}\text{Pb}$ compositions found in the islands. Found predominantly...
Figure 2. Pb isotope ratios of the four isotopic domains [Hoernle et al., 2000] in Figure 1. Data from: Allan and Simkin [2000], Blichert-Toft and White [2001], Cullen et al. [1987], Cullen et al. [1989], Geist et al. [2002], Geist et al. [2005], Handley et al. [2011], Harpp and White [2001], Kurz and Geist [1999], Naumann et al. [2002], Pik et al. [1999], Reynolds and Geist [1995], Saal et al. [2007], Standish et al. [1998], Teasdale et al. [2005], Thompson et al., 2004, Vicenzi et al. [1990], White and Hofmann [1978], White [1979], and White et al. [1993].

at the island of Floreana this endmember is termed FLO [Harpp and White, 2001]. This endmember appears to consist mostly of high μ (HIMU) with some Enriched Mantle II (EMII) [Harpp and White, 2001; Harpp et al., 2013], but plots outside of the major mantle reservoirs [Hart et al., 1992; White, 1985; Zindler and Hart, 1986]. This anomalous isotopic signal was previously thought to be the result of a recycled component or metasomatized asthenosphere [Bow and Geist, 1992; Harpp and White, 2001]. The presence of high εHf and low εSr is taken to indicate the presence of recycled sediment, which is also consistent with a component of EMII [Blichert-Toft and White, 2001]. More recent work in Floreana [Harpp et al., 2013] suggests that the isotopic and trace element geochemistry of Floreana points to a source which is comprised of recycled oceanic crust (FLO) that has been altered by hydrous fluids, DUM, and PLUME.

[5] Islands north of the Galápagos platform, with the exception of Genovesa, exhibit a slight enrichment in 207Pb and 208Pb which cannot be reconciled by the mixing of the PLUME, DM, and FLO endmembers of Harpp and White [2001]. Most evident in the Wolf-Darwin Lineament, the source of the Pb enrichment is termed WD [Harpp and White, 2001]. In their 2001 paper, Harpp and White proposed a number of different possibilities for the source for the WD endmember. These include recycled sediment [Dupré and Allegre, 1983], subducted oceanic crust, and subcontinental lithosphere [Hart, 1984]. The source of the isotopic anomaly remains unresolved; however, the contribution of the plume north of the platform is seemingly supported by isotopic data, despite its distance from the plume axis [Harpp and White, 2001].

[10] Hot spot tracks on the Cocos plate preserve a record of the Northern, Central, and Eastern Isotopic Domains [Hoernle et al., 2000] back to ~14.5 Ma, while tracks on the Nazca plate indicate the presence of the Southern, Central, and Eastern Isotopic Domains [Hoernle et al., 2000] to at least 17.3 Ma [Werner et al., 2003]. The Northern Isotopic Domain [Hoernle et al., 2000] was first evident in the accreted seamount at Quepos, Costa Rica (60–65 Ma) [Geldmacher et al., 2003] and then later as a major component of the Seamount Province of the Cocos Ridge (Figure 3) [Werner et al., 2003]. According to Hoernle et al. [2002, 2004], the geochemistry of CLIP lavas establish the activity of the Galápagos plume back perhaps as far as 139 Ma. Geldmacher et al. [2003] show that CLIP lavas 70–90 Ma are isotopically similar to the modern Eastern, Central, and Southern Isotopic Domains [Hoernle et al., 2000] (Figure 3). Gorgona (90 Ma) has a wide spectrum of depleted and enriched isotopic compositions, yet it melted when the plume was hottest [Herzberg and Gazel, 2009].

3. Petrological Methods

3.1. Analytical Method

[11] We analyzed olivine phenocrysts in 64 samples from 19 volcanoes across the Galápagos Archipelago. A total of 4677 high-precision analyses were obtained from olivine phenocrysts of varying sizes and forsterite contents (Table S1; supporting information1). Single core analyses were taken from olivine phenocrysts identified as homogeneous from reconnaissance EMP (electron microprobe) work and backscatter images. Traverses were taken from olivines that exhibited zoning.

[12] Olivine, major element, and trace element data were obtained using the method of Sobolev et al. [2007]. Analyses were conducted on the JEOL JXA-8200 electron microprobe at Rutgers University. A current of 300 nA and an accelerating voltage of 20 kV were used for all analyses. Count

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1Additional supporting information may be found in the online version of this article.
times on the elements are as follows: 50 s for Si, 80 s for Mg, 100 s for Fe, and 150 s for Mn, Ni, and Ca. The San Carlos olivine standard was analyzed at regular intervals throughout every run to correct for drift. Detection limits at a 3σ (99%) confidence level and errors (2σ) were obtained from the Probe for Electron Probe Micro Analyser (EPMA) program [Donovan, 2012]. Average detection limits for Si, Mg, and Fe are 30, 26, and 40 ppm, respectively. Average detection limits for trace elements Ni, Mn, and Ca are 27, 28, and 16 ppm, respectively. These detection limits are higher than those reported in other studies which applied this analytical method using JEOL software [Gurenko et al., 2009, 2013; Sobolev et al., 2007, 2011]. Analyses of major elements Si, Mg, and Fe have an average relative 2σ error of ~0.09%, ~0.11%, and ~0.23%, respectively. Average 2σ errors for trace elements Ni, Mn, and Ca are 31, 37, and 22 ppm, respectively. These errors are slightly higher than those reported in previous studies. The errors we report are from counting statistics from the analyses of the Galápagos olivines, while other studies [Gurenko et al., 2009, 2013; Sobolev et al., 2007, 2011] report two standard errors of the analyses of the San Carlos olivine standard. Two standard deviations of Ni, Mn, Ca, and Fo for the San Carlos olivine analyses (n = 680) in our study are 32 ppm, 19 ppm, and 0.1%, respectively. These values are in agreement with the values reported in Gurenko et al. [2009] which are 76 ppm, 37 ppm, 16 ppm, and 0.1%, for Ni, Mn, Ca, and Fo, respectively. Analyses with oxide totals greater than ±2% deviation from 100% were excluded. Chemical formulas were calculated for all analyses. Analyses with deviations in stoichiometry greater than ±1% were excluded.

3.2. Computational Method

[13] Source lithology is inferred by comparing the compositions of observed olivine phenocrysts with those that have been computed for primary and derivative melts of mantle peridotite [Herzberg, 2011b]. Olivine phenocrysts with compositions that cannot be matched to a peridotite-source provenance are assumed to have crystallized from magmas of a pyroxenite source (e.g., Figures 5 and 6). The black colored forms represent model Mg numbers (i.e., 100Mg/(Mg + Fe) in cation mol fractions), Ca, Mn, and Ni contents of olivine phenocrysts of primary magmas of a fertile peridotite composition [Herzberg, 2011b].

[14] When mantle peridotite partially melts and the primary magmas are transported readily to the surface, the liquidus phase is invariably olivine [O’Hara, 1968]. The effect of decompression, however, is to expand the liquidus crystallization
fields of both olivine and clinopyroxene [O’Hara, 1968; Herzberg, 1992]; the importance of sub-Moho clinopyroxene fractionation in Galápagos petrogenesis has been recognized previously [Geist et al., 1998]. Relative to olivine-only crystallization, deep fractionation of clinopyroxene can produce derivative magmas with lower CaO, SiO₂, MnO, higher FeO, and higher NiO [Herzberg et al., 2013]. Olivines that then crystallize from such melts at the surface will exhibit depletions in Ca and Mn, elevated Fe/Mn, and elevated Ni. Therefore, olivines that crystallize from peridotite-source magmas that exhibit a prior history of deep “pyroxenite” fractionation can be confused with olivines that crystallize from primary magmas of a pyroxenite source. Modeling

Figure 4. Example of (a) modeled olivine and (b) liquid compositions for Cerro Azul. Figure 4a shows compositions of peridotite partial melts (gray shaded area) and initial pressures of melting (crosses within circles). The green line represents the peridotite/pyroxenite source discrimination line of Herzberg and Asimow [2008]. First step in the modeling procedure, illustrated by the round symbols, is the calculation of the primary magma [Herzberg and Asimow, 2008] by the addition and subtraction of olivine from a given starting composition (Cerro Azul Sample CA-46). In the second step, olivine and liquid compositions (solid lines) are calculated from the primary magma accounting for variable Cpx fractionation. Calculated olivine compositions are plotted against the actual data in order to infer source and crystallizing mineral assemblage.
Figure 5. Olivine phenocryst compositions from (a–d) Cerro Azul and (e–h) Fernandina compared with model olivine compositions crystallized from primary magmas and their derivatives after variable olivine and clinopyroxene fractionation. Primary magma compositions from peridotite sources were calculated from whole rock lava compositions of CA-46 [Naumann et al., 2002] for Cerro Azul and F425 [Allan and Simkin, 2000] for Fernandina. Primary magma compositions for a pyroxenite bearing source (blue lines, Figure 5e–5h) were calculated from whole rock sample G192-1 [Kurz and Geist, 1999] from Santa Cruz. There is a shift from a predominantly peridotite source in the southernmost dredges (D38A, D39G, and D40C) to magmas derived from a hybrid source with a component of pyroxenite closer to the island. Sample D30A has the strongest pyroxenite signal. Blue bars represent the $2\sigma$ error calculated from repeated analyses of San Carlos olivine standard.
the compositions of olivines along the liquid line of descent, allows for the discrimination of olivines derived from a primary magma of a pyroxenitic source and primary magmas from peridotite sources which have fractionated olivine and pyroxenes concurrently.

[15] The method for calculating a liquid line of descent begins with identification of a primary magma composition. Primary magma compositions for peridotite sources have been computed from primitive lava compositions using PRIMELT2 [Herzberg and Asimow, 2008]. Results for lavas from the Galápagos Archipelago and older CLIP lavas have been reported earlier [Herzberg and Asimow, 2008; Herzberg and Gazel, 2009]. Those primary magma solutions that permit olivine calculation and comparison with our observed olivines are reported in Table S2 of the supporting information. Whole rock NiO contents often provide unreliable primary magma NiO contents due to olivine sorting; for primary magmas of peridotite sources, we use Ni (ppm) = 21.6MgO - 0.32Mg² + 0.051Mg³ [Herzberg, 2011b]. From these identified primary magmas, both olivine and clinopyroxene were permitted to crystallize along a liquid line of descent; the method for this calculation was described previously [Herzberg et al., 2013]. The Ol/Cpx proportions have been varied arbitrarily because the exact T-P paths at which melts interact with wall rocks are not known.

[16] An example of the modeling procedure for Cerro Azul is provided in Figure 4. The liquid line of descent is calculated in PRIMELT2 [Herzberg and Asimow, 2008] by adding and subtracting olivine in 1% increments. Starting with Cerro Azul sample CA-46, the resulting calculated liquid compositions (4A) as well as olivine in equilibrium with the liquids (4B) are shown by the blue and red symbols. PRIMELT2 [Herzberg and Asimow, 2008] yields a primary magma solution (green triangle) when ~1.6 wt % olivine is subtracted from the original whole rock composition. Forward modeling of olivine compositions crystallized from this primary magma is shown by the solid lines. Olivine that crystallizes without Cpx is represented by the black line. Co-crystallization of olivine and Cpx results in liquids and olivine compositions that are depleted in Ca relative to the solutions of the model in which Cpx fractionation is absent. As the proportion of Cpx relative to olivine increases, so do the depletions of Ca in the calculated liquids and olivines (colored lines; Figure 4). Fields representing Indian Ocean and Pacific MORB [Sobolev et al., 2007] are included as an example of a peridotite source. At high Mg#, these analyses overlap with predicted primary magmas of Herzberg [2011]. Primitive olivine compositions from Mauna Kea [Sobolev et al., 2007], on the other hand, do not overlap with this field, rather they trend toward a primitive magma that is lower in Ca than is expected from peridotite-source magmas consistent with pyroxenite melting for Mauna Kea. Analyses of olivines from Cerro Azul sample CA-16 fall between the models in which 0–60% Cpx has fractionated from a peridotite-source magma. The same modeling procedure is repeated for Mn, Ni, and Fe. All four plots (Mn, Fe/Mn, Ni, and Ca; Figure 5) yield a reasonably consistent fit between sample CA-16 and the modeled olivine compositions which extrapolate to a peridotite source, primary magma, thus a peridotite-source lithology for Cerro Azul is inferred.

[17] In some cases, observed olivine phenocryst compositions are too high in Ni and Fe/Mn and too low in Ca and Mn to have crystallized from primary and derivative magmas of a peridotite source. We interpret these as pyroxenite source signatures and compare them with computed olivines from a model pyroxenite source. As with peridotite-source magmas, we begin the calculation by computing a primary magma composition from a primitive whole rock analysis. However, instead of using PRIMELT2, which is appropriate for peridotite sources, we add olivine to the candidate lava and backtrack to a set of primary magmas along model pyroxenite cotectic liquid compositions [Herzberg, 2011b, Table A2]. For these pyroxenite-source primary magmas, we have computed the effects of olivine and clinopyroxene fractionation on both derivative liquid compositions and the olivines from which they crystallized. Of all the volcanos covered in this study, results from Santa Cruz show the widest variation between peridotite and relatively pure pyroxenite sources. Distinctive olivine populations for Santa Cruz reveal hybrid magmas mixed from partial melts of peridotite and pyroxenite [Sobolev et al., 2007], and we attempt to model their proportions. Whole rock compositions of most primitive lavas from Santa Cruz [Gibson and Geist, 2010; Saal et al., 2007; White et al., 1993] have low CaO and SiO₂ contents, indicating a low SiO₂-type pyroxenite melt with no restrictions to mixing with peridotite-source melts [Herzberg, 2011b].

[18] Gurenko et al. [2010] provide an equation for inferring the amount of pyroxenite from olivine phenocryst compositions, an algorithm that is a
4. Petrological Results

4.1. Dominantly Peridotite-Source Magmas

One of the most important conclusions of this study is that peridotite is the dominant source lithology that melts in the Galápagos mantle plume. A detailed discussion of each volcano now follows (also see the Appendix).

4.1.1. Cerro Azul (Central Isotopic Domain)

Of all the samples in this study, those obtained from the 1998 eruption of Cerro Azul provide the best example of high Fo olivines crystallized from a peridotite source (Figures 5a–5d). Olivines from five samples are internally consistent with those expected from ~20 to 40% olivine and ~60 to 80% clinopyroxene fractionation of a primary, peridotite-source magma. However, olivines from sample CA-14 are an exception. The extremes of Fe/Mn in sample CA-14 are not consistent with a peridotite source, and olivines with the highest Mg numbers are unusually elevated in Mn and depleted in Ni. As Mn is strongly compatible in garnet [Balta et al., 2011; Herzberg et al., 2013; Pertermann and Hirschmann, 2003], we tentatively interpret the olivines in sample CA-14 to crystallization from a melt that has selectively dissolved garnet.

4.1.2. Fernandina (Central Isotopic Domain)

Olivines from dredge samples taken from the submarine sector of Fernandina indicate that the source lithology of submarine lavas is variable. There is a shift from a predominantly peridotite source in the southernmost dredges (D38A, D39G, and D40A) to magmas derived from a hybrid peridotite–pyroxenite source with a component of pyroxenite closer to the island (Figure 5). Sample D30A has the strongest pyroxenite signal and is similar to olivines from Volcan Ecuador (see section 4.2.2).

4.1.3. Northern Isotopic Domain

Olivines from Wolf Island, Darwin Island, Marchena, and Genovesa (Figures 6a–6d) have compositions consistent with a peridotite source. Unfortunately, the magmas from the northern half of the archipelago are too evolved to calculate primary magma compositions, and model olivine compositions could not be obtained reliably. However, modeled olivine compositions from sample SC-99 from Santa Cruz (see section 4.2.1 for details) provide a reasonable fit for the data and indicate that Cpx fractionation has affected the olivine chemistry. Analyses of olivines from Pinta sample P-24 show clear offsets in Mn, Fe/Mn/Ni, and Ca at high forsterite contents from the other samples of the northern archipelago. When compared to modeled olivines from a hybrid source consisting of 40% pyroxenite and 60% peridotite (see section 4.2.1), it is apparent that these data are more consistent with a source that has a minor pyroxenitic component. Magmas from a peridotite source that have crystallized 20% olivine and 80% Cpx fit the Pinta data for Ni and Ca; however the fit is poor for Mn and Fe/Mn.

4.1.4. Floreana (Southern Isotopic Domain)

Floreana olivines are dominated by peridotite melting, but a significant pyroxenite component is indicated in some samples. Our interpretation of the presence of a pyroxenitic component from this olivine data is consistent with previous work on isotopes and trace elements [Harpp et al., 2013]. A single magnetically reversely polarized sample (FL-03-135) shows the strongest peridotite signal from Floreana (Figures 6e–6h). A wide range of Ca contents in the olivines in this and other Floreana samples is observed, which we interpret as mantle xenocrysts. This interpretation is consistent with previous studies that have indicated that xenocrysts and disaggregated xenoliths are virtually ubiquitous in the magmas of Floreana [Bow and Geist, 1992; Harpp et al., 2013; Lyons et al., 2007]. Anomalously low Ca (<1000 ppm) in the cores of these olivine crystals is followed by an increase in Ca out toward the edges of the crystals. Calcium concentrations at the edges are consistent with the calculated compositions of olivines crystallized from a basaltic magma of a peridotite source. Conversely, there is no drop in Mg# from core to rim corresponding with the increase in Ca. These grains have subtle undulatory extinction and show signs of embayment, which indicates
Figure 6. Olivine phenocryst compositions from the (a–d) northern archipelago and (e–h) Floreana compared with model olivine compositions. Primary peridotite-source magma composition for Floreana was calculated from lava composition FL-26 [Bow and Geist, 1992]. The pyroxenite-source was calculated from Santa Cruz (see section 4.2.1.), and the green crosses are olivines that crystallized from the mixed proportions indicated. Green curves are olivines that crystallize from these mixed magmas by 100% olivine crystallization. The blue curves are model olivine compositions that crystallize from a 40% pyroxenite mix, by fractional crystallization of 40% olivine and 60% clinopyroxene. The drop in Fe/Mn with differentiation is likely caused by magnetite fractionation [Herzberg, 2011b]. Blue bars represent the 2σ error calculated from repeated analyses of San Carlos olivine standard.
disequilibrium between the olivine and the host liquid. It is our interpretation that the elevated Ca contents at the edges of the grains are the result of partial re-equilibration of the olivine xenocrysts with the host magma. Evidence of similar xenocrysts is observed in some of the olivine data from Santa Cruz (Samples GHZ-8, GHZ-9, and GHZ-11, Figure 7d) and San Cristobal (Figure A2).

[24] Normally magnetically polarized, Main Stage [Bow, 1979; Bow and Geist, 1992] Floreana samples (FL-03-01, FL-03-120) have Ni and Ca contents and Fe/Mn that are difficult to attribute to peridotite melting. However, their compositions are compatible with crystallization from a melt that was a mixture of ~60–80% peridotite-source melt, as modeled with FL-26 [Bow and Geist, 1992] and ~20–40% pyroxenite-source melt modeled from Santa Cruz (Figures 6e–6h; see section 4.2.1 for details). The Late Flank Stage [Bow, 1992] reveals the strongest evidence for pyroxenite-source melting with low Ca, high Ni, and high Fe/Mn.

4.1.5. Other Dominantly Peridotite-Source Occurrences

[25] Evidence for the importance of peridotite-source melting is provided by olivine compositions from two lava samples from Volcan Wolf, and one from Alcedo (Appendix). However, our interpretations of a dominantly peridotite source for these volcanoes may be compromised by the limited number of samples available. Additionally, these olivines have low Mg numbers, generally <80, and all petrological models of olivine and clinopyroxene fractionation become less secure for these evolved compositions.

4.2. Dominantly Pyroxenite-Source Magmas

4.2.1. Pyroxenite-Source Magmas in the Eastern Isotopic Domain

[26] In the Galápagos, the greatest variability in source composition as inferred from olivine chemistry is found on Santa Cruz (Figure 7). Unlike the examples discussed above, there is little evidence for a pure peridotite source in magmas from Santa Cruz. Out of 79 lava samples from Geochemistry of Rocks of the Oceans and Continents (GEOROC) [Gibson and Geist, 2010; Saal et al., 2007; White et al., 1993] and Bow [1979], we could only obtain five primary magma solutions for a peridotite source. All other samples were either too evolved, or resulted in a pyroxenite source input error in PRIMELT2 [Herzberg and Asimow, 2008]. Model olivine compositions from a peridotite source, shown in Figure 7, were calculated from SC-99 [Bow, 1979]. It is unfortunate that we do not have olivine data from any of the five possible peridotite-source primary magmas and their derivatives.

[27] Lava sample G192-1 [Kurz and Geist, 1999] was used to provide a primary magma composition of a pyroxenite bearing source. Fractional crystallization of 40–100% olivine and 0–60% clinopyroxene from this primary magma yields derivative magmas that crystallize olivines that are an excellent match to observed olivines in samples GHZ-9 to 11, E-31, and 39.3 (Figure 7). The match, however, is not as good for olivines in other samples that show systematically higher Fe/Mn and Ni, and lower Ca and Mn, evidence that they crystallized from magmas with a larger pyroxenite component. Having the greatest offsets from calculated olivine compositions from a peridotite-source primary magma, we assume that these olivines crystallized from a pure pyroxenite melt, and the primary magma G192-1 is a mixture of primary magmas from the pure peridotite SC-99 and this pure pyroxenite primary magma. We then solve the mass balance equation: G192-1 = X PX + (1–X)SC-99 where PX is the composition of the pure pyroxenite melt, and to this we compute the fractional crystallization of 20–100% olivine and 80–100% clinopyroxene. By iteratively varying X and solving for PX, the computed olivines for a 100% pyroxenite melt provide an excellent match to many observed olivines from Santa Cruz when X = 0.4. Olivines from sample 49.4 are consistent with 100% crystallization of olivine from the pure pyroxenite melt; olivines from sample 34.8 are more consistent with substantial clinopyroxene crystallization from the same pure pyroxenite melt. At the other extreme, olivines in sample GHZ-10 are consistent with crystallization of a melt that was a mixture of ~80% peridotite and 20% pyroxenite (Figure 7).

[28] Evidence for the importance of pyroxenite as a source lithology is also obtained from olivines from Santiago, Volcan Darwin, Española, and Santa Fe (Appendix). Unfortunately, there are fewer samples from these localities available, and a detailed analysis of the relative contributions of peridotite and pyroxenite is not as secure. However, using our analysis of Santa Cruz as a guide, we can infer substantial contributions of pyroxenite for Santiago and Volcan Darwin, and greater contributions of peridotite for San Cristobal and
Figure 7. Olivine phenocryst compositions from (a–d) Santa Cruz and (e–h) Volcan Ecuador, Roca Redonda, and Sierra Negra compared with model compositions crystallized from primary magmas and their derivatives after variable olivine and clinopyroxene fractionation. See text for primary magma compositions for Santa Cruz. Primary magma composition for Volcan Ecuador was calculated from whole rock lava compositions of E-9740 [Geist et al., 2002]; it contains 0.164% MnO, and Figure 7f illustrates how Fe/Mn in olivine compositions propagates with small changes in MnO. Mixing between different proportions of peridotite and pyroxenite-source melts are evident in the Santa Cruz samples. Blue bars represent the 2σ error calculated from repeated analyses of San Carlos olivine standard.
Española (Appendix). Collectively, these volcanoes highlight the importance of pyroxenite melting in the isotopically depleted volcanoes.

4.2.2. Pyroxenite-Source Magmas in the Central Isotopic Domain

[28] Some of the strongest evidence for pyroxenite melting is indicated by olivines in four lava samples from Volcan Ecuador, four samples from Roca Redonda, and one sample from Sierra Negra (Figures 7e–7h). Olivines from all four samples from Volcan Ecuador are essentially identical, and they are compared with model olivines calculated from a pyroxenite-source primary magma. Fractional crystallization of 30–100% olivine and 0–70% clinopyroxene from this primary magma provides a good match to observed olivine compositions from Volcan Ecuador (Figure 7) and is consistent with geochemical evidence of olivine/Cpx fractionation [Geist et al., 2002]. Unfortunately, the substantial uniformity in olivine compositions from Volcan Ecuador does not permit an evaluation of the proportions of pyroxenite and peridotite melt that mixed to yield the primary magma. However, if Santa Cruz is a guide, then the primary magma for Volcan Ecuador may have formed from a mixture of ~40% peridotite and 60% pyroxenite. Results for Roca Redonda are similar, but slightly more pyroxenite melt may have been important for Sierra Negra.

[30] Olivines from 3 of the 4 Roca Redonda picrite samples are similar to those from Volcan Ecuador. Olivine phenocrysts in the samples from Roca Redonda are generally subhedral with somewhat rounded edges and few embayments. All samples with the exception of R952 exhibit extreme zoning, which permeates deep into the olivine crystals. Olivines from sample R952 and a single grain from R959 are anomalous. At Mg numbers of ~80%, olivines in these anomalous samples exhibit strong enrichments in Mn and depletions in Fe/Mn, Ni, and Ca (Figure 7). As Mn is strongly compatible in garnet [Herzberg et al., 2013], we tentatively interpret the trends in Mn and Fe/Mn as reflecting the dissolution of garnet into the liquid from which the olivines crystallized.

5. A Lithological Source Map of the Galápagos Archipelago

[31] A map summarizing the source lithology of the Galápagos plume inferred from the olivine data is presented in Figure 8. Basemaps used for Figures 8 and 11 were produced in GeoMapApp, (http://www.geomapapp.org) [Ryan et al., 2009]. As with samples obtained from presently active volcanoes, the results obtained from inactive volcanoes are assumed to reflect the source lithology of the present-day plume. Paleopositions of older samples were calculated from the equations of Cox and Hart [1986] using available K/Ar ages and the NUVEL-1A Euler vector for the Nazca plate relative to the hot spot [Gripp and Gordon, 2002]. It should be noted that none of the samples used in this study has been dated and exact coordinates is not known for many of the samples. As such, the mapping of the results should be taken as an approximation.

[32] Samples from San Cristóbal come from both the southwestern and northeastern ends of the island. Argon ages obtained from these regions range between 0.05 and 2.35 Ma [White et al., 1993]. With that in mind, the corresponding units inferred from our analysis cover an area spanning the location of the present-day island to ~50 km to the west. K/Ar ages from the Platform Stage [Bow, 1979] of Santa Cruz range from 1.12 to 1.31 Ma [White et al., 1993], which places these flows near the center of the archipelago. Older ages obtained in earlier studies are not considered due to imprecision. Dated flows from Española range from 2.1 to 3.31 Ma [Bailey, 1976] placing the paleoposition of this island in a region southeast of present-day Floreana. Most of the available ages for Santa Fe fall in the range of 2.5–2.85 Ma [Bailey, 1976; White et al., 1993], however, a single younger age of 0.72 [White et al., 1993] makes it impossible to assign our results to a specific region.

6. Lithologic-Isotopic Relations

6.1. Isotope Systematics

[33] Harpp and White [2001] proposed that the isotopic characteristics of lavas from the Galápagos Archipelago can be understood by the mixing of four mantle endmembers: PLUME, DUM, WD, and FLO. Based on inferences drawn from olivine chemistry, we have identified peridotite as the dominant source lithology of the volcanoes which represent these compositional extremes. Furthermore, peridotite is also a likely source lithology for Volcan Wolf, Cerro Azul, and Volcan Alcedo, and these volcanoes have isotope ratios that are more representative of intermediate mixtures of the four endmembers. The implication is that peridotite is the dominant and widespread mixtures of the Galápagos mantle plume, with heterogeneous isotopic compositions.
Before we discuss the implications of the lithology-isotope relations in detail, an important assumption needs to be considered. Ideally we would have provided olivine chemistry data on samples for which isotopic data exist, a truly integrated study. However, few isotopic data exist for the samples in this study, and in some cases our olivine data were acquired from a single sample that may not be representative of the entire volcano. For example, of the 284 samples for which isotopic data have been compiled from GEOROC, we collected olivine chemistry on only five (i.e., two from Volcan Ecuador, two from Cerro Azul, and one from San Cristobal). This problem arose from the lack of available rock chips for which many isotopic studies have been performed, and from our requirement to work on primitive lavas from which we could expect to find olivines with high Mg numbers. Isotopic variations of each volcano are generally quite small when compared to that of the entire archipelago (Figure 9). Exceptions include the two stages of activity for Santa Cruz [White et al., 1993] and Floreana [Harpp et al., 2013], both of which have a considerable range of isotopic compositions. Given the narrow range of isotopic compositions for most of the volcanos, we assume in the discussion that follows that the published isotope ratios represent the isotopic characteristics of the source lithologies we have inferred from olivine chemistry.
6.2. The PLUME Component

[35] A disproportionate amount of Ni is found in the olivines of sample D38A, which is not consistent with olivine expected to crystallize from normal mantle peridotite having 1960 ppm Ni, yet Ca, Mn, and Fe/Mn are consistent with a peridotite source. The apparent correlation of high Ni with high $^{3}$He/$^{4}$He is seen not only in Fernandina but also in samples D28C and D11A from Baffin Island, West Greenland, and Gorgona [Herzberg et al., 2013]. This observation has been interpreted as evidence of input from the core and thus places the high $^{3}$He/$^{4}$He reservoir in the source of some OIBs near or above the core/mantle boundary [Herzberg et al., 2013]. Sample D30A, which has the strongest pyroxenite signal in Fernandina, has a $^{3}$He/$^{4}$He of 26.85, whereas samples D28C and D11A have $^{3}$He/$^{4}$He ratios of 27.71 and 25.67, respectively [Kurz et al., 2009]. We infer a smaller contribution from pyroxenite in samples D28C and D11A thus there is no apparent connection between the proportion of pyroxenite in the source and helium.

Figure 9. Plot of mean Sr, Nd, and Pb isotopic ratios and inter-quartile range for each volcano covered in this study against the isotopic range of the Galapagos Archipelago [Allan and Simkin, 2000; Blichert-Toft and White, 2001; Cullen and Mcbriney, 1987; Cullen et al., 1989; Geist et al., 2002, 2005; Handley et al., 2011; Harpp and White, 2001; Kurz and Geist, 1999; Naumann et al., 2002; Reynolds and Geist, 1995; Saal et al., 2007; Standish et al., 1998; Teasdale et al., 2005; Thompson et al., 2004; White and Hofmann, 1978; White, 1979; White et al., 1993].
[36] Relationships between He isotope ratios and source lithology may be obscured by many variables, such as the contamination by depleted upper mantle thought to pervade the sources of the Galápagos plume [Harpp and White, 2001], and the possibility that helium might diffuse between lithologies [Hart et al., 2008]. Basalts from Mid-ocean ridges are degassed twice, first upon eruption [Craig et al., 1975] and then again during subduction [Moreira and Kurz, 2001]. In this process, both isotopes of helium are lost, however 3He can be replenished by radioactive decay of Th and U. Thus elevated 3He/4He is not expected in recycled oceanic crust. The range (n = 72) of 3He/4He at Fernandina is 10.52–29.53 [Kurz et al., 2009; Raquin and Moreira, 2009]. Volcan Ecuador, which is directly adjacent to Fernandina, has a range (n = 13) of 3He/4He of 8.24–11.42 [Kurz et al., 2009]. This observed decrease in 3He/4He between Fernandina and Volcan Ecuador is consistent with the presence of more pyroxenite below Volcan Ecuador as our results suggest. The proximity of Fernandina to Volcan Ecuador makes it likely that the two volcanoes share a similar recycled source, but in different proportions. If this is the case, then it is the peridotite component in the hybrid magmas of Fernandina that carries the primordial He signal.

6.3. The FLO Component

[37] Having the most radiogenic 206Pb, 208Pb, and 87Sr enriched samples in the archipelago it is surprising that Floreana shows only a small amount of pyroxenite in its olivine chemistry. It has been proposed that recycled crust may be stretched and stirred into mantle peridotite to the point it is no longer present as a separate lithology [Allègre and Turcotte, 1986]. Melts produced from a peridotite source refertilized in such a way would have the isotopic signature of recycled crust imprinted upon it, yet would crystallize olivine phenocrysts with no apparent pyroxenite signal [Gurenko et al., 2009; Herzberg, 2011b]. This mechanism is supported by rare earth element patterns that suggest a high modal concentration of Cpx in the source [Bow and Geist, 1992; Harpp and White, 2001; Harpp et al., 2013].

[38] A reversely polarized Main Stage [Bow, 1979; Bow and Geist, 1992] sample (FL-03-135) has the strongest peridotite signal at Floreana, in contrast with the younger Flank Stage [Bow, 1979; Bow and Geist, 1992] sample (FL-03-93) that has one of the strongest pyroxenite signals (Figures 6e–6h). The Main Stage [Bow, 1979; Bow and Geist, 1992] magmas are also the most enriched in 87Sr and 206Pb isotopes, while the youngest Flank Stage [Bow, 1979; Bow and Geist, 1992] magmas show a relative depletion in radiogenic Pb and Sr (Figure 10). We suggest that the pyroxenite component is derived by the incorporation of a small amount of the pyroxenite from the east into the source of Floreana as it drifted farther from the plume axis over its 1.5 Ma of activity [White et al., 1993]. That is, in the later stages of activity Floreana was closer to the depleted Eastern Pyroxenite Domain, which we have shown to consist of a significant amount of recycled crust, thus it tapped a different part of the plume which included a greater amount of pyroxenite than the earlier stage. Our interpretation of the source of the slight depletion of Floreana in the younger lavas is largely speculative given that: (a) none of the samples in this study have been dated; and (b) none of the isotopic data from Floreana presented here are directly from the samples we have analyzed.

6.4. The Wolf-Darwin (WD) Component

[39] Northward migration of plume material to the Galápagos Spreading Center (GSC) is unequivocal given the geochemical, bathymetric, and seismic data collected at the ridge [Canales et al., 1997; Christie et al., 2005; Detrick et al., 2002; Fisk et al., 1982; Schilling et al., 2003; Sinton et al., 2003; Villagomez et al., 2007] and the geochemistry of the islands Wolf, Darwin, and Pinta [Cullen and Mcbirney, 1987; Harpp and Geist, 2002]. Magmas of the Wolf-Darwin Lineament consist primarily of WD and PLUME with minor amounts of DUM [Harpp and White, 2001]. Given that there is no pyroxenite signal in the olivine chemistry in these samples, if the source of the 207Pb and 208Pb enrichment is recycled materials, they have been mixed into a peridotite host.

6.5. The “Depleted Upper Mantle (DUM)” Component

[40] Depleted in both radiogenic isotopic ratios and trace elements, magmas from Genovesa are virtually indistinguishable from near-ridge seamounts and are more depleted than the plume influenced Galápagos spreading center [Harpp et al., 2003, 2012; Harpp and White, 2001]. Genovesa is also similar to Pacific MORB, and has been interpreted by some as originating from depleted upper mantle [Geist et al., 1988; Harpp and White, 2001; White et al., 1993]. Olivine
chemistry of samples from Genovesa indicates the presence of a peridotite source (Figure 6).

PRIMELT2 modeling for rare peridotite-source lavas from Santiago and Santa Cruz in the Eastern Isotopic Domain [Hoernle et al., 2000] reveals high mantle potential temperatures (Figure 1), consistent with the presence of plume material and inconsistent with entrainment of ambient upper mantle [Geist et al., 1988; Harpp and White, 2001; White et al., 1993]. Additionally, the 90 Ma komatiites from Gorgona are isotopically depleted (Figure 3) and melted from a peridotite source that was hot [Herzberg and Gazel, 2009]. We conclude that both peridotite and pyroxenite contributed to plume volcanism in the Eastern Isotopic Domain [Hoernle et al., 2000], and recommend that the term “DUM component” of Harpp and White [2001] be replaced with “DM” in order to relax the restriction that it originates in the upper mantle.

Figure 10. Sr, Nd, and Pb isotope ratios of (left) peridotite and (right) pyroxenite-source magmas (Sources as in Figure 9) Fernandina, Pinta, Floreana, Genovesa, and islands of the Wolf-Darwin Lineament are thought to be most representative of the isotopic endmembers proposed by Harpp and White [2001]. Western pyroxenite-source volcanoes Volcan Ecuador, Roca Redonda, and Sierra Negra are more enriched isotopic signatures than pyroxenite-source volcanoes Santa Cruz, Darwin, and western Santiago to the east. Santa Fe is not included given the wide span of ages [Bailey, 1976; White et al., 1993] and isotopic compositions [Graham et al., 1993; Thompson et al., 2004; White et al., 1993] which may reflect temporal changes; thus it is not known if it belongs with the Eastern or Western Pyroxenite Domains. San Cristobal is not included because it is apparent that the pyroxenite component is minor in all samples.
6.6. Galápagos Pyroxenite Domains: The Hidden Isotopic Endmembers

[43] Isotopes of pyroxenite-source volcanoes form two, spatially distinct populations, which show little overlap in isotope space (Figures 10d and 10f). Santiago, Santa Cruz, and Volcan Darwin in the Eastern Isotope Domain (Hoernle et al., 2000) have isotopically depleted signatures; Roca Redonda, Volcan Ecuador, and Sierra Negra in the Central Isotope Domain (Hoernle et al., 2000) are relatively enriched in terms of their isotopic ratios. Volcanoes which have the strongest pyroxenite signals have isotopic compositions which do not appear to correspond to any one of the proposed Galápagos endmembers (Harpp and White, 2001, Figure 10). Each pyroxenite source region appears to be mixtures of the PLUME, DUM, and FLO endmembers (Harpp and White, 2001), which we have identified as dominantly peridotite sources. However, the pyroxenitic component found in these two domains could not have formed from the mixing of these endmembers as it is impossible to form a pyroxenite directly from the mixing of two or more peridotite sources. Thus it is likely that there are more than four isotopic endmembers contributing to the magmatism in the Galápagos.

An important outcome of this study is that MORB-like isotopically depleted lavas can melt from either peridotite (e.g., Genovesa) or pyroxenite (e.g., Santa Cruz). The presence of isotopically depleted and relatively enriched pyroxenite-source magmas that have compositions overlapping depleted and enriched peridotite sources in the Galápagos indicates that it is not possible to infer source lithology from isotopes. This conclusion sheds new light on the geodynamic circumstances that were likely required for the incorporation of depleted recycled crust into the Galápagos mantle plume.

7. Peridotite-Pyroxenite Relations

[45] Many models have been proposed in which the melting of peridotite and pyroxenite derived from recycled crust can yield isotopically depleted and enriched OIB, respectively [e.g., Hofmann and White, 1982; Allegre and Turcotte, 1986; Chauvel et al., 1992; Morgan and Morgan, 1999; Ito and Mahoney, 2005; Sobolev et al., 2005]. These models stem from the effects of partial melting and crustal production on fractionating parent/daughter isotopes. However, the expectation of isotopic enrichment correlated to a recycled source is in conflict with integrated olivine chemistry and isotope studies of the Canary Islands (Gurenko et al., 2009). These studies show that more depleted Sr-Nd-Pb isotope ratios are characteristic of pyroxenite-source melting (Gurenko et al., 2009), and more enriched volcanoes are generated from peridotite (Gurenko et al., 2009). Isotopic enrichment associated with peridotite-source magmas in the Canary Islands has been interpreted as the mixing of a HIMU-type recycled crustal component into peridotite (Gurenko et al., 2009). In the Canary Islands, the HIMU-like source is thought to originate from ancient (>2 Ga) subducted crust that has had time to be stirred into mantle peridotite and accumulate radiogenic isotopes. Isotopically depleted pyroxenite-source magmas, interpreted as being derived from younger (<1 Ga) subducted oceanic crust, have not had an adequate residence time in the mantle to be dispersed into peridotite, or develop a radiogenic isotopic signal (Gurenko et al., 2009). The proposed isotopic composition of the young (<1 Ga) pyroxenitic component of the Canary plume (Gurenko et al., 2009) is strikingly similar to the pyroxenite bearing magmas the Eastern Pyroxenite Domain (Figure 10).

[46] As discussed in section 6.1, there are few isotopic data on the samples from the Galápagos Islands for which we obtained olivine chemistry data, and as such the problem of mantle source lithologic–isotopic relations in each domain is unconstrained. That is, we cannot tell whether pyroxenite within each domain is isotopically enriched or depleted relative to peridotite in that domain. Moreover, research on the Canary Islands (Gurenko et al., 2009; Day et al., 2010) is sufficient reason to be skeptical of any model that attributes all cases of isotopic enrichment to the presence of recycled crust as a distinct source lithology. However, an important observation is that all volcanic rocks that plot within the Central and Eastern Isotopic Domains (Hoernle et al., 2000) in the Galápagos overlap little with each other (Figure 10). We therefore infer that the isotope ratios of each domain characterize both peridotite and
pyroxenite, and future work is necessary to resolve their differences. The implication of our conclusion is that differences in the isotopic ratios between the Central and Eastern pyroxenite domains are likely to be the result of two separate bodies of recycled crust with different origins.

Several lines of evidence indicate that the isotopic identities of pyroxenite and peridotite in each pyroxenite domain are not unrelated issues. As the pyroxenite source of basaltic magmas is thought to be a second stage reaction product of mantle peridotite and either subducted oceanic crust [Herzberg, 2011b] and/or the partial melts of subducted oceanic crust [Sobolev et al., 2007, 2005]. The second stage pyroxenite produced will have a major element, trace element, and isotopic compositions that are directly inherited by both lithologies. It is therefore possible that the wide variations in the isotopic compositions of the two pyroxenite domains in the Galápagos are inherited by enriched peridotite sources in the Central Isotopic Domain [Hoernle et al., 2000] and depleted peridotite sources in the Eastern Isotopic Domain [Hoernle et al., 2000]. For example, within the Central Isotopic Domain [Hoernle et al., 2000], Sr, Nd, and Pb isotope ratios of pyroxenite-source lavas from Volcan Ecuador are similar to those of peridotite-source lavas from Cerro Azul (Figure 10). This prospect shifts the problem to requiring an understanding of the origin of the isotopic identities of the host peridotite.

Mixing of recycled crust back into a host peridotite is thought to occur by convective thinning, stretching, recrystallization, and solid state diffusion [Allègre and Turcotte, 1986; Herzberg, 2011b]. Mixing will be limited by low solid state diffusion rates and enhanced by partial melting [Hofmann and Hart, 1978]. There is a growing body of evidence for the existence of partial melt at the base of the lower mantle [Williams and Garnero, 1996; Fiquet et al., 2010; Collerson et al., 2010; Coltice et al., 2011; Herzberg et al., 2013], and we speculate that the formation of stage two pyroxenite by solid state reaction and its possible mixture into its host peridotite [Herzberg, 2011b] may be favored kinetically by the presence of melt. Whether the melt is catalytic or it imparts its chemical signature to the source [Herzberg et al., 2013; Collerson et al., 2010] is the subject of future work. Furthermore, there is the potential that recycled water may play a catalytic role, as there is evidence for aqueous fluid-rock interaction in an independent component analysis of Pb, Nd, and Sr isotope ratios of OIB [Iwamori et al., 2010]. One possible consequence of deep melt/water–catalysis/kinetics is that it might help to promote homogenization of initially heterogeneous peridotite and pyroxenite isotopic sources.

8. A Galápagos Mantle Plume Model

Results of our olivine analyses, coupled with isotopic variations in the archipelago lead us to the conclusion that there are two separate, isotopically distinct pyroxenite domains (Figure 10). We propose that filaments of enriched pyroxenite and peridotite ascend nearly vertically beneath Isabela and Fernandina [Farnetani and Samuel, 2005]. Separate, relatively depleted and perhaps younger filaments of recycled crust ascend below the archipelago, east of Isabela along with depleted peridotite. Although seismic tomography studies have yet to image the Galápagos plume to the core-mantle boundary [Hooft et al., 2003; Montelli et al., 2006; Villagomez et al., 2007, 2011; Zhao, 2007], other lines of evidence suggest that these components of the Galápagos plume may be traced to the deepest parts of the lower mantle [Herzberg et al., 2013].

The presence of OIBs derived from a deep, lower mantle source may be inferred not only from elevated $^3$He/$^4$He [Kurz et al., 2009] and anomalously high Ni contents in olivine [Herzberg et al., 2013], but also by the presence of large low shear velocity provinces (LLSVPs). These low seismic velocity anomalies have been linked to upwellings just above the core-mantle boundary [Garnero and McNamara, 2008; Steinberger and Torsvik, 2012; Thorne et al., 2004]. Many mantle plumes are located above the edges of these thermochemical piles at the core-mantle boundary [Steinberger and Torsvik, 2012; Thorne et al., 2004]. It has been shown that the Galápagos plume may be near the eastern edge of a LLSVP beneath the Pacific [Steinberger and Torsvik, 2012]. Moreover it has been suggested that the spatial variations in geochemistry exhibited by the Hawaiian, Kerguelen, and Tristan da Cunha hot spots [Weis et al., 2011] and the Galápagos and Easter hot spots [Harpp et al., 2012] may be the result of the sampling of two different reservoirs at the edges of LLSVPs. For Hawaii, more enriched material constituting Loa trends is inferred to be derived from the LLSVP, whereas the more depleted Kea trend is may be derived from more depleted, ambient lower mantle [Weis et al., 2011]. A similar conclusion may be drawn in the Galápagos, as the
LLSVP is located west of the Archipelago and the more enriched magmas are generally erupted from volcanoes in the west and south. Therefore the location of the Galápagos hot spot relative to the LLSVP is not only evidence for a deeper origin than indicated by seismic tomography, but may also provide an explanation for the geochemical/petrologic variations.

[51] Mantle plume dynamics and plume ridge interaction have been the subject of many experimental studies and numerical models [e.g., Farnetani and Richards, 1996; Griffiths and Campbell, 1990; Ito et al., 1997; Richards and Griffiths, 1989]. Given the compositional diversity of the Galápagos plume, studies dealing with the entrainment of ambient mantle [Geist et al., 1988; Griffiths and Campbell, 1990; Harpp and White, 2001; Richards and Griffiths, 1989; White et al., 1993] are most pertinent to this discussion. Early experimental studies support the entrainment of ambient mantle into an ascending plume head [Griffiths and Campbell, 1990]. The later tail stage is thought to undergo little contamination during ascent through the mantle until it is sheared laterally [Griffiths and Campbell, 1990; Richards and Griffiths, 1989]. The Galápagos plume is currently in the tail stage, and it is subject not only to shear imposed by the eastward motion overriding Nazca plate [Gripp and Gordon, 2002] but it is also evident that plume material is migrating northward to the GSC [Ito et al., 1997; Morgan, 1978]. Results of recent numerical models suggest that the tail stage does not significantly entrain ambient mantle during ascent at any depth or stage [Farnetani et al., 2002].

[52] If the numerical modeling of Farnetani et al. [2002] and Farnetani and Richards [1996] are applicable, then a mantle plume origin for young depleted recycled crust in the Eastern Pyroxenite Domain would require recycled crust to subduct to the lower mantle, become thermally buoyant, and rise within the plume conduit before acquiring an enriched isotopic signature (Figure 11). A study of melt inclusions in Mauna Loa [Sobolev et al., 2011] determined that the pyroxenitic source of these magmas have been derived from young recycled oceanic crust (200–650 Ma) based on 86Sr/87Sr systematics. If the recycling of oceanic crust over such short time scales is not an isolated case, as suggested for the Canary plume [Gurenko et al., 2009, 2013] and the Hawaiian plume [Sobolev et al., 2011], then it provides a plausible mechanism for the presence of the depleted eastern pyroxenitic domain without invoking a model of shallow entrainment [Geist et al., 1988; Harpp and White, 2001; White et al., 1993].

9. Conclusions

[53] We have analyzed the contents of Ni, Ca, and Mn in olivine phenocrysts from volcanoes in the Galápagos Archipelago to infer the mantle source lithologies that melted to produce them. Results show that peridotite is the dominant source
lithology for Fernandina, Floreana, Genovesa, Wolf Island, and Darwin Island, and these volcanoes largely characterize the PLUME, WD, FLO, and DUM isotopic endmembers of Harpp and White [2001]. We also find that peridotite is the dominant source lithology for Volcan Wolf, Alcedo, Marchena, and Cerro Azul, and that these have isotopic compositions that can be generated by mixing the four endmembers of Harpp and White [2001]. Minor pyroxenite melting contributes to Fernandina and Floreana. Peridotite is therefore the dominant melting lithology in the Galápagos mantle plume.

[54] Pyroxenite melting is significant in two spatially separated domains that are also isotopically distinct from one another. The western pyroxenite domain is comprised of Roca Redonda, Volcan Ecuador, and Sierra Negra; and the eastern pyroxenite domain that includes Santiago, Santa Cruz, Santa Fe, and Volcan Darwin on Isla Isabela. The western and eastern pyroxenite domains have the isotopic characteristics of the enriched Central and depleted Eastern domains of Hoernle et al. [2000]. One implication of this discovery is that the two pyroxenite domains likely represent two separate bodies of recycled crust within the Galápagos mantle plume. We propose that the distribution of these pyroxenite bodies is consistent with numerical models of stretched filaments in the plume [Farnetani et al., 2002; Farnetani and Richards, 1996]. None of the pyroxenite-source volcanoes have isotopic compositions that are representative of any one of the four endmembers of Harpp and White [2001]. Instead the volcanoes of both pyroxenite domains appear to be mixtures of the PLUME, DUM, or FLO endmembers. Because it is not petrologically possible to form a pyroxenite by mixing peridotite sources, the apparent mixing relationship is an illusion.

[55] Isotopically enriched and depleted domains of the archipelago melted from both peridotite and pyroxenite, and there is no apparent relationship between source lithology and its isotopic characteristics. Basaltic magmas with an enriched isotopic signal may indicate the presence of recycled crust in the source, however if the pyroxenite is disaggregated within a peridotite host, the petrologic signal can be lost. Melts produced from the resulting refertilized peridotite would fractionate olivines that only have a peridotite-source signal. Our discovery of a pyroxenite source with depleted MORB-like isotope ratios suggests that the reservoir may have formed from recycled crust that experienced a residence time in the mantle too short to develop an enriched isotopic signal. The identification of peridotite-source melting in volcanoes with isotopic characteristics that have been attributed to recycled crust (i.e., Floreana and the Wolf-Darwin Islands) points to the importance of mixing in OIB genesis, in agreement with studies on the Canary Islands [Gurenko et al., 2009]. Mixing of crust back into its host peridotite during subduction and return in a mantle plume may be facilitated by deep partial melting and recycled water in the lower mantle.

Appendix

Sources of Other Galápagos Volcanoes

A1. Volcanoes of Northern Isabela Island

[56] Analyses of olivines from Volcan Darwin indicate the presence of a hybrid source consisting of a mixture of 60–80% pyroxenite melt and 20–40% peridotite melt (Figures A1a–A1d). Olivines from Volcan Wolf and Alcedo, located just north and south of Volcan Darwin, respectively, appear to be derived from a source that is predominantly peridotite with signs of significant amounts of Cpx fractionation (Figures A1e–A1h). Due to the evolved nature of the magmas, olivine compositions could not be modeled directly from whole rock data from these volcanoes. Instead the data presented in the diagrams of Figures A1a–A1d are plotted against calculated olivine compositions from Santa Cruz [Bow, 1979; Kurz and Geist, 1999]. Modeled olivine compositions from a peridotite source in Figures A1e–A1h were calculated from Fernandina sample F425 [Allan and Simkin, 2000]. The lack of consistency between the Mn and Fe/Mn plots and the Ni and Ca plots in Figures A1e–A1h is likely due to the fact that the sample used in the modeling is from a different volcano. Although the sources of Wolf and Darwin appear to be quite different, isotopic data [White et al., 1993] indicate that isotopically they are similar. Because few samples were available for analysis, it is possible that the results presented in Figure A1 are not fully representative of the source variations of these three volcanoes.

A2. Volcanoes of the Central Archipelago

[57] Olivine analyses from Española, San Cristobal, Santa Fe, and Santiago are plotted against calculated olivine compositions from Santa Cruz samples (see supporting information Table S2). Analyses of olivines from the Española sample indicate a source that is predominantly peridotite, however, a small amount of pyroxenite may be present (Figures A2a–A2d). Given the ages of Española [Bailey, 1976; White et al., 1993], it was probably just south/south west of the present location of Floreana when this magma erupted. Thus
the depleted nature of Española relative to Floreana [White et al., 1993] may simply reflect a larger component of depleted mantle in its source due to this island being closer to the periphery of the plume.

Results from seven San Cristobal samples indicate that the source of these flows consists of no more than \(20\%\) pyroxenite melt. Many of these samples show a remarkable similarity to the analyses from the

Figure A1. Olivine phenocryst compositions from (a–d) Volcan Darwin and (e–h) Volcans Wolf and Alcedo. Model olivine compositions presented in Figures A1a–A1d were calculated from whole rock compositions from Santa Cruz [Bow, 1979; Kurz and Geist, 1999]. Peridotite-source olivine compositions presented in Figures A1e–A1h were calculated from Fernandina sample F425 [Allan and Simkin, 2000]. Despite the relatively low Fo of the olivines present, a strong pyroxenite signal is seen in Volcan Darwin. A peridotite source is indicated by the olivine analyses from Volcan Wolf and Alcedo with variable amounts of Cpx fractionation. Blue bars represent the 2\(\sigma\) error calculated from repeated analyses of San Carlos olivine standard.
Española sample (Figures A2a–A2d). Samples analyzed in this study come from the western half (older) and eastern half (younger) of the island. Although no direct ages exist for these magmas, given the geographic distribution, our data suggest that little if any change has occurred in the source of San Cristobal with time. This position is bolstered by the observation of only subtle variations in the chemistry of San Cristobal magmas [Geist et al., 1986] from the oldest to the most recent flows.

Figure A2. Olivine phenocryst compositions from (a–d) Española, San Cristobal, and Santa Fe and (e–h) Santiago (E-H). Model olivine compositions presented in Figure A2a–A2d were calculated from whole rock compositions from Santa Cruz [Bow, 1979; Kurz and Geist, 1999] (see supporting information Table S2 for details). A significant pyroxenite component is seen in the olivines of Santa Fe, while San Cristobal and Española appear to have a very minor component of recycled crust in their sources. Magmas derived from both a relatively pure pyroxenite source (SO-1) and a mixture of pyroxenite and peridotite source melts (E-76) is observed in Santiago. Blue bars represent the 2σ error calculated from repeated analyses of San Carlos olivine standard.
[59] Both samples analyzed from Santa Fe show a significant component of pyroxenite in the source of this volcano (Figures A2a–A2d). Difficulty arises when attempting to attribute the pyroxenite component in this volcano to either the Eastern or Western Pyroxenite Domains. Data presented in White et al. [1993] indicate that older magmas erupted between 2.76 and 2.50 Ma have relatively unradiogenic isotopic ratios whereas a single sample dated at 0.72 Ma has slightly more radiogenic isotopic ratios. This is quite unusual as the older magmas erupted when Santa Fe was very close to present-day Sierra Negra, yet their isotopic compositions bear little resemblance to this volcano. The youngest Santa Fe sample has \(^{87}\text{Sr} / ^{86}\text{Sr}, ^{142}\text{Nd} / ^{144}\text{Nd}, \) and \(^{207}\text{Pb} / ^{206}\text{Pb} \) that are well within the range of the Sierra Negra samples, however at the time of eruption Santa Fe would have been much closer to present-day Santa Cruz. These limited age and isotopic data from Santa Fe suggests that localized variations in the source of the Galápagos plume may occur on time scales that are on the order of a few million years, which is in agreement with the recent study of Floreana conducted by Harpp et al. [2013].

[60] Santiago sample SO-1 appears to have a large component of recycled crust in its source, whereas sample E-76 may have been derived from a source with more peridotite (Figures A2e–A2h). E-76 is from Buccaneer Cove in the western half of the island, and SO-1 is from Sullivan Bay in the eastern corner of the island. This is unusual given that the magmas in the eastern half of Santiago generally have more depleted isotopic and incompatible trace element abundances [White et al., 1993; Gibson et al., 2012]. An extensive geochemical study has shown that variations in the mantle beneath Santiago may occur over kilometer length scales. In order to determine if a correlation exists between the geographic variations in the geochemistry of Santiago [White et al., 1993; Gibson et al., 2012] with source lithology, the analysis of olivines from a larger suite of samples needs to be performed.

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