A radiogenic isotopic (He-Sr-Nd-Pb-Os) study of lavas from the Pitcairn hotspot: Implications for the origin of EM-1 (enriched mantle 1)

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Abstract

We present new He-Sr-Nd-Pb-Os isotopic compositions and major and trace-element concentrations for ten subaerially-erupted lavas and one seamount lava associated with the Pitcairn hotspot. The most geochemically-enriched lavas at the Pitcairn hotspot have signatures that are consistent with recycled sediments derived from upper continental crust. Pitcairn lavas have elevated Ti, which also supports the presence of a mafic protolith in the Pitcairn mantle. A subset of Pitcairn seamount samples, including the seamount sample presented here, are tholeiitic. Tholeiitic lavas are uncommon at ocean hotspots located far from mid-ocean ridges. Like tholeiites that erupted in Hawaii, the presence of tholeiites in the Pitcairn magmatic suite can be explained by melting a silica-saturated recycled mafic component in the Pitcairn mantle source. We also present the highest ⁴He/³He ratio (12.6 Ra, ratio to atmosphere) from the Pitcairn hotspot. This sample anchors the high ²⁰⁶Pb/²⁰⁴Pb portion of the Pitcairn array and provides evidence for a plume component in the Pitcairn mantle. In contrast, Pitcairn lavas that have the lowest ²⁰⁶Pb/²⁰⁴Pb are the most geochemically enriched, and have the highest ⁸⁷Sr/⁸⁶Sr and lowest ¹⁴³Nd/¹⁴⁴Nd in the Pitcairn suite; these EM-1 end-member lavas have MORB-like ³He/⁴He (~ 8 Ra, ratio to atmosphere). Recycled oceanic crust and sediment suggested to be in the Pitcairn EM-1 mantle are expected to have low ³He/⁴He (~ 0.1 Ra). Therefore, the higher, MORB-like ³He/⁴He in Pitcairn EM-1 lavas is paradoxical, but might be explained by diffusive exchange of helium, but not the heavy radiogenic isotopes, with the ambient mantle over billion-year timescales.

1. Introduction

Geochemical analyses of ocean island basalts (OIB) erupted at hotspots reveal that the Earth's mantle is compositionally heterogeneous (e.g., Gast et al., 1964; Hofmann, 1997, 2003; Stracke, 2012; White, 2010; Zindler and Hart, 1986). Compilations of radiogenic isotopic measurements made on OIB show that several distinct isotopic compositions emerge from the global dataset (Zindler and Hart, 1986), including EM-1 (enriched mantle 1), which is characterized by the unradiogenic ²⁰⁶Pb/²⁰⁴Pb ratios with moderately high ²⁰⁸Pb/²⁰⁴Pb and ⁸⁷Sr/⁸⁶Sr, EM-2 (enriched mantle 2) which is characterized by intermediate ²⁰⁶Pb/²⁰⁴Pb ratios and highly radiogenic ⁸⁷Sr/⁸⁶Sr, and HIMU (high ³He/³²He or high ²³⁸U/²³⁵U) component in the oceanic mantle. Geochemically-depleted compositions are also sampled by hotspots, and the origin of the depleted component in plumes is not well understood and may host elevated (primordial) ³He/⁴He ratios (Farley et al., 1992; Hanan and Graham, 1996; Hart et al., 1992). Mixing of the EM-1, EM-2 and HIMU end-members with the depleted composition is thought to generate much of the diversity in OIB.

While the field of mantle geochemistry has advanced to a state of careful description and classification of the various mantle species, and their possible mixing relationships, the origin of these species still remains poorly understood. Models explaining the sources of the mantle end-members often call for subduction injection of surface materials, including continental crust, oceanic crust, and sediments into the mantle over geologic time (Hofmann and White, 1982). Following mixing with ambient mantle, the subducted protoliths are sampled by regions of upwelling mantle, called plumes, and melted beneath hotspots. Alternative hypotheses for the origin of the various end-members include delamination of subcontinental lithosphere (Mahoney et al., 1991; McKenzie and O'Nions, 1983), CO₂-flux induced melting of pristine mantle domain in the lower mantle (Collerson et al., 2010), and more exotic metasomatic processes, where fluids in the mantle impart enriched geochemical signatures on specific domains of the mantle (e.g., Geldmacher et al., 2008; Menzies, 1983; Menzies and Murthy, 1980; Niu and O'Hara, 2003; Pilet...
et al., 2011; Salters and Sachi-Kocher, 2010; Workman et al., 2004). However, a consensus is emerging for the origin of EM-2 as recycled marine sediment (Workman et al., 2008) with a terrigenous origin (Jackson et al., 2007). Similarly, the long-standing hypothesis that HIMU represents recycled oceanic crust (Eiler et al., 1997; Hofmann and White, 1982; Zindler et al., 1982) is gaining strength (Cabral et al., 2013; Hanuš et al., 2011, 2014; Kawabata et al., 2011).

Among all mantle end-members, no consensus has emerged for the origin of EM-1 (e.g., Hofmann, 1997; Hart, 2011) and yet numerous mechanisms have been proposed (e.g., Blichert-Toft et al., 1999; Brandenburg et al., 2008; Chauvel et al., 1992; Collerson et al., 2010; Eiler et al., 1995; Eisele et al., 2002; Escrig et al., 2004; Gasperini et al., 2000; Geldmacher et al., 2008; Hanan et al., 2004; Honda and Woodhead, 2005; Lassiter and Hauri, 1998; Mahoney et al., 1991; Rehkämper and Hofmann, 1997; Salters and Sachi-Kocher, 2010; Tatsumi, 2000; Weaver, 1991; Willbold and Stracke, 2010; Woodhead and Devey, 1993; Woodhead and McCulloch, 1989). Here, armed with a new geochemical dataset from the Pitcairn hotspot, we argue for the presence of recycled sediment in the Pitcairn EM-1 mantle (e.g., Eisele et al., 2002; Honda and Woodhead, 2005), and we suggest that Pitcairn lavas also sample recycled oceanic crust. We also report new helium data on Pitcairn lavas, including the highest 3He/4He ratio from Pitcairn, and the new data provide insights into the evolution of the poorly-understood high 3He/4He mantle and EM-1 mantle reservoirs.

2. Methods

The ten subaerial lavas from Pitcairn island reported here were collected by Ken Farley during the 1989 Helios Expedition. We do not report ages for the samples in this study, but subaerial lavas from Pitcairn have ages between 0.4 and 1.0 Ma (Duncan et al., 1974). Fig. 1 shows locations for the subaerial samples presented in this study. Based on comparison with the geological formations as mapped by Carter (1967), the samples in this study were collected from the Tedside (Pit-1, Pit-3, Pit-4, Pit-7, Pit-8) and Pulawana volcanic formations (Pit-6, Pit-16, Pit-11, Pit-12, Pit-13). Samples Pit-6, 12, 13 and 16 were collected in place from outcrops. Samples Pit-1, 3, 4a, 7, 8 and 11 were collected as cobbles on the wave cut terrace; it is very likely that cobbles on the terrace derive from the overlying cliff as there are no rivers to transport cobbles and longshore transport is not plausible.

We also report new geochemical data on a single submarine sample, 65DS-4, dredged between 1503 and 1276 m water depth on Adams Seamount (25°21'26", 125°18'59"), located ~90 km ESE of Pitcairn. This sample was recovered aboard the FS Sonne on December 1989 on the 65th cruise of this vessel (Stoffers et al., 1990; Woodhead and Devey, 1993). The seafloor extending ~90 km ESE of Pitcairn Island is populated by a submarine volcanic field hosting approximately 90 volcanic cones and seamounts, including Adams Seamount (Hekinian et al., 2003), the largest seamount in this volcanic field (Hekinian et al., 2003; Woodhead and Devey, 1993). Several of these volcanic features exhibit evidence of being active, and submarine dredges returned samples with fresh volcanic rocks and glass; there was little evidence for sediment cover (Stoffers et al., 1990). Seafloor backscatter images revealed that several of the volcanic edifices are highly reflective, further supporting a young age for these seamounts, and substantiating the hypothesis that these seamounts (including Adams Seamount) mark the active portion of the Pitcairn hotspots (Hekinian et al., 2003; Stoffers et al., 1990; Woodhead and McCulloch, 1989). K/Ar age determinations of lavas recovered from Adams Seamount range from 2 to 7 kyr (Guillou et al., 1997) and volcanic rocks recovered from this seamount exhibit evidence of only slight alteration (Hekinian et al., 2003). Additionally, geochemical data obtained on seven dredges of Adams Seamount, including new data presented here on sample 65DS-4, indicate that this seamount has a Pitcairn hotspot pedigree (Woodhead and Devey, 1993). Given this significant body of evidence, we adopt the finding that Adams Seamount marks the active end of the Pitcairn hotspot.

Cut rock slabs were crushed in plastic bags to avoid contamination with metal from the hammer, and the freshest chips were separated and powdered in agate. With the exception of sample 65DS-4, major elements and a subset of trace elements were measured on powders by XRF in the laboratory of Michael Rhodes at the University of Massachusetts Amherst, and the methods and associated errors are reported in Rhodes (1988). The remaining trace elements were measured by ICP-MS at the Geoanalytical Lab at Washington State University (WSU) (Knaack et al., 1994). For sample 65DS-4, major and trace elements were measured at WSU by XRF and ICP-MS (analytical methods and errors are reported in Johnson et al. (1999); Knaack et al. (1994)). At WSU, precision for the analyses of SiO₂, Al₂O₃, TiO₂ and P₂O₅ in basalts is 0.11 - 0.33% (1σ) of the amount present, and 0.38 - 0.71% (1σ) of the amount present for FeO, MgO, CaO, Na₂O, MnO and K₂O (Johnson et al., 2000; Geldmacher et al., 2008; Hanan et al., 2004; Honda and McCulloch, 2005; Lassiter and Hauri, 1998; Mahoney et al., 1991; Willbold and Stracke, 2010; Woodhead and Devey, 1993; Woodhead and McCulloch, 1989). Here, armed with a new geochemical dataset from the Pitcairn hotspot, we argue for the presence of recycled sediment in the Pitcairn EM-1 mantle (e.g., Eisele et al., 2002; Honda and Woodhead, 2005), and we suggest that Pitcairn lavas also sample recycled oceanic crust. We also report new helium data on Pitcairn lavas, including the highest 3He/4He ratio from Pitcairn, and the new data provide insights into the evolution of the poorly-understood high 3He/4He mantle and EM-1 mantle reservoirs.

Fig. 1. Map of Pitcairn island with sample locations from this study. The subaerial samples, and the single submarine sample, were collected prior to GPS availability. The submarine sample 65DS-4 was obtained from volcano 1 located ~90 km ESE of Pitcairn.
Table 1
Whole rock major, trace and isotope analyses for Pitcairn samples from this study, and Os-isotopic analyses on olivines.

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<th>Sample</th>
<th>SiO2 (wt%)</th>
<th>CaO</th>
<th>MgO</th>
<th>Al2O3</th>
<th>FeO</th>
<th>FeOT</th>
<th>TiO2</th>
<th>Na2O</th>
<th>K2O</th>
<th>MnO</th>
<th>Nb</th>
<th>Y</th>
<th>Ho</th>
<th>Tb</th>
<th>Gd</th>
<th>Eu</th>
<th>Nd</th>
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</table>

Data in italics are previously published in Eiler et al. (1995; 1997) where NA indicates "present but not analyzed." *He/He on sample PIT-16, was measured twice, on coarse (11.8 Ra; 4.3*10^-6 cc STP/g) and fine (13.3 Ra; 5.5*10^-6 cc STP/g) olivine fractions, to verify the elevated *He/He ratio.
$^{86}$Sr/$^{87}$Sr = 0.1194, and $^{87}$Sr/$^{88}$Sr ratios were adjusted to an SRM987 value of 0.710240. The external precision for the $^{87}$Sr/$^{86}$Sr ratio is estimated to be 45 ppm, and is based on repeat analyses of the SRM987 Sr standard. Nd-isotopic compositions were fractionation corrected relative to a $^{143}$Nd/$^{144}$Nd value of 0.7219. The $^{143}$Nd/$^{144}$Nd ratios were adjusted to a value of 0.511847 for the La Jolla Nd standard. The external precision on the $^{143}$Nd/$^{144}$Nd is estimated to be 40 ppm, and is based on repeat measurements of the La Jolla Nd standard. Pb isotopic data for the subaerial samples were previously reported for all samples in Eiler et al. (1995), except for sample Pit-6, which is reported here; analytical procedures and fractionation correction are the same as outlined in Hauri and Hart (1993), and Pb-isotopic ratios for sample Pit-6 were adjusted to SRM981 values from (Todt et al., 1996) to be consistent with the data reported in Eiler et al. (1995). Sr, Nd and Pb chemical separation protocols and isotopic analyses for the submarine lava, 65DS-4, follow those outlined in Hart and Blusztajn (2006); Sr, Nd and Pb isotopic measurements were made on the Neptune multi-collector ICP-MS at WHOI.

The new isotopic data are presented in Table 1, together with major element abundances for olivine phenocrysts from subaerial Pitcairn samples. Samples were dissolved by HF–HCl dissolution, subjected to a gentle dry down at <100 °C, then loaded in Carius Tubes as described in Shirey and Walker (1995); chemical separation of Os is as described in Shirey and Walker (1995). The isotopic analyses were made at DTM by negative thermal ionization mass spectrometry (N-TIMS) (Hauri and Kurz, 1997). In-run precision was better than 0.1% (2σ standard error of the mean) on the $^{187}$Os/$^{188}$Os ratio for all runs. Total procedural blanks were <5 pg Os and were within analytical errors. Analyses of the DTM osmium standard yielded an average $^{187}$Os/$^{188}$Os of 0.11378 ± 10 (2σ, n = 5). Helium isotopic compositions were measured on 0.9 to 3.8 g of olivine (on coarse (11.8 Ra) and fine (13.3 Ra) olivine fractions (the average value of the two measurements, 12.6 Ra, is used hereafter), to verify the elevated $^{3}$He/$^{4}$He ratio; the raw ratios were 105 and 11.5 Ra, so even without blank correction the sample is unusual. This sample was collected on a sea-cliff, where erosion enhanced by continuous wave action precludes long-term exposure at the surface, which enables us to exclude a cosmogenic helium origin for the elevated isotope ratio in this sample.

The new isotopic data are presented in Table 1, together with previously published data on these samples. Additionally, in Table 2 we report major element abundances for olivine phenocrysts from submarine sample 65DS-4; these data complement previously-published olivine compositional data on subaerially-erupted lavas from the hotspot (Eiler et al., 1995). Microprobe analyses were made on Cameca SX-100 at UC Santa Barbara with the following operating conditions: 15 kV accelerating voltage, 15 nA beam current and 1 μm beam diameter.
the triangle outlining the Pitcairn dataset in Pb-isotope space in Fig. 4), and this component is characterized by lower $^{208}\text{Pb}/^{204}\text{Pb}$ at a given $^{206}\text{Pb}/^{204}\text{Pb}$. Curiously, this third component is only visible in $^{208}\text{Pb}/^{204}\text{Pb}$ versus $^{206}\text{Pb}/^{204}\text{Pb}$ isotope space, and is not evident in other isotope projections. Sample 65DS-4 defines the end-member composition for this new component, and it plots closer to Hawaiian Koolau lavas in Pb-isotopic space than any previously examined Pitcairn lava. We emphasize that a subset of prior analyses of lavas from the Pitcairn hotspot have Sr, Nd and Pb isotopic compositions that are similar to 65DS-4, but this sample slightly expands the field of the Pitcairn hotspot in $^{208}\text{Pb}/^{204}\text{Pb}$ versus $^{206}\text{Pb}/^{204}\text{Pb}$ isotopic space.

Fig. 5 shows $^{3}\text{He}/^{4}\text{He}$ ratios in olivine in subaerial samples (reported here for the first time) and seamount glasses [reported in] (Honda and Woodhead, 2005), plotted against $^{206}\text{Pb}/^{204}\text{Pb}$. At a given $^{206}\text{Pb}/^{204}\text{Pb}$ ratio, the $^{3}\text{He}/^{4}\text{He}$ of the olivines is bracketed by the $^{3}\text{He}/^{4}\text{He}$ of the glass dataset; only the Pit-16 olivine $^{3}\text{He}/^{4}\text{He}$ measurement is higher than any of the $^{3}\text{He}/^{4}\text{He}$ measurements on glasses. The $^{3}\text{He}/^{4}\text{He}$ of the Pitcairn olivines presented here show no evidence for reduced $^{3}\text{He}/^{4}\text{He}$ by radiogenic ingrowth of $^{4}\text{He}$. Fig. 5 also shows a possible relationship between $^{206}\text{Pb}/^{204}\text{Pb}$ and $^{3}\text{He}/^{4}\text{He}$, where the highest $^{3}\text{He}/^{4}\text{He}$ ratio is found in the sample with the highest $^{206}\text{Pb}/^{204}\text{Pb}$. In comparison, the two lavas with the lowest $^{206}\text{Pb}/^{204}\text{Pb}$ have the lowest $^{3}\text{He}/^{4}\text{He}$ ratios, 7.7 Ra (measured in a glass) and 8.0 Ra (measured in olivine), indicating that EM-1 lavas at Pitcairn have MORB-like $^{3}\text{He}/^{4}\text{He}$. Fig. 6 shows $^{187}\text{Os}/^{188}\text{Os}$ ratios and Os concentrations measured on olivines from samples from this study compared to the whole rock measurements of Pitcairn lavas from previous studies (Eisele et al., 2002; Reisberg et al., 1993). $^{187}\text{Os}/^{188}\text{Os}$ ratios in olivines from this...
study range from 0.1342 to 0.1432, which falls within the range previously identified in whole-rock measurements of the most Os-rich Pitcairn samples (0.131 to 0.148 for samples with Os > 50 pg/g) (see Fig. 2b in Eisele et al. (2002)). As observed by Eisele et al. (2002), there are no clear relationships between Os isotopic ratios and the other radiogenic isotopic compositions presented in this study. Rhenum was not measured on the olivines, so an age-correction cannot be performed. However, recent studies on magmatic olivines from Samoa (Jackson and Shirey, 2011), Iceland (Debaille et al., 2009) and the Cook Islands (Hanyu et al., 2011) indicate that Re/Os ratios in magmatic olivines are low, and the subaerial lavas from Pitcairn are young, so any age-correction to the $^{187}$Os/$^{188}$Os ratio of the olivine will be small.

Fig. 4. Isotopic data for Pitcairn lavas reveal a typical EM-1 signature: low $^{143}$Nd/$^{144}$Nd, moderately high $^{87}$Sr/$^{86}$Sr and highly unradiogenic $^{206}$Pb/$^{204}$Pb. The plot of $^{208}$Pb/$^{204}$Pb against $^{206}$Pb/$^{204}$Pb reveals the presence of a third component in Pitcairn lavas, which is the second component that is highlighted in the dashed triangle. The third component (called low-$\kappa$ EM-1) is anchored by geochemically-more depleted rejuvenated lavas. Data fields for Samoa and Hawaii (including Koolau volcano) are shown with the global OIB database, and these data are taken from Georoc database and verified with the original references. The Pitcairn isotopic data are from the following sources: (Eisele et al., 2002; Honda and Woodhead, 2005; Woodhead and Devey, 1993; Woodhead and McCulloch, 1989; Woodhead et al., 1993).

Fig. 5. $^{3}$He/$^{4}$He ratios and He concentrations in olivine (in subaerial samples, reported here for the first time) and seamount glasses (reported in Honda and Woodhead, 2005). Only the data for glasses with $^{4}$He $> 10^{-7}$ cc STP/g are plotted in the right panel. The line in the left panel connects two replicate measurements on coarse and fine olivine fractions (sample PIT-16). The diagram to the right shows that the sample with the highest $^{3}$He/$^{4}$He ratio (which is shown as the average of two measurements of this sample, 11.8 Ra and 13.3 Ra, measured on coarse and fine olivine fractions, respectively) also has the weakest EM-1 signature and instead trends toward the depleted mantle component (FOZO?) sampled by Pitcairn lavas. The two lavas with the lowest $^{206}$Pb/$^{204}$Pb have the lowest $^{3}$He/$^{4}$He ratios, 7.7 Ra (measured in a glass) and 8.0 Ra (measured in olivine), indicating MORB-like $^{3}$He/$^{4}$He signature in the EM-1 mantle.

Fig. 6. The new olivine $^{187}$Os/$^{188}$Os measurements in this study fall in the same range as identified in the most Os-rich whole rocks (>50 pg/g; Eisele et al., 2002) and support the previous observations that the EM-1 end-member has radiogenic Os, which is consistent with a recycled origin for EM-1 lavas in Pitcairn (Eisele et al., 2002).
geochemical feature of EM-1 as sampled by Pitcairn lavas: 1. Pitcairn
lavas exhibit a range of highly unradiogenic 206Pb/204Pb ratios associ-
eted with relatively high 208Pb/204Pb, and 2. The most geochemically-
enriched Pitcairn lava with Hf-isotopic data departs from the global
array formed by 143Nd/144Nd and 176Hf/177Hf and trends toward a com-
ponent with high 176Hf/177Hf at a given 143Nd/144Nd (Eisele et al., 2002).
The former geochemical characteristic indicates that the protolith con-
tributing to the enriched mantle source beneath Pitcairn must also
have high Th/U (Eisele et al., 2002). Sediments can have high Th/U,
and pelagic clays have among the highest Th/U, as they are the
weathering products of mature upper continental crust (where U has
been preferentially mobilized relative to Th over time) (Plank and
in pelagic sediments will generate Pb-isotopic systematics appropriate
for Pitcairn hotspot lavas over time. Pelagic clays also satisfy the
176Hf/177Hf systematics in Pitcairn lavas since they do not contain detri-
tal zircon, a Hf-rich phase. The presence of detrital zircon in a sediment
would yield lower Lu/Hf ratios at a given Sm/Nd ratio, and the departure
of Pitcairn lavas from the mantle array suggests the presence of a zircon-
poor pelagic sediment. Therefore, among possible subducted protoliths
in the EM-1 mantle, pelagic sediments have the combination of high Th/
U and high Lu/Hf required to generate the Nd, Hf and Pb isotopic com-
positions in Pitcairn EM-1 lavas. Therefore, based on the geochemical
characteristics inferred for the Pitcairn EM-1 mantle source, incorpora-
tion of a pelagic sedimentary protolith into the mantle sourced by
Pitcairn EM-1 lavas remains a plausible model for the origin of this man-
tle end-member (Eisele et al., 2002; Honda and Woodhead, 2005;
Woodhead and Devey, 1993; Woodhead and McCulloch, 1989).

4.2. The presence of subducted oceanic crust in the EM-1 Pitcairn mantle

Eisele et al. (2002) modeled the mantle source of the EM-1 Pitcairn
mantle and found that, in addition to supporting a role for recycled pel-
agic sediment in the EM-1 mantle, the model results permit the pres-
ence of a mafic component in the EM-1 Pitcairn mantle. High
concentrations of TiO2 in OIB lavas, including lavas from the Pitcairn
suite, have also been used to argue for a recycled mafic component in
their mantle sources (Prytulak and Elliot, 2007). This is because melting
of a primitive or depleted mantle peridotite cannot generate the elevat-
ed TiO2 observed in primitive OIB lavas from many hotspots, even at
very low degrees of melting. We emphasize that elevated Ti concen-
trations do not translate to particularly striking Ti, Ta and Nb (TITAN)
anomalies (Jackson et al., 2008) in the new dataset; this is because the
concentrations of these elements are generally complemented by
higher concentrations of elements of similar incompatibility. While
the presence of a mafic component in the Pitcairn mantle is indicated
from elevated TiO2 in Pitcairn lavas, it is important to evaluate the na-
ture of this mafic component.

With the exception of the tholeiitic seamount samples from the
Pitcairn hotspot (including sample 65DS-4), most Pitcairn lavas are
alkaline and silica undersaturated, which suggests that the mafic compo-
nent in the mantle source is silica-undersaturated (i.e., a silica-deficient
pyroxenite; Herzberg, 2011). This silica-undersaturated mafic compo-
nent may be responsible for elevating the TiO2 in alkalic Pitcairn lavas.
The less common tholeiitic samples from the Pitcairn hotspot offer
important clues to silica-saturated recycled mafic compositions in the
Pitcairn mantle. Together with Koolau lavas from Hawaii (e.g., Hauri,
1996), lavas from the Pitcairn hotspot, in particular the submarine
Pitcairn seamount lavas (e.g., Eisele et al., 2002; Hekinian et al., 2003;
Woodhead and Devey, 1993), anchor the SiO2-rich portion of the global
OIB array (Fig. 7, Jackson and Dasgupta, 2008). The similarity in SiO2
contents in both the Pitcairn seamount lavas (including sample 65-
DS4) and Koolau lavas may suggest that mantle sources at both EM-1 lo-
calities may generate SiO2-rich lavas. The observation of tholeiitic sam-
ples at the Pitcairn hotspot is important and can give important clues to
the nature of the EM-1 mantle, as oceanic hotspot lavas erupted far from
mid-ocean ridges (i.e., excluding near-ridge hotspots like Iceland and Galapagos) are predominantly alkaline. Among hotspots that erupted far from mid-ocean ridges, excluding Hawaii, tholeiitic lavas are uncommon (<10%). Of 14 such hotspots tabulated by (Jackson and Dasgupta, 2008), all plot, on average, in the alkali basalt field of Macdonald and Katsura (1964).

The new tholeiitic Pitcairn seamount sample, 65DS-4, not only has the highest olivine-corrected SiO₂ abundance in the Pitcairn dataset, but it also extends the Pitcairn field closer to the Koolau EM-1 field in Pb-isotopic space than any previously examined Pitcairn lava (Fig. 4). The geochemical affinities suggest that there may be broad similarities, including geochemically-enriched Sr and Nd isotopic compositions and elevated SiO₂, in the mantle sources sampled by the low 208Pb/204Pb component in Pitcairn (which is exhibited in purest form by 65DS-4; Fig. 4) and the EM-1 component in Koolau lavas. The origin of the SiO₂-rich nature of Koolau lavas from Hawaii is argued to be the result of eclogite melting in the Hawaiian mantle plume (Hauri, 1996; Herzberg, 2011; Sobolev et al., 2005), where the eclogite is suggested to be a recycled oceanic crust that preserved low U/Pb ratios through the subduction zone and generated relatively low time-integrated 206Pb/204Pb (Jackson and Dasgupta, 2008; Jackson et al., 2012). Hauri (1996) showed that existing peridotite melting experiments at pressures >2 GPa (i.e., pressures that are slightly lower than the pressure at the base of mature oceanic lithosphere beneath Pitcairn and Hawaii) cannot generate the major element compositions observed in Koolau lavas, and more recent compilations of experimental data support this observation (see discussion in Jackson et al., 2012). Hauri (1996) Hauri (1996) also showed that elevated FeO at a given SiO₂ in Koolau lavas is a result of eclogite melting. Given existing experimental constraints, the high FeO and SiO₂-rich tholeiites from Hawaii are best explained by melting a silica-saturated, mafic protolith in the mantle source (Hauri, 1996; Jackson et al., 2012). Sample 65DS-4 and a subset of Pitcairn seamount samples plot within or close to the Hawaiian Koolau field in a plot of SiO₂ vs. FeO (Fig. 8); sample 65DS-4 plots in the field for Mauna Loa lavas, which also host elevated FeO at a given SiO₂ abundance, and like Koolau lavas, are argued to be melts of an eclogite-bearing mantle source (Hauri, 1996), (Lasitter and Hauri, 1998), (Blanchert-Toft et al., 1999), (Huang and Frey, 2005), (Sobolev et al., 2005), (Herzberg, 2006), (Jackson et al., 2012). Therefore, we suggest that a subset of EM-1 Pitcairn lavas, which have SiO₂ abundances that anchor the high-SiO₂ portion of the data arrays in Figs. 7 and 8, sample a mantle source that is lithologically similar to the mantle source of high-SiO₂ Hawaiian lavas (Mauna Loa and Koolau).

4.3. Helium isotopic constraints on the mantle source beneath Pitcairn

4.3.1. Origin of the depleted high ³He/⁴He component hosted in Pitcairn lavas: entrained depleted upper mantle or a depleted plume (FOZO?) component?

The new data provide a window into the high ³He/⁴He component in the Pitcairn suite. The Pitcairn lava (Pit-16) with the highest ³He/⁴He (12.6 Ra, ratio to atmosphere) is also the sample with the highest 206Pb/204Pb that has been examined for helium isotope (Fig. 5). Additionally, with increasing 206Pb/204Pb, Pitcairn lavas exhibit increasingly depleted geochemical signatures, including higher 143Nd/144Nd and lower 87Sr/86Sr (Fig. 4). While the high ³He/⁴He Pitcairn sample, Pit-16, is not in itself geochemically-depleted (142Nd/144Nd is 0.512590, 87Sr/86Sr is 0.704687), it plots in a region of isotopic space that is closer to the most depleted Pitcairn component (but farther from the EM-1 domain) than other lavas from Pitcairn that have been characterized for helium isotopes (Fig. 4). Therefore, going in the direction of the depleted Pitcairn component in isotopic space, ³He/⁴He appears to increase. Since MORB lavas tend to have ³He/⁴He ratios that cluster near 8 ± 1 Ra (e.g., Graham et al., 1992), the new ³He/⁴He measurement suggests that the depleted component in the Pitcairn mantle is unlike the typical upper mantle material sampled at mid-ocean ridges. Instead, this mixing component may be the high ³He/⁴He component that is proposed to reside in the lower mantle (Hart et al., 1992).

Based on radiogenic Sr-Nd-Pb isotopic compositions, four distinct mantle components EM-1, EM-2, HIMU and the depleted MORB mantle are suggested to encompass the global OIB dataset, and these end-members have been plotted as the vertices of a three-dimensional tetrahedron in Sr-Nd-Pb isotopic space (e.g., Hart et al., 1992). Each hotspot forms arrays within the tetrahedron that appear to converge on a common component with geochemically-depleted Sr and Nd isotopic compositions, and Hart et al. (1992) argued that this component, called FOZO (Focus Zone), is distinct from (i.e., less geochemically-depleted than) the MORB mantle and has high ³He/⁴He. Indeed, the Pitcairn lava with the highest ³He/⁴He is far from being as depleted as the MORB mantle.

Like other hotspot suites, geochemically-depleted lavas from the Pitcairn hotspot appear to converge on the common component of Hart et al. (1992), but the range of ³He/⁴He data in the subset of depleted Pitcairn lavas examined by Honda and Woodhead (2005) were inconsistent with a FOZO component in the Pitcairn hotspot. Instead, Honda and Woodhead (2005) proposed that the MORB-mantle is the depleted component sampled by geochemically-depleted Pitcairn lavas. The new ³He/⁴He isotopic data presented here suggest that the depleted component in the Pitcairn mantle is not like the mantle sampled at mid-ocean ridges. Instead, the terrestrial mantle reservoir hosting high ³He/⁴He, like that sampled by sample Pit-16, is proposed to reside in the lower mantle (Hart et al., 1992). If this is the case, then the depleted Pitcairn component was incorporated into the Pitcairn plume in the lower mantle, and is not a component entrained in the upper mantle. While the ³He/⁴He ratios in the geochemically-depleted Pitcairn lavas are not extraordinarily elevated, they do provide hints of a non-MORB component in the geochemically depleted domains of the Pitcairn plume. Honda and Woodhead (2005) showed from neon isotope data that Pitcairn samples of clear EM-1 signature had a major component of solar neon, but the data did not fall near the N-MORB correlation line.

4.3.2. MORB-like ³He/⁴He in the EM-1 mantle

A notable feature of the new dataset is the observation that the Pitcairn lavas with the most geochemically-enriched isotopic signatures have ³He/⁴He similar to MORB. The two Pitcairn lavas that have the most extreme EM-1 signatures (i.e., lowest 143Nd/144Nd and
suggested to host recycled crustal components, including recycled 
206Pb/204Pb and highest 87Sr/86Sr), and that have also been character-
ized for helium isotopic ratios, are found to have MORB-like 3He/4He 
(Fig. 5). However, the other mantle end-members associated with 
recycled crustal materials, EM-2 and HIMU, have 3He/4He ratios lower 
than MORB, as do lavas from several other hotspot localities 
(e.g., Tristan, Gough, Comores, Canaries) that do not sample end-
member heavy radiogenic isotope compositions (Barford et al., 1999; 
Class et al., 2005; Day and Hilton, 2011; Graham et al., 1992; Hanyu 
and Kaneoka, 1997; Jackson et al., 2007; Kurz et al., 1982; Moreira 
et al., 2011; Parai et al., 2009). 3He/4He ratios range down to 4.3 Ra 
in Samoan EM-2 lavas with 87Sr/86Sr of 0.7186 (Jackson et al., 2007), 
(Jackson et al., 2009), and the low 3He/4He is interpreted to be the result 
of the incorporation of terrigenous sediment with low 3He/4He (down 
to 0.01 Ra; (Podosek et al., 1980), (Ballentine and Burnard, 2002)) 
into peridotitic mantle (~8 Ra); such sediment is U and Th-rich and 
will generate 4He, and the mixture of sediment and mantle peridotite 
will evolve relatively low 3He/4He in the EM-2 reservoir over time 
(Jackson et al., 2009). Similarly, lavas sampling the HIMU mantle 
generally have 3He/4He < 7 Ra (e.g., St. Helena, Mangaia, Tubuai, older 
series at Rurutu; (Graham et al., 1992), (Hanyu and Kaneoka, 1997), 
(Parai et al., 2009), (Hanyu et al., 2011)) or ~8 in OIB with less extreme 
HIMU signatures (e.g., Canaries; (Day and Hilton, 2011)). The sub-
MORB 3He/4He ratios may relate to the presence of recycled U and Th-
rich oceanic crust into the HIMU mantle (e.g., (Hofmann and White, 1982)). 
Note that this U–Th enrichment is opposite the low U/Pb assumed above for the eclogitic component of Koolau EM-1 lavas. If the high 
HIMU mantle is host to recycled oceanic crust, which is degassed and 
will have highly radiogenic 4He (3He/4He ~0.1 Ra; (Moreira 
and Kurz, 2001)); an important question is how lavas sampling this 
mantle reservoir have 3He/4He that is significantly higher than a 
recycled oceanic crust. A number of solutions have been presented 
that attempt to explain the observation of sub-MORB 3He/4He ratios in 
HIMU lavas that are not as low as degassed oceanic crust (e.g., Class 
et al., 2005; Day and Hilton, 2011; Hanyu and Kaneoka, 1997; Hanyu 
et al., 1999, 2011; Hart et al., 2008; Hilton et al., 2000; Moreira 
and Kurz, 2001; Moreira et al., 2011; Parai et al., 2009).

Like the HIMU and EM-2 reservoirs, Pitcairn EM-1 lavas are also 
suggested to host recycled crustal components, including recycled 
sediment and oceanic crust. However, Pitcairn EM-1 lavas have 
MORB-like 3He/4He, which is higher than end-member HIMU and EM-
2 lavas and is therefore notable. Recycled crustal materials, like those 
suggested to exist in the EM-1 mantle beneath Pitcairn, should generate 
4He by U and Th decay over time, and this should generate mantle 
reservoirs with low 3He/4He (<0.1 Ra). A clue to the apparent paradox 
of MORB-like 3He/4He in the Pitcairn may be provided by the higher 
diffusivity of He relative to the other radiogenic isotopic tracers (Sr, 
Nd and Pb). Owing to the high diffusivity of helium, Hart et al. (2008) 
suggested that mantle heterogeneities with length-scales smaller than 
5 to 10 km will not preserve their helium isotopic signatures over 
time scales of 1.5 Ga, but their Sr, Nd and Pb isotopic signature will 
remain relatively unchanged. If the recycled sediment heterogeneities 
in the EM-1 mantle are distributed over relatively short length scales 
(5 to 10 km or less), and if the EM-1 mantle is sufficiently old 
(~1.5 Ga), then the helium isotopic composition of the EM-1 mantle 
may become overwhelmed by the 3He/4He signature of the mantle 
reservoir hosting the recycled materials, here inferred to be ambient 
upper mantle, which is 8 Ra. Of course, this requires that the 4He 
production of the recycled component is low compared to the ambient 
mantle helium concentration (Hart et al., 2008). If the time-integrated 
helium concentrations of the upper mantle greatly exceed the concen-
trations of the recycled oceanic crust and sediment (Day and Hilton, 
2011), and if the length scales of the recycled materials are small 
(~5–10 km; (Hart et al., 2008), the 3He/4He of the resulting mantle 
reservoir is more likely to have a 3He/4He similar to the ambient 
upper mantle. A diffusive decoupling mechanism like that proposed 
by Hart et al. (2008) may explain the presence of MORB-like 3He/4He 
in a mantle reservoir that hosts recycled oceanic crust and sediments. 
Because neon diffuses many orders of magnitude slower than helium 
in olivine, Cherniak et al. (2014), the solar neon signature of Honda 
and Woodhead (2005) could be retained in the recycled material 
while a MORB-like helium is imprinted by diffusive exchange. Of course, 
a more sinister explanation would simply postulate a helium (but not 
Sr, Nd or Pb) isotope exchange in the uppermost mantle between 
migrating EM-1 melts (or magma chambers) and the ambient MORB 
mantle country rocks. It is of course possible that EM-1 has the same 
helium value as MORB by coincidence.

5. Conclusions

The extreme enriched mantle EM-1 signature in Pitcairn lavas likely 
hosts an enriched protolith derived from upper continental crust, and 
this protolith exhibits many geochemical similarities with pelagic 
sediment. Elevated primary-melt Ti in the EM-1 lavas suggests the 
presence of recycled pyroxenite in the mantle source. We identify a 
thalassic sample (65–D54) from Adams Seamount at the leading edge 
of the Pitcairn hotspot, and the thalassic composition of this sample comple-
ments tholeiites previously described from the Pitcairn hotspot. The new 
thalassic Pitcairn seamount sample not only has the highest olivine-
corrected SiO2 abundance in the Pitcairn dataset, but it also extends the 
Pitcairn field closer to the Hawaiian Koolau field in Pb-isotopic space 
that any previously examined Pitcairn lava. The geochemical affinities 
suggest that there may be broad similarities including geochemically-
enriched Sr and Nd isotopic compositions and elevated SiO2 in the mantle 
sources sampled by thalassic Pitcairn lavas and the EM-1 component in 
Koolau lavas. The origin of the SiO2-rich nature of Koolau lavas from 
Hawaii is argued to be the result of eclogite melting in the Hawaiian 
mantle plume. The isotopic and major element affinities between the 
Pitcairn EM-1 thalassic sample 65S5–4 and the EM-1 component sampled 
by thalassic Koolau lavas from Hawaii suggests a common origin, and we 
argue that a silica-saturated mafic protolith is in the source of the EM-1 
mantle at both hotspots. We also present the highest 3He/4He (12.6 Ra) 
reported for Pitcairn lavas, and it is associated with a third component 
with the highest 206Pb/204Pb and the most geochemically-depleted 
87Sr/86Sr and 143Nd/144Nd ratios in the Pitcairn suite. We also show that 
EM-1 end-member lavas from Pitcairn have 3He/4He that is indistinguish-
able from the MORB mantle. If the recycled sedimentary and mafic 
components in the Pitcairn EM-1 occur at relatively short length scales, 
distributed in a peridotite matrix with 4He/He of 8 Ra, diffusive exchange 
of helium over timescales >1.5 Ga will permit the 3He/4He of the Pitcairn 
EM-1 mantle source to evolve toward values that typify the ambient 
upper mantle sampled by MORB. Alternatively, exchange of helium 
between EM-1 melts and melt conduits in the upper mantle host rock 
may result in EM-1 helium isotopic ratios evolving toward values 
typical of the upper mantle.

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