Recycled crust in the Galápagos Plume source at 70 Ma:

Implications for plume evolution

Jarek Trela¹, Chris Vidito², Esteban Gazel^{1*}, Claude Herzberg², Cornelia Class³,

William Whalen¹, Brian Jicha⁴, Michael Bizimis⁵, Guillermo E. Alvarado⁶

1. Department of Geosciences, Virginia Tech, Blacksburg, VA 24061, USA

2. Earth and Planetary Sciences, Rutgers University, Piscataway, NJ 08854,

USA

3. Lamont-Doherty Earth Observatory of Columbia University, Palisades, NY

10964, USA

4. Department of Geoscience, University of Wisconsin-Madison, Madison, WI

53706, USA

5. Department of Earth and Ocean Sciences, University of South Carolina,

Columbia, SC 29208, USA

6. Instituto Costarricense de Electricidad, Area de Amenazas y Auscultacion

Sismologica y Volcanica, Apdo 10032-1000, Costa Rica

**Corresponding author: egazel@vt.edu*

Revised manuscript for EPSL

(Word count, Abstract 218, Text 5364)

ABSTRACT

Galápagos plume-related lavas in the accreted terranes of the Caribbean and along the west coast of Costa Rica and Panama provide evidence on the evolution of the Galápagos mantle plume, specifically its mantle temperature, size and composition of heterogeneities, and dynamics. Here we provide new $^{40}\text{Ar}/^{39}\text{Ar}$ ages, major and trace element data, Sr-Nd-Pb isotopic compositions, and high-precision olivine analyses for samples from the Quepos terrane (Costa Rica) to closely examine the transitional phase of the Galápagos Plume from Large Igneous Province (LIP) to ocean island basalt (OIB) forming stages. The new ages indicate that the record of Quepos volcanism began at 70 Ma and persisted for 10 Ma. Petrological evidence suggests that the maximum mantle potential temperature (T_p) of the plume changed from $\sim 1650^\circ$ to $\sim 1550^\circ\text{C}$ between 90-70 Ma. This change correlates with a dominant pyroxenite component in the Galapagos source as indicated by high Ni and Fe/Mn and low Ca olivines relative to those that crystallized in normal peridotite derived melts. The decrease in T_p also correlates with an increase in high-field strength element enrichments, e.g., Nb/Nb*, of the erupted lavas. Radiogenic isotope ratios (Nd-Pb) suggest that the Quepos terrane samples have intermediate (Central Domain) radiogenic signatures. The Galápagos plume at 70 Ma represents elevated pyroxenite melt productivity relative to peridotite in a cooling lithologically heterogeneous mantle.

KEYWORDS

Galápagos plume, mantle evolution, olivine chemistry, source composition, LIP-OIB

1. INTRODUCTION

The initial melting of mantle plumes produces unparalleled volumes (5-40 x 10⁶ km³) of lava that form continental flood basalts and oceanic plateaus, which represent the most significant igneous structures on Earth (e.g., Coffin and Eldholm, 1994; Mahoney and Coffin, 1997; Saunders, 2005; Coffin et al., 2006). This type of intra-plate magmatism originates at a deep thermal boundary layer below the upper mantle, (possibly the core-mantle boundary, e.g., Morgan, 1972; Tolstikhin and Hofmann, 2005; Torvisk et al., 2010; Burke et al., 2011). During ascent, a plume may develop a volumetrically large “head”, followed by a narrower conduit or “tail”. Geodynamic models suggest that melting of a 500-2000 km wide plume head (Arndt, 2000) forms large igneous provinces (LIPs), characterized by high degrees of partial melting, high mantle potential temperatures (T_p), and widespread lava flows (> 10⁵ km²) (Richards et al., 1989; Coffin and Eldholm, 1994, Herzberg and Gazel, 2009). These events were sometimes so large that they affected environmental change leading to catastrophic mass extinctions or oceanic anoxic events (e.g., Courtillot and Renne, 2004; Kerr, 2005; Blackburn et al., 2013). Subsequent melting of a plume conduit will form ocean island basalts (OIBs), which may manifest themselves as tracks of linear ocean islands and seamount chains. Most older LIPS were hotter and melted more extensively than their younger OIB counterparts (Herzberg and Gazel, 2009).

The Galápagos hotspot provides an exceptional opportunity to reconstruct plume evolution due to the on-land accessibility of LIP terranes in the Caribbean and Costa Rica (Alvarado et al., 1997; Denyer and Gazel, 2009; Loewen et al., 2013)

and accreted OIB terranes along the west coast of Panama and Costa Rica (Hoernle et al., 2002). Recent petrological studies suggested that the Caribbean LIP (CLIP) melted more extensively and at higher temperatures than lavas from the modern Galápagos Islands (Herzberg and Gazel, 2009). The maximum T_p of the plume was ~ 1620 °C during the Cretaceous and cooled to ~ 1500 °C at present time at a rate of ~ 1 °C/Ma assuming a linear decrease (Herzberg and Gazel, 2009). Therefore, understanding the evolution of the Galápagos mantle plume has significant global implications for understanding mantle dynamics.

The amount, age, and contribution of subducted slab sources (eclogite, pyroxenite, or refertilized peridotite) fingerprinted in mantle plume geochemistry are also important. Although a recycled crust signature has been identified in plumes through the use of radiogenic isotopes and trace-element signatures (e.g., Zindler and Hart, 1986; Hofmann, 1997; Chauvel et al., 2008) and more recently through the use of major element signatures (e.g., Jackson and Dasgupta, 2008; Shorttle and MacLennan, 2011; Jackson et al., 2012) and numerical models (Ballmer et al., 2013) the exact nature of geochemical exchanges and mixing of pyroxenite with mantle end members requires further investigation. Global scale recycling of crustal material undoubtedly plays a key role in the development and evolution of deep geochemical reservoirs as well as affecting the thermodynamic cooling of Earth. Furthermore, when compared with a primitive mantle reference, typical OIB trace element signatures show positive high field strength element (e.g., Nb and Ta) anomalies, suggesting a subducted slab reservoir in the mantle (McDonough, 1991; Rudnick et al., 2000; Kamber and Collerson, 2000; Jackson et al., 2008). This

reservoir could represent the mantle domain component common in all plumes (e.g., C, FOZO) (Zindler and Hart, 1986; Hart et al., 1992; Stracke et al., 2005).

Our objective is to characterize the sources of the lavas present in the Quepos terrane and their effect on plume evolution during the important LIP-OIB transition. This “transition” marks a major decrease in the magmatic productivity when head stage melting ceases and tail stage melting begins during a plume’s evolution. In this study we use integrated petrological, geochemical, isotopic, and high-precision olivine geochemical analyses to compare our new results to preexisting data sets to show a decrease in T_p evident in the Quepos lavas that corresponds to a dominant pyroxenite component in the Galápagos Plume.

2. BACKGROUND

The formation of the Caribbean Large Igneous Province (CLIP) began ~90 Ma with the incipient melting of the Galápagos plume head (Duncan and Hargraves, 1984; Richards et al., 1989; Pindel and Barrett, 1990; Kerr, et al., 1996a; Kerr et al., 1996b; Hauff et al., 1997; Sinton et al., 1997; Hauff et al., 2000a;). The Caribbean plate represents an over-thickened section of oceanic crust (up to 20 km) and is interpreted as the product Galápagos plume head melting (Duncan and Hargraves, 1984; Hauff et al., 1997, 2000a,b; Sinton et al., 1998; Kerr et al., 1996a; Hoernle et al., 2002, 2004). The CLIP then migrated northeast with the Farallon Plate where it collided with the Greater Antilles Arc (e.g., Duncan and Hargraves, 1984; Hauff et al., 2000; Geldmacher et al., 2003, Gazel et al., 2011). This collision triggered a reversal in subduction polarity, which initiated subduction along the western edge of the

CLIP margin (Duncan and Hargraves, 1984; Geldmacher et al., 2003). The reversal in polarity facilitated the migration of the CLIP farther northeast between the converging North and South Americas between Late Cretaceous to Tertiary time (e.g., Duncan and Hargraves, 1984; Hauff et al., 2000a; Hoernle et al., 2002; Geldmacher et al., 2003).

Petrological and geochemical research of the Burica, Osa, and Quepos terranes in western Costa Rica (Fig. 1) together with Azuero and Sona terranes in Panama, provide strong evidence that these areas represent accreted Galápagos OIB terranes, which resulted from melting of the plume tail post CLIP formation (Hauff et al., 2000a; Hoernle et al., 2002; Denyer and Gazel, 2009). These terranes are markedly younger than CLIP related lavas (85-95 Ma) with $^{40}\text{Ar}/^{39}\text{Ar}$ ages between 65-40 Ma and represent ancestral Galápagos plume tail segments accreted to Central America (Sinton et al., 1997; Hauff et al., 2000a; Hoernle et al., 2002). Around 23 Ma the Farallon Plate began spreading to form the Nazca and Cocos plates (e.g., Lonsdale, 2005). During the next 20 Ma the Galápagos hotspot and the Cocos-Nazca ridge experienced a series of complex interactions (Werner et al., 2003). The ridge-hotspot interactions subsequently formed the Cocos and Carnegie aseismic ridges located on the northeastward and eastward migrating Cocos and Nazca Plates respectively.

The present day Galápagos archipelago is currently divided into four domains, characterized by unique isotopic variations in Nd-Sr-Pb and $^3\text{He}/^4\text{He}$ (Hoernle et al., 2000; Harpp and White, 2001). The Eastern Domain represents the most depleted isotopic end-member (Hoernle et al., 2000; Harpp and White, 2001)

with compositions similar mid ocean ridge basalt (MORB). Lavas from Floreana define the most isotopically enriched end member (Southern Domain) with high radiogenic $^{87}\text{Sr}/^{86}\text{Sr}$ and $^{206}\text{Pb}/^{204}\text{Pb}$ ratios (Harpp and White, 2001). Lavas in the Wolf-Darwin Lineament (Northern Domain) typically display minor ^{207}Pb and ^{208}Pb enrichments and $^3\text{He}/^4\text{He}$ ratios between 8.8-6.9 R/R_A (Harpp and White, 2001; Vidito et al., 2013) though they are depleted in $^{87}\text{Sr}/^{86}\text{Sr}$ and $^{206}\text{Pb}/^{204}\text{Pb}$ relative to Floreana lavas (Harpp and White, 2001). Islands in the Central Domain (e.g., Fernandina) most likely resemble the true “plume” composition and show intermediate enrichments in Sr, Nd, Pb (Harpp and White, 2001) with high $^3\text{He}/^4\text{He}$ (up to 30.3 R/R_A; Kurz et al., 2014).

The Quepos terrane (focus of this study) is one of six fault-bounded oceanic igneous terranes located on the Pacific Costa Rican coast (Fig. 1). An intra-plate Galápagos origin is now generally accepted to explain the formation of Quepos based on petrological evidence, field-based observations, and trace element data and represents the oldest accreted segments of the Galápagos OIB stage (Frisch et al., 1992; Hauff et al., 1997; Denyer and Gazel, 2009). The lava stratigraphy of Quepos is dominated by a transitional tholeiitic to alkaline series with incompatible trace element patterns indicative of an OIB source with a clear Galápagos isotopic signature (Hauff et al., 2000; Geldmacher et al., 2003).

3. MATERIALS AND METHODS

Picrite, diabase, and basalt samples were collected in the Quepos terrane, from in-situ outcrops or fresh, meter-size boulders along the shoreline to take

162 advantage of active surf erosion exposures. Sample locations can be found in Table
163 S1A in the Supplementary Materials.

164 At the University of Wisconsin-Madison Rare Gas Geochronology Laboratory,
165 $^{40}\text{Ar}/^{39}\text{Ar}$ incremental heating experiments were conducted on groundmass and
166 mineral separates using a 25 Watt CO_2 laser and analyzed using a MAP 215-50
167 following the procedures in Jicha and Brown (2014). Isotope data was reduced
168 using ArArCalc software version 2.5 (<http://earthref.org/ArArCALC/>). Ages are
169 reported with 2σ uncertainties (includes the J uncertainty) and are calculated
170 relative to a Fish Canyon standard age of 28.201 ± 0.046 Ma (Kuiper et al., 2008)
171 and a value for $\lambda^{40}\text{K}$ of $5.463 \pm 0.107 \times 10^{-10} \text{ yr}^{-1}$ (Min et al., 2000) (Supplementary
172 Tables S2A-E).

173 Alteration-free rock chips were selected under stereoscopic microscope and
174 powdered in an alumina mill at the Department of Geosciences at Virginia Tech.
175 From these powders, whole rock major and trace element compositions were
176 collected at Washington State University by XRF and ICP-MS (protocols in Johnson
177 et al., 1999) and are reported in Table S1A in the Supplementary Materials. Curacao
178 picrite samples were processed and analyzed in the geochemistry laboratory at the
179 Department of Geosciences at Virginia Tech following the procedures described in
180 Mazza et al. (2014). Major elements were collected by X-ray fluorescence (XRF), the
181 Relative Standard Deviation (RSD) for 10 replicates of BHVO-2g was $<2.70\%$ for all
182 major elements and accuracy better than 1.74% , with the exception of P_2O_5 . Trace
183 elements as well as P_2O_5 were collected from the same fluxed glasses with an Agilent
184 7500ce ICPMS coupled with a Geolas laser ablation system, following the

procedures detailed in Mazza et al. (2014). Data were calibrated against USGS standards BCR-2, BHVO-2, G-2, and BIR-1a, using Ti from XRF as an internal standard and the standard element values reported in Kelley et al. (2003). The accuracy for 4 replicates of BHVO-2g was better than 5 % for all elements except for Ba, Hf, and Ge (7-9 %). The accuracy for 4 replicates of BIR-1a was better than 5 % for most elements except for Ga, Ge, Ce, Pr, Eu, Tb, Dy, Ho, Tm, Er, Yb, and Lu (5-9 %).

Acid-digested aliquots of the same powdered material was used to collect radiogenic isotope ratios of Pb, Nd, and Sr on a Neptune multi collector ICPMS at the Center for Elemental Mass Spectrometry (CEMS), University of South Carolina (USC). Pb isotope ratios were determined by the Tl-addition technique (White et al., 2000) using $^{203}\text{Tl}/^{205}\text{Tl} = 0.418922$ and assuming identical fractionation factors between Tl and Pb. The Pb/Tl ratio ($^{208}\text{Pb}/^{205}\text{Tl} \sim 7$) of the samples was kept near identical to the NBS 981 standard by first performing dip checks in the samples and spiking Tl to the appropriate level. The NBS-981 standard was determined at $^{206}\text{Pb}/^{204}\text{Pb} = 16.936 \pm 0.001$, $^{207}\text{Pb}/^{204}\text{Pb} = 15.490 \pm 0.001$, $^{208}\text{Pb}/^{204}\text{Pb} = 36.694 \pm 0.003$ (2 standard deviations, $n = 13$). Isotopic ratios for Nd were normalized to $^{146}\text{Nd}/^{144}\text{Nd} = 0.7219$ and the Nd standard JNd_i was measured at $^{143}\text{Nd}/^{144}\text{Nd} = 0.5121$ and 0.512112 ± 0.000007 ($n=6$) at USC. Isotopic ratios for Sr were normalized to $^{86}\text{Sr}/^{88}\text{Sr} = 0.1194$ and replicate analyses of $^{87}\text{Sr}/^{86}\text{Sr}$ at 0.710320 ± 0.000012 . All Sr measurements are reported relative to NBS-987 where $^{87}\text{Sr}/^{86}\text{Sr} = 0.710250$.

Radiogenic isotope ratios of seven new Quepos samples (this study) were age corrected to initial eruptive values at 65 Ma using parent/daughter values reported in Table S1A. Additionally, we calculated the radiogenic composition of the Quepos and CLIP sources at 65 Ma and 90 Ma respectively by inverting primary magma compositions of Quepos and CLIP lavas assuming a modal composition of a refertilized peridotite and a melt fraction of 10%. We then projected these values to 0 Ma (assuming a radiogenic ingrowth corresponding to the source parent/daughter ratios). This method allows for proper comparison of Quepos and CLIP lavas to the present day isotopic domains in the Galápagos archipelago. We tested the sensitivity of radiogenic source compositions to the degree of partial melting and found negligible changes 10, 20, and 30%. Inverted source compositions for Quepos and CLIP samples, as well as additional details are included in Table S1D.

We performed high-precision trace element analyses on Quepos olivines using Rutgers University's JEOL JXA-8200 Superprobe. A modified version of the Sobolev et al. (2007) protocol was used to obtain high precision trace-element data. Samples were analyzed with a focused beam ($\sim 1 \text{ } \mu\text{m}$) at 20 kV and 300 nA. Peak count times were as follows: Si : 50s; Mg : 80s; Fe : 100s; Ni : 150s; Ca : 150s and Mn : 150s. The San Carlos olivine standard was analyzed at regular intervals during each run in order to correct for instrumental drift. A summary of the primary standards, statistics for the secondary standards, and statistics for the olivine analyses are given in Table S1B in Supplementary Materials. All oxide totals from the olivine analyses were normalized to 100%. Olivine data are also reported from

Curacao lavas (Kerr et al., 1996b for sample 92-8 and Revillon et al., 1999 for sample 94-54).

4. Petrological Modeling

Our goal is to use high precision olivine data to infer the lithology of the source that melted to produce the Quepos picrites; additionally, we examine olivines from the older CLIP lavas from Curacao in order to provide constraints on how the source lithology changed from ~90 to 70 Ma. A detailed description of the method that was adopted for petrological modeling was provided previously (Herzberg, 2011; Herzberg et al., 2013; 2014), and is given again in the Supplementary Materials. The model has the following components: 1) identification of a primary magma composition, 2) calculation of a liquid line of descent after variable proportions of olivine, clinopyroxene, and plagioclase have been subtracted, 3) calculation of the compositions of olivine that crystallize from these derivative magmas, 4) comparison of model and observed olivine compositions. The modeling requires use of the olivine/melt distribution coefficients D ; for Ni we have chosen the Beattie-Jones model (Jones, 1984; Beattie et al., 1991); for Ca and Mn we use the models in Herzberg and O'Hara (2004) and for Fe-Mg exchange between olivine and melt we use the model of Toplis (2005). There are other olivine/liquid distribution models for Ni, and all are parameterizations of experimental data. Of these, the Beattie-Jones model has the minimum root mean square error (RMSE) (reproducing experimentally collected data (Herzberg et al., 2013; 2014).

252 Matzen et al. (2013) proposed that the partitioning of Ni between olivine and
 253 melt decreases as temperature increases, and the inference is that deep melting of
 254 peridotite will produce Ni rich melts that crystallize Ni-rich olivine near the surface.
 255 They proposed that the NiO contents of olivines greater than 0.37% in mantle
 256 peridotites are proportional to the difference in temperature between melting in the
 257 mantle and crystallization near the surface, i.e., the ΔT effect. The difficulties with
 258 this model were previously examined (Herzberg et al., 2013; 2014) and we will
 259 draw attention to several points that compromise its use for interpreting the
 260 Curacao and Quepos olivine phenocrysts. The Beattie-Jones Ni partitioning model is
 261 more accurate than the Matzen et al. (2013) in describing the experimental data
 262 (Herzberg et al., 2013; 2014). The 1 RMSE for the Beattie-Jones model is 1.1; the 1
 263 RMSE for the Matzen model is 2.5 (Herzberg et al., 2014). Therefore, the difference
 264 in olivine NiO between average Hawaii and spinel peridotite (i.e., 0.46% and 0.37%)
 265 discussed by Matzen et al. (2013) is fully accounted for by the 1 RMSE of 2.5. Thus,
 266 the ΔT interpretation for the high NiO in Hawaiian olivines is undermined by the
 267 uncertainty. In the Beattie-Jones model the effect of T on D_{Ni} is accounted for in the
 268 MgO content of the melt, but in the Matzen et al. (2013) model, the separation of T
 269 and melt MgO as independent variables likely accounts for the larger RMSE. As
 270 discussed below, olivine compositions from Curacao crystallized from hot
 271 peridotite-source primary magmas, yet they display no elevations in Ni.
 272 Additionally, the highest olivine Ni contents worldwide are found in subduction
 273 zone lavas from the Mexican Volcanic Belt (Straub et al., 2008), yet there is no
 274 thermal anomaly below this region. Finally, we reiterate that the inference of a

pyroxenite or refertilized peridotite source from elevated Ni using the Beattie-Jones model is internally consistent with Ca and Fe/Mn, but no such consistency was provided in the Matzen et al. (2013) model.

5. RESULTS

The matrix of three samples from Quepos (two basalts and one picrite) were dated by $^{40}\text{Ar}/^{39}\text{Ar}$, and yielded weighted mean plateau ages of 61.4 ± 0.6 , 68.6 ± 0.3 , 70.6 ± 1.2 Ma. Supporting data summarizing the $^{40}\text{Ar}/^{39}\text{Ar}$ age dating results are located in the Supplementary Materials (Tables 2A-E). Previously published $^{40}\text{Ar}/^{39}\text{Ar}$ ages of Quepos basalts yielded ages at 60-65 Ma (Sinton et al., 1997; Hoernle et al., 2002). Our new measurements agree with the ~60 Ma pulse and extend the maximum age of Quepos volcanism to 70 Ma, indicating that the formation of this complex may have occurred over a 10 Ma interval.

New major and trace element results are plotted in Figures 2 and 3. Quepos lavas belong to both alkaline and sub-alkaline series (Fig. 2A). These lavas are mostly classified as basalts, with some examples of basaltic andesites, trachybasalts, and one basanite (Fig. 2A). Quepos picrites fall within the picro-basalt/basalt compositional fields (Fig. 2A) but textural and petrological evidence (Supplementary Materials, Section S1) suggest that they are not liquids but cumulates produced by olivine accumulation. Nevertheless, their olivine-controlled high MgO (>10 wt%) compositions and abundance of fresh olivines make them ideal for source composition determinations and petrological studies. Quepos picrites plot below the peridotite-pyroxenite discrimination line (Fig. 2B) defined by

Herzberg and Asimow (2008). In contrast, the older Curacao picrites (Kerr et al., 1996b, this study) plot on or above the line, in range with other CLIP samples consistent with a peridotite source. Primitive mantle normalized spider-diagrams from Quepos samples display broad enrichment in incompatible elements (Fig. 3). These lavas show marked positive high-field strength element enrichments (positive Ta, Nb, and Ti anomalies) as well as depletions in fluid mobile elements (K, Rb, Th, U) relative to elements with similar compatibilities. Additionally, the trace element abundances of our samples closely resemble the incompatible element patterns of current Galápagos hotspot tracks; the Cocos, Coiba, and Carnegie ridges (Fig. 3).

High precision data on olivine interpreted as derived from a mantle peridotite source from a wide range of tectonic environments contain a range of ~ 2800-3100 ppm Ni (Herzberg et al., 2013). These contents are consistent with values from olivine in primitive MORB from the East Pacific Rise and olivines inferred to have crystallized from primary magmas of Archean komatiites (Herzberg, 2011a; Herzberg et al., 2013). CLIP olivines from the ~ 90 Ma lavas of Curacao are another example of peridotite-source olivines (Fig. 4). Olivine compositions trend towards 3000 ppm Ni and 2000 ppm Ca, consistent with magmas derived from a peridotite source. Importantly, these olivines crystallized from hot primary magmas ($T_p = 1540^\circ\text{C}$; $\text{MgO} = 18\%$; Herzberg and Gazel, 2009; Hastie and Kerr, 2010), yet there is no evidence for elevated Ni in Curacao olivines that would support the ΔT model (Matzen et al., 2013). Curacao olivines have low Ni contents (Fig. 4), unlike high Ni Hawaiian olivines (Fig. 5).

When olivine Mg numbers are greater than 90, pyroxenite or refertilized peridotite formed by reaction of partial melts of recycled crust with peridotite are often revealed in olivine phenocrysts with high Ni, low Ca, low Mn and high Fe/Mn compared to olivines that crystallize from melts of normal MORB-like peridotite (Sobolev et al., 2005; 2007; Gurenko et al., 2009; 2010; Herzberg, 2011; Herzberg et al., 2014). Olivines from Quepos (Fig. 5) are too high in Ni and Fe/Mn and too low in Ca to have crystallized from normal mantle peridotite such as the source of MORB, Archean komatiites, and Curacao lavas (Fig. 4). They are also similar to olivines from Mauna Kea for which a pyroxenite source has been inferred (Sobolev et al., 2007; Herzberg, 2011a).

Seven new sets of Sr-Nd-Pb isotope ratios are provided in this study (Table S1A and Fig. 6). These ratios were first age corrected using an average age of 65 Ma and then used to project the radiogenic ingrowth of the source, as described in the methods above. These values are plotted in Figure 6 along with modern Galápagos Plume isotopic domains (Hoernle et al. 2000; Harpp and White, 2001) and previously published CLIP and Nicoya data, also projected to present day (Hauff et al., 2000a, 2000b; Hoernle et al., 2004). A detailed description and discussion of the isotope age corrections and source projections can be found in Supplementary Materials.

6. DISCUSSION

The new $^{40}\text{Ar}/^{39}\text{Ar}$ ages presented in this study, suggest that Quepos volcanism lasted for approximately 10 Ma beginning at ~70 Ma and terminating at

~60 Ma. Our new data suggest that the initial melting event, which formed this terrane occurred ~5 Ma earlier than previously described (Hoernle et al., 2002). The trace-element patterns reveal similarities with the current Galápagos hotspot tracks; the Cocos, Coiba, and Carnegie Ridges (Fig. 3). Therefore, the Quepos terrane is possibly the oldest accreted Galápagos OIB terrane found in Central America and records the transitional phase between LIP and OIB-type volcanism.

CaO-MgO bulk rock compositions can be used as a first order discrimination between peridotite and pyroxenite source-derived melts (Herzberg and Asimow, 2008). Samples from the CLIP plot in the area consistent with a dominantly peridotitic source composition (Fig. 2B), although it is also possible that some pyroxenite lithologies can also produce high CaO magmas (Herzberg, 2011). Our new samples from Quepos plot below that line and are similar to the range exhibited by Mauna Kea (Hawaii) lavas, which have been interpreted to be derived from a pyroxenite source (Sobolev et al., 2007; Herzberg, 2011a). We note that no experimental melt compositions of peridotites have been reported that plot below the discrimination line, supporting evidence that low CaO primary magmas originated by pyroxenite melting. Modern Galápagos archipelago lavas plot above and below the discrimination line, consistent with the presence of both peridotite and pyroxenite sources in the plume (Vidito et al. 2013). Also, one lava composition from Quepos reported by Tournon (1984) plots above the line, and this was used to model the olivines for peridotite source melts (Fig. 5).

New high-precision data from Quepos olivine phenocrysts show that they have higher Ni and Fe/Mn and lower Ca contents than those expected of a normal

peridotite source (Fig. 5). While Quepos and Mauna Kea olivines share many similarities (Fig. 5), there are also differences that may be interpreted as variable amounts of olivine in the source, which may be present in Quepos and variably present or even absent in Mauna Kea (Sobolev et al., 2005; Herzberg, 2011a).

The observed trends in Ni and Ca in Quepos olivines suggest that sole crystallization of olivine is not sufficient to explain the data. Quepos olivines are too low in Ca and too high in Ni to be derived from a pure pyroxenite end member (Fig. 5). The best fit occurs when olivine and clinopyroxene crystallize along a cotectic. However, this is problematic for several reasons. First, Quepos picrites lack clinopyroxene. Although magmas can crystallize olivine + clinopyroxene at high-pressures and then only olivine within shallower crustal magma chambers (e.g. Thompson et al., 1980; O'Donnell and Presnall, 1980; Elthon et al., 1995), this process would be preserved in the geochemical signature of these picrites. The lack of decreasing Sc/Al and Ca/Al with decreasing MgO does not support polybaric crystallization for Quepos samples. However, fractional crystallization coupled with magma recharge and mixing from a pyroxenite source provides a viable explanation to the Ni and Ca trends observed in Quepos olivines (Fig. 5). The recharge model shows that Quepos olivines crystallized from mixtures between the pyroxenite-derived primary magma and a derivative liquids with Mg# of 75 to 84 (Fig. 5). It is important to note that Ni and Ca are elements sensitive to magma recharge and mixing while Fe/Mn remains insensitive to this process. A detailed discussion of magma chamber recharge and mixing model is provided in the Supplementary Materials section.

Source-projected isotope variations in $^{143}\text{Nd}/^{144}\text{Nd}$ and $^{208}\text{Pb}/^{204}\text{Pb}$ and $^{206}\text{Pb}/^{204}\text{Pb}$ reveal that the Quepos terrane is most similar to the Central Domain of the Galápagos archipelago (Fig. 6). Nicoya and Curacao basalts also show similarity to the Central Domain, although are typically less radiogenic in Pb isotopic composition and more radiogenic in Nd isotopic composition than Quepos lavas. In the modern archipelago the Central Domain is by far the most widespread and defines lavas from Fernandina and Isabella (Harpp and White, 2001). This component dominates the center of the archipelago, whereas the other components prevail in the peripheral areas (Harpp and White, 2001). The trace element chemistry, radiogenic isotope ratios, and $^3\text{He}/^4\text{He}$ ratios (Graham et al., 1993; Kurz and Geist, 1999) of Fernandina all suggest that lavas erupted in the western reaches of the archipelago represent the most undegassed, primitive part of the Galápagos plume (Harpp and White, 2001).

Previous studies suggested some possible mechanisms leading to the formation of isotopically distinct domains in the Galápagos plume source include: 1) recycling and entrainment of ancient oceanic crust into the plume (Hart, 1984), 2) recycling of subducted sediment (Dupré and Allégre, 1983), 3) delamination of subcontinental lithosphere (Hart, 1984). Based on the new data presented here, we favor the interpretation that recycled oceanic crust is responsible for the more radiogenic Pb isotopic nature of the Quepos terrane, relative to CLIP lavas.

Our new $^{40}\text{Ar}/^{39}\text{Ar}$ ages and geochemical data provide valuable insight into a crucial phase during the life cycle of the Galápagos plume during the LIP-OIB stage transition, thereby elucidating the “head-tail plume evolution”. A fundamental

413 question that remains is: what effect does recycled oceanic crust in the form of
 414 pyroxenite contribution have on the evolutionary history of the Galápagos plume?
 415 Herzberg and Gazel (2009) proposed that CLIP lavas melted hotter and more
 416 extensively than lavas from the 0-15 Ma archipelago and Carnegie and Cocos tracks
 417 (Fig. 7A). A significant decrease in T_p ($\sim 75^\circ\text{C}$) is also observed between ~ 90 Ma
 418 CLIP lavas and ~ 65 Ma Quepos lavas (Herzberg and Gazel, 2009). Interestingly,
 419 there is also an increasing trend in high-field strength elements (HFSE), particularly
 420 Nb/Nb^* (defined as $\text{Nb}_N/\sqrt{(\text{Th}_N \times \text{La}_N)}$) from ~ 90 to 0 Ma (Fig. 7B). HFSE's such as Nb
 421 are sequestered in refractory mineral phases, e.g., rutile, in a subducting slab, thus
 422 the increase in Nb/Nb^* between 90 and 65 Ma may reflect a significant increase in
 423 recycled oceanic crust as the pyroxenite source for Quepos lavas. Jackson et al.,
 424 (2008) presented evidence for a global enriched HFSE reservoir commonly sampled
 425 by OIBs with correspondingly high $^3\text{He}/^4\text{He}$, and radiogenic $^{187}\text{Os}/^{188}\text{Os}$ that
 426 collectively suggest the existence of a mantle source with a refractory eclogite
 427 (pyroxenite) component (Jackson et al., 2008). This result advocates important
 428 implications for the development of global mantle geochemical heterogeneity. The
 429 high $^3\text{He}/^4\text{He}$ ratios of erupted lavas at some hotspots (Galápagos, Hawaii, Samoa,
 430 Iceland) are interpreted to originate from a relatively primitive, undegassed mantle
 431 reservoir (Kurz et al., 1982; Farley et al., 1992), named PHEM (Farley et al., 1992),
 432 FOZO (Hart et al., 1992), or C (Hanan and Graham, 1996). Understanding the nature
 433 of such a primitive, undegassed mantle reservoir and the development of a
 434 nonprimitive (nonchondritic?) mantle component is therefore crucial for
 435 understanding the formation of the high HFSE – high $^3\text{He}/^4\text{He}$ domain frequently

436 sampled by some plumes (Galápagos, Iceland, Hawaii, Samoa), balancing Earth's
437 HFSE budget from subduction to deep recycling, and evaluating large-scale
438 planetary differentiation processes (Jackson et al., 2008).

439 Evidence is accumulating that the amount of recycled crust in the Galápagos
440 mantle plume has been increasing with time. Peridotite-source melting is consistent
441 with the ~90 Ma CLIP lavas that have high CaO contents (Fig. 2B) and olivine data
442 that indicate a peridotite source provenance (Fig. 4). The first evidence for
443 participation of a dominant recycled crust component is at ~ 70 Ma as documented
444 in this paper by lavas from the Quepos terrane. And in modern lavas, olivine
445 chemistry reveals the widespread occurrence of pyroxenite throughout the present-
446 day Galápagos archipelago (Vidito et al., 2013). However, determining secular
447 changes in the absolute amount of recycled crust contained in the Galápagos plume
448 is difficult because of its lower solidus and higher melt productivity relative to
449 peridotite (e.g., Morgan, 2001; Pertermann and Hirschmann, 2003; Ito and
450 Mahoney, 2005; Sobolev et al., 2007; Shorttle and MacLennan, 2011). That is, given
451 the same lithological mixture of peridotite and pyroxenite, if the plume is cooler for
452 any reason, pyroxenite will preferentially melt owing to its lower melting
453 temperature, and its elevated productivity will mean that pyroxenite-source melts
454 will be overly abundant. However, these petrological constraints provide a
455 qualitative framework for explaining the larger geodynamical framework for
456 cooling the plume with time (Fig. 7A).

457 Petrological modeling suggests that most LIPS (Paleocene and older) were
458 hotter and melted more extensively than younger ocean islands, maybe only with

the exception of Hawaii (Herzberg and Gazel, 2009). It would be difficult to imagine a scenario of LIPs-producing plumes rooted in a hotter boundary layer at the base of the mantle than younger ocean island-producing plumes. If we assume that the Galápagos is representative of global plume evolution, and that recycled crust remains negatively buoyant relative to peridotite at all depths in the mantle (Hirose, 2006; Ricolleau et al., 2010), then it is possible that mantle plumes were triggered near the core-mantle boundary by instabilities of the buoyantly-favored perovskite-bearing peridotite equivalent, and that they progressively entrained more recycled crust with time. According to Davaille et al. (2002) entrainment of dense crust would require a hotter mantle to rise, which is not consistent with the Galápagos plume. An alternative explanation is that relative to the recycled crust-free peridotite equivalent, mantle plumes that entrain a limited amount of recycled crust of limited volume are expected to have a longer transit ascent time, creating opportunities for greater conductive heat loss to their surroundings (Farnetani and Samuel, 2005; Nolet et al., 2006; Kumagai et al., 2008). The problem with this model is that increased recycled crust entry into the root of the plume can be confused with increased melt productivity of recycled crust that is expected from cooling. Detailed work on the oldest 90 Ma CLIP lavas and additional Galápagos-related terranes might break this interpretative circularity if it can constrain whether the LIP source was free of recycled crust or if it was present, but completely exhausted and diluted during partial melting.

7. CONCLUSIONS

482 The new age constraints presented here indicate that the LIP-OIB transition
483 in the history of the Galápagos Plume occurred at about 70-60 Ma, i.e. 20-30 Ma
484 after the onset of CLIP volcanism. Our results show that a dominant pyroxenite
485 source lithology is observed for the first time at ~70 Ma in the record of the Quepos
486 terrane.

487 The maximum T_p of the plume changed from ~1650 to ~1550 °C between
488 90-70 Ma. This change corresponds to a dominant pyroxenite component in the
489 Galápagos plume evident in the Quepos data. Furthermore, there is also an increase
490 in HFSE contents relative to equally incompatible elements throughout the
491 evolution of the plume. The petrological requirement of elevated melt productivity
492 of pyroxenite relative to peridotite in a cooling lithologically heterogeneous mantle
493 is well-illustrated by the Galápagos plume. However, with no obvious reason for
494 plume material being cooler at the time of tail versus plume head formation, we
495 consider an increased incorporation of pyroxenite into the plume tail as the cause
496 for the secular cooling of the plume tail relative to the head most likely. This secular
497 cooling would be caused due to the decreased buoyancy of the plume material with
498 a higher pyroxenite proportion resulting in a slower ascent rate through the mantle
499 and thus, a higher cooling rate. The increased pyroxenite contribution through time
500 would in this case reflect the effect of an increased proportion in the plume tail
501 combined with an increased melt production rate.

502 Sr-Nd-Pb radiogenic isotopic compositions suggest that the source of the ~65
503 Ma Quepos terrane contains intermediate signatures similar to the classically
504 defined Central Domain and are more radiogenic than CLIP lavas, suggesting an

enrichment in the source of the plume, probably the result of a recycled oceanic crust component.

ACKNOWLEDGEMENTS

The National Science Foundation Award EAR- 12019033, Evolution of the Galápagos Mantle Plume to Gazel and Class supported this project. Reviews and comments by N. Sou and S. Mazza improved the original manuscript. Thanks go to Andrew Kerr for providing samples from Curacao and Anne Davaille for comments. This is Lamont-Doherty Earth Observatory (LDEO) Contribution # (number provided when the manuscript is accepted for publication). The authors are very grateful to Catherine Chauvel and an anonymous reviewer for thoughtful and critical comments that greatly improved this paper.

REFERENCES

- Allan, J.F., Simkin, T., 2000. Fernandina Volcano's evolved, well-mixed basalts: Mineralogical and petrological constraints on the nature of the Galápagos plume. *J. Geophys. Res.* 105, 6041-6071.
- Alvarado, G. E., Denyer, P., Sinton, C. W., 1997. The 89 Ma Tortugal komatiitic suite, Costa Rica: Implications for a common geological origin of the Caribbean and Eastern Pacific region from a mantle plume. *Geology*, 25 (5), 439-442.

- 528 Arndt, N., 2000. Hot heads and cold tails. *Nature*, 407, 458-461.
- 529 Ballmer, M.D., Ito, G., Wolfe, C.J., Solomon, S.C., 2013. Double layering of a
 530 thermochemical plume in the upper mantle beneath Hawaii. *Earth Planet. Sci.*
 531 *Lett.* 376, 155-164.
- 532 Blackburn, T.J., Olsen, P.E., Bowring, S.A., McLean, N.M., Kent, D.V., Puffer, J., McHone,
 533 G., Rasbury, E.T., Et-Touhami, M., 2013. Zircon U-Pb geochronology links the
 534 end-Triassic extinction with the Central Atlantic Magmatic Province. *Science*,
 535 340, 941-945.
- 536 Blichert-Toft, J., White, W. M., 2001. Hf isotope geochemistry of the Galápagos
 537 Islands. *Geochem. Geophys. Geosyst.* 2, <http://dx.doi.org/2000GC000138>.
- 538 Burke, K., 2011. Plate Tectonics, the Wilson Cycle, and Mantle Plumes: Geodynamics
 539 from the Top. *Ann. Rev. Earth Planet. Sci.* 39 (1), 1-29.
- 540 Beattie, P., Ford, C., Russell, D., 1991. Partition coefficients for olivine-melt and
 541 orthopyroxene-melt systems. *Contrib. Mineral. Petrol.* 109, 212-224.
- 542 Danyushevsky, L. V., and Plechov, P., 2011. Petrolog3: Integrated software for
 543 modeling crystallization processes. *Geochem. Geophys. Geosyst.* 12 (7).
 544 <http://dx.doi.org/10.1029/2011GC003516>.
- 545 Chauvel, C., Lewin, E., Carpentier, M., Arndt, N. T., Marini, J.-C., 2008. Role of
 546 recycled oceanic basalt and sediment in generating the Hf-Nd mantle array.
 547 *Nat. Geo.* 1 (1), 64-67.
- 548 Coffin, M. F., Duncan, R. A., Eldholm, O., Fitton, J. G., Frey, F. A., Larsen, H. C., Mahoney,
 549 J. J., Saunders, A. D., Schlich, R., Wallace, P. J., 2006. Large Igneous Provinces
 550 and Scientific Ocean Drilling status quo and a look ahead. *Oceanography*. 19,

551 150-160.

552 Coffin, M. F., Eldholm, O., 1994, Large igneous provinces: crustal structure,
553 dimensions, and external consequences. *Rev. Geophys*, 32, 1-36.

554 Courtillot, V.E., Renne, P.R., 2003. On the ages of flood basalt events. *Comptes*
555 *Rendus Geoscience*. 335, 113-140.

556 Denyer, P., Gazel, E., 2009. The Costa Rican Jurassic to Miocene oceanic
557 complexes: Origin, tectonics and relations. *J. South Am. Earth Sci.* 28 (4), 429-
558 442.

559 Duncan, R., Hargraves, R., 1984. Plate tectonic evolution of the Caribbean region
560 in the mantle reference frame. *Geol. Soc. Am. Mem.* 162, 81-94.

561 Dupré, B., and Allegre, C. J., 1983. Pb-Sr isotope variation in Indian Ocean basalts
562 and mixing phenomena. *Nature*. 303, 142-146.

563 Duran, P., 2013. Estudio vulcano-estructural y sedimentario del mélange en el
564 Promontorio de Quepos, Pacífico Central, Costa Rica. University of Costa Rica.
565 Central American School of Geology. Masters Thesis.

566 Elthon, D., Ross, D.K., Meen, J.K., 1995. Compositional Variations of Basaltic Glasses
567 from the Mid-Cayman Rise Spreading Center. *J. Geophys. Res.* 100 (B7),
568 12497-12512.

569 Farley, K. A., Craig, H., 1992. Mantle Plumes and Mantle Sources. *Science*, 258
570 (5083), 821-822.

571 Farnetani, C. G., Samuel, H., 2005. Beyond the thermal plume paradigm:
572 *Geophys. Res. Lett.* 32, 1-4.

573 Frisch, W., Meschede, M., Sick, M., 1992. Origin of the Central American

- ophiolites: Evidence from paleomagnetic results. *Geol. Soc. Am. Bull.* 104 (10), 1301-1314.
- Gale, A., Dalton, C.A., Langmuir, C.H., Su, Y., Schilling, J.-G., 2013. The mean composition of ocean ridge basalts. *Geochem. Geophys. Geosyst.* 14, 489-518. [http://dx. doi:10.1029/2012GC004334](http://dx.doi.org/10.1029/2012GC004334)
- Gazel, E., Abbott R. N, Draper, G. 2011., Garnet-bearing ultramafic rocks from the Dominican Republic: Fossil mantle plume fragments in an ultra high pressure oceanic complex? *Lithos.* 125 (1-2), 393-404.
- Geist, D., White, W.M., Albarede, F., Harpp, K., Reynolds, R., Blichert-Toft, J., Kurz, M.D., 2002. Volcanic evolution in the Galápagos: The dissected shield of Volcan Ecuador. *Geochem. Geophys. Geosyst.* 3 (1). [http://dx. doi:10.1029/2002GC000355](http://dx.doi.org/10.1029/2002GC000355) of 32-32 of 32.
- Geldmacher, J., Hanan, B. B., Blichert-Toft, J., Harpp, K., Hoernle, K., Hauff, F., Werner, R., Kerr, A. C., 2003. Hafnium isotopic variations in volcanic rocks from the Caribbean Large Igneous Province and Galápagos hot spot tracks. *Geochem.Geophys.Geosyst.* 4 (7). <http://dx.doi.org/10.1029/2002GC000477>
- Graham, D. W., Christie, D. M., Harpp, K. S., Lupton, J. E., 1993. Mantle Plume Helium in Submarine Basalts from the Galápagos Platform. *Science.* 262, 2023-2026.
- Hanan, B. B., Graham, D. W., 1996. Lead and helium isotope evidence from oceanic basalts for a common deep source of mantle plumes. *Science.* 272 (5264), 991-995.
- Harpp, K. S., White, W. M., 2001. Tracing a mantle plume: Isotopic and trace

597 element variations of Galápagos seamounts. *Geochem. Geophys. Geosyst.* 2,
 598 <http://dx.doi.org/2000GC000137>.

599 Hart, S., Hauri, E., Oschmann, L., Whitehead, J., 1992. Mantle plumes and
 600 entrainment: isotopic evidence. *Science*. 256, (5056) 517-520.

601 Hart, S. R., 1984. A large-scale isotope anomaly in the Southern Hemisphere mantle.
 602 *Nature*. 309, 753-757.

603 Hastie, A.R., Kerr, A.C., 2010. Mantle plume or slab window?: physical and
 604 geochemical constraints on the origin of the Caribbean oceanic plateau. *Earth*
 605 *Sci. Rev.*, 98, 283-293.

606 Hauff, F., Hoernle, K., Schmincke, H.U., Werner, R., 1997. A Mid Cretaceous
 607 origin for the Galápagos hotspot: volcanological, petrological, and
 608 geochemical evidence from Costa Rican oceanic crustal segments. *Geol. Rund.*
 609 86, 141-155.

610 Hauff, F., Hoernle, K., Tilton, G., Graham, D. W., Kerr, A. C., 2000a. Large volume
 611 recycling of oceanic lithosphere over short time scales: geochemical
 612 constraints from the Caribbean Large Igneous Province. *Earth Planet Sci.*
 613 *Lett.* 174, 247-263.

614 Hauff, F., Hoernle, K., van den Bogaard, P., Alvarado, G., Garbe-Schönberg, D.,
 615 2000b. Age and geochemistry of basaltic complexes in western Costa Rica:
 616 Contributions to the geotectonic evolution of Central America. *Geochem.*
 617 *Geophys. Geosyst.* 1 (5). <http://dx.doi.org/10.1029/1999GC000020>.

618 Herzberg, C., 2011a. Identification of Source Lithology in the Hawaiian and Canary
 619 Islands: Implications for Origins. *J. Petrol.* 52, 113-146.

620 Herzberg, C., Asimow, P. D., 2008. Petrology of some oceanic island basalts:
621 PRIMELT2.XLS software for primary magma calculation. *Geochem. Geophys.*
622 *Geosyst.* 9 (9). <http://dx.doi.org/10.1029/2008GC002057>

623 Herzberg, C., Asimow, P.D., Ionov, D.A., Vidito, C., Jackson, M.G., Geist, D., 2013. Nickel
624 and helium evidence for melt above the core-mantle boundary. *Nature* 493,
625 393-397.

626 Herzberg, C., Cabral, R.A., Jackson, M.D., Vidito, C., Day, J.M.D., Hauri, E., 2014.
627 Phantom Archean crust in Mangaia hotspot lavas and the meaning of
628 heterogeneous mantle. *Earth Planet. Sci. Lett.* 396, 97-106.

629 Herzberg, C., Gazel, E., 2009. Petrological evidence for secular cooling in mantle
630 Plumes. *Nature*. 458, 619-623.

631 Herzberg, C., O'Hara, M.J., 2002. Plume-associated ultramafic magmas of
632 Phanerozoic age. *J. Petrol.* 43, 1857-1883.

633 Hirose, K., 2006. Postperovskite phase transition and its geophysical implications.
634 *Rev. Geophys.* 44, 2005RG000186, RG3001.

635 Hoernle, K., Hauff, F., van den Bogaard, P., 2004, 70 m.y. history (139–69 Ma) for the
636 Caribbean large igneous province. *Geology*. 32 (8), 697-700.

637 Hoernle, K., Werner, R., Phipps Morgan, J., Garbe-Schönberg, C.D., Bryce, J.G., Mrazek,
638 J., 2000. Existence of complex spatial zonation in the Galápagos plume for at
639 least 14 m.y. *Geology*. 28, 435-438.

640 Hoernle, K., van den Bogaard, P., Werner, R., Lissinna, B., Hauff, F., Alvarado, G.,
641 Garbe-Schönberg, D., 2002. Missing history (16–71 Ma) of the Galápagos
642 hotspot: Implications for the tectonic and biological evolution of the

- 643 Americas. *Geology*. 30 (9), 795-798.
- 644 Hofmann, A. W., 1997. Mantle geochemistry: the message from oceanic volcanism.
645 *Nature*. 385, 219-229.
- 646 Ito, G., Mahoney, J.J., 2005. Flow and melting of a heterogeneous mantle: 1. Method
647 and importance to the geochemistry of ocean island and mid-ocean ridge
648 basalts. *Earth. Planet. Sci. Lett.*, 230, 29-46.
- 649 Jackson, M.G., Dasgupta, R., 2008. Compositions of HIMU, EM1, and EM2 from global
650 trends between radiogenic isotopes and major elements in ocean island
651 basalts. *Earth. Planet. Sci. Lett.*, 276, 175-186.
- 652 Jackson, M. G., Hart, S. R., Saal, A. E., Shimizu, N., Kurz, M. D., Blusztajn, J. S.,
653 Skovgaard, A. C., 2008. Globally elevated titanium, tantalum, and niobium
654 (TITAN) in ocean island basalts with high $^3\text{He}/^4\text{He}$. *Geochem. Geophys.*
655 *Geosyst.* 9. <http://dx.doi.org/10.1029/2007GC001876>.
- 656 Jackson, M.G., Weis, D., Huang, S., 2012. Major element variations in Hawaiian shield
657 lavas: Source features and perspectives from global ocean island basalt (OIB)
658 systematics. *Geochem. Geophys. Geosyst.* 13.
659 <http://dx.doi:10.1029/2012GC004268>
- 660 Jicha, B. R., Brown, F. H., 2014. An age on the Korath Range and the viability of
661 $^{40}\text{Ar}/^{39}\text{Ar}$ dating of kaersutite in Late Pleistocene volcanics, Ethiopia, Quat.
662 *Geochron.* 21, 53-57.
- 663 Johnson, D. M., Hooper, P. R., Conrey, R. M., 1999. XRF Analysis of rocks and
664 minerals for major and trace elements on a single low dilution Li-tetraborate
665 fused bead. *Advances in X-ray Anal.* 41, 843-867.

- 666 Kamber, B., Collerson, K., 2000. Zr/Nb systematics of ocean island basalts
 667 reassessed—the case for binary mixing. *J. Petrol.* 41 (7), 1007-1021.
- 668 Kerr, A. C., 2005. Oceanic LIPs: The Kiss of Death: *Elements*. 1, 289-292.
- 669 Kerr, A. C., Marriner, G. F., Arndt, N., Tarney, J., Nivia, A., Saunders, A. D., Duncan,
 670 R. A., 1996a. The petrogenesis of Gorgona komatiites, picrites and basalts:
 671 new field, petrographic and geochemical constraints. *Lithos.* 37, 245-260.
- 672 Kerr, A. C., Tarney, J., Marriner, G. F., Klaver, G. T., Saunders, A. D., Thirlwall, M.
 673 F., 1996b. The geochemistry and petrogenesis of the late-Cretaceous picrites
 674 and basalts of Curaçao, Netherlands Antilles: a remnant of an oceanic
 675 plateau: *Cont. Mineral. Petrol.* 124 (1), 29-43.
- 676 Kuiper, K.F., Deino, A., Hilgen, F.J., Krijgsman, W., Renne, P.R., Wijbrans, J.R., 2008.
 677 Synochronizing Rock Clocks of Earth History. *Science* 320, 500-504.
- 678 Kumagai, I., Davaille, A., Kurita, K., Stutzmann, E., 2008. Mantle plumes: Thin, fat,
 679 successful, or failing? Constraints to explain hot spot volcanism through time
 680 and space. *Geophys. Res. Lett.* 35 (16).
 681 <http://dx.doi.org/10.1029/2008GL035079>
- 682 Kurz, M. D., Jenkins, W. J., Hart, S. R., 1982. Helium isotopic systematics of oceanic
 683 islands and mantle heterogeneity. *Nature.* 297, 43-47.
- 684 Kurz, M. D., Geist, D., 1999. Dynamics of the Galápagos hotspot from helium isotope
 685 geochemistry. *Geochim. Cosmochim. Acta.* 63 (23/24), 4139-4156.
- 686 Kurz, M. D., Rowland, S. K., Curtice, J., Saal, A. E., Naumann, T., 2014. Eruption Rates
 687 for Fernandina Volcano, The Galápagos. In Harpp, K.S., Mittelstaedt, E.,
 688 d'Ozouville, N., Graham, D.W. (Eds.), *A New Chronology at the Galápagos*

- 689 Hotspot Center. John Wiley & Sons Inc., Chapter 4, pp. 41-54.
- 690 Li, C., Ripley, E.M, 2010. The relative effects of composition and temperature on
691 olivine-liquid Ni partitioning: Statistical deconvolution and implications for
692 petrologic modeling. *Chem. Geol.* 275, 99-104.
- 693 Loewen, M. W., Duncan, R. A., Kent, A. J. R., Krawl, K., 2013. Prolonged plume
694 volcanism in the Caribbean Large Igneous Province: New insights from
695 Curaçao and Haiti. *Geochem. Geophys. Geosyst.* 14 (10).
696 <http://dx.doi.org/10.1002/ggge.20273>.
- 697 Lonsdale, P., 2005. Creation of the Cocos and Nazca plates by fission of the Farallon
698 plate. *Tectonophys.* 404 (3-4), 237-264.
- 699 Mahoney, J. J., Coffin, F. F., 1997. Large Igneous Provinces: Continental, oceanic, and
700 planetary flood volcanism, Washington. *Am. Geophys. Union Mono.* 100.
701 <http://dx.doi.org/10.1029/GM100>.
- 702 Matzen, A.K., Baker, M.B., Beckett, J.R., Stolper, E.M., 2013. The temperature and
703 pressure dependence of nickel partitioning between olivine and silicate melt.
704 *J. Petrol.* 54, 2521-2545.
- 705 Meschede, M., Frisch, W., 1994, Geochemical characteristics of basaltic rocks from
706 the Central American ophiolites. *Profil.* 7, 71-85.
- 707 Morgan W. J., 1971. Convection plumes in the lower mantle. *Nature* 230, 42–43.
- 708 Morgan, J. P., 2001. Thermodynamics of pressure release melting of a veined plum
709 pudding mantle. *Geochem. Geophys. Geosys.* 2, 2000GC000049.
- 710 McDonough, W., 1991. Partial melting of subducted oceanic crust and isolation of its
711 residual eclogitic lithology. *Phil. Trans. Royal Soc. London. Series A. Physical*

- 712 and Engineering Sciences. 335 (1638), 407-418.
- 713 Min K., Mundil R., Renne P. R., Ludwig K. R., 2000. A test for systematic errors in
714 $^{40}\text{Ar}/^{39}\text{Ar}$ geochronology through comparison with U/Pb analysis of a 1.1Ga
715 rhyolite. *Geochem. Cosmochem. Acta.* 64, 73-98.
- 716 Morgan, W. J., 1972. Deep Mantle Convection Plumes and Plate Motion. *Am. Assoc.*
717 *Petrol. Geol. Bull.* 56 (2), 203-213.
- 718 Naumann, T.R., Geist, D.J., 2002. Petrology and Geochemistry of Cerro Azul Volcano
719 and the Origin of the Petrologic Diversity of the Western Galápagos Shield
720 Volcanoes: An inter-volcano comparison of Cerro Azul, Alcedo, and Sierra
721 Negra. *J. Petrol.* 43, 859-883.
- 722 Nolet, G., Karato, S., Montelli, R., 2006. Plume fluxes from seismic tomography. *Earth*
723 *Planet. Sci. Lett.* 248 (3-4), 685-699.
- 724 O'Connor, J. M., Stoffers, P., Wijbrans, J. R., Worthington, T. J., 2007. Migration of
725 widespread long-lived volcanism across the Galápagos Volcanic Province:
726 Evidence for a broad hotspot melting anomaly? *Earth Planet. Sci. Lett.* 263
727 (3-4), 339-354.
- 728 O'Donnell, T. H., Presnall, D.C., 1980, Chemical Variations of the Glass and Mineral
729 Phases in Basalts Dredged from 25-Degrees-30-Degrees-N Along the Mid-
730 Atlantic Ridge. *Am. J. Sci.* 280, 845-868.
- 731 O'Hara, M.J. 1968, The bearing of phase equilibria studies in synthetic and natural
732 systems on the origin of basic and ultrabasic rocks. *Earth Sci. Rev.* 4, 69-133.

- 733 Pertermann, M., Hirschmann, M.M., 2003. Anhydrous partial melting experiments on
 734 MORB-like eclogite: phase relations, phase compositions and mineral-melt
 735 partitioning of major elements at 2-3 GPa. *J. Petrol.* **44**, 2173-2201.
- 736
- 737 Pindell, J. L., Barrett, S. F., 1990. Geological evolution of the Caribbean region: a plate
 738 tectonic perspective: The Caribbean region. In: Dengo, G., Case, J.E., (Eds.),
 739 Boulder, Colorado, Geological Society of America, *The Geology of North*
 740 *America*, H, pp. 405-432.
- 741 Richards, M. A., Duncan, R. A., Courtillot, V. E., 1989. Flood basalts and hot-spot
 742 tracks: Plume heads and tails. *Science*. 246 (4926), 103-107.
- 743 Ricolleau, A., Perrillat, J.-P., Fiquet, G., Daniel, I., Matas, J., Addad, A., Menguy, N.,
 744 Cardon, H., Mezouar, M., Guignot, N., 2010. Phase relations and equation of
 745 state of a natural MORB: implications for the density profile of subducted
 746 oceanic crust. *J. Geophys. Res.* 115, B08202, doi:10.1029/2009JB006709.
- 747 Rudnick, R. L., Barth, M., Horn, I., McDonough, W. F., 2000. Rutile-bearing refractory
 748 eclogites: missing link between continents and depleted mantle. *Science*. 287
 749 (5451), 278-281.
- 750 Rudnick, R.L., Gao, S., 2003. The Composition of the Continental Crust, In: Holland,
 751 H.D. and Turekian, K.K. (Eds.), *Treatise on Geochemistry*, Elsevier-Pergamon,
 752 Oxford. 3, pp. 1-64.
- 753 Saunders, A. D., 2005. Large Igneous Provinces: Origin and Environmental
 754 Consequences. *Elements*. 1, 259-263.
- 755 Sinton, C. W., Duncan, R. A., Denyer, P., 1997. Nicoya Peninsula, Costa Rica: A single

756 suite of Caribbean oceanic plateau magmas. *J. Geophys. Res.* 102 (B7), 15507-
 757 15520.

758 Sinton, C. W., Duncan, R. A., Storey, M., Lewis, J., Estrada, J. J., 1998. An oceanic flood
 759 basalt province within the Caribbean plate: *Earth Planet. Sci. Lett.* 155, 221-
 760 235.

761 Shorttle, O., MacLennan, J., 2011. Compositional trends of Icelandic basalts:
 762 Implications for short-length scale lithological heterogeneity in mantle
 763 plumes. *Geochem. Geophys. Geosyst.* 12, Q11008,
 764 doi:10.1029/2011GC003748.

765 Sobolev, A.V., Hofmann, A.W., Sobolev, S.V., Nikogosian I.K., 2005. An olivine-free
 766 mantle source of Hawaiian shield basalts. *Nature.* 434, 590-597.

767 Sobolev, A. V., Hofmann, A. W., Kuzmin, D. V., Yaxley, G. M., Arndt, N. T., Chung, S. L.,
 768 Danyushevsky, L. V., Elliott, T., Frey, F. A., Garcia, M. O., Gurenko, A. A.,
 769 Kamenetsky, V. S., Kerr, A. C., Krivolutsкая, N. A., Matvienkov, V. V.,
 770 Nikogosian, I. K., Rocholl, A., Sigurdsson, I. A., Sushchevskaya, N. M., Teklay,
 771 M., 2007. The amount of recycled crust in sources of mantle-derived melts.
 772 *Science.* 316, 412-417.

773 Stolper, E., Sherman, S., Michael, G., Baker, M., Seaman, C., 2004. Glass in the
 774 submarine section of the HSDP2 drill core, Hilo, Hawaii. *Geochem. Geophys.*
 775 *Geosyst.* 5 (7). <http://dx.doi.org/10.1029/2003GC000553>.

776 Stracke, A., Hofmann, A. W., Hart, S. R., 2005. FOZO, HIMU, and the rest of the mantle
 777 zoo: *Geochem. Geophys. Geosyst.* 6 (5).
 778 <http://dx.doi.org/10.1029/2004GC000824>.

- 779 Straub, S.M., LaGatta, A.B., Pozzo, A.L.M.-D. Langmuir, C.H., 2008. Evidence from
780 high Ni olivines for a hybridized peridotite/pyroxenite source for orogenic
781 andesites from the central Mexican Volcanic Belt. *Geochem. Geophys. Geosys.*
782 9, Q03007, doi: 10.1029GC001583.
- 783 Thompson, G., Bryan, W.G., Melson, W.G., 1980. Geological and geophysical
784 investigation of the Mid-Cayman-Rise-Spreading-Center - geochemical
785 variation and petrogenesis of basalt glasses. *J. Geol.* 88 (1), 41-55.
- 786 Toplis, M. J., 2005. The thermodynamics of iron and magnesium partitioning
787 between olivine and liquid: criteria for assessing and predicting equilibrium
788 in natural and experimental systems. *Contrib. Mineral. Petrol.* 149 (1), 22-39.
- 789 Tolstikhin, I., Hofmann, A. W., 2005. Early crust on top of the Earth's core. *Phys.*
790 *Earth Planet. Interiors.* 148 (2-4), 109-130.
- 791 Torsvik, T. H., Burke, K., Steinberger, B., Webb, S. J., Ashwal, L. D., 2010. Diamonds
792 sampled by plumes from the core-mantle boundary. *Nature.* 466 (7304), 352-
793 355.
- 794 Vidito, C., Herzberg, C., Gazel, E., Geist, D., Harpp, K., 2013. Lithological structure of
795 the Galápagos Plume. *Geochem. Geophys. Geosyst.* 14 (10). [http://dx.](http://dx.doi.org/10.1002/ggge.20270)
796 [doi.org/10.1002/ggge.20270](http://dx.doi.org/10.1002/ggge.20270).
- 797 Walter, M. J., 1998. Melting of Garnet Peridotite and the Origin of Komatiite and
798 Depleted Lithosphere, *J. Petrol.*, 39 (1), 29-60.
- 799 Wegner, W., Worner, G., Harmon, R.S., Jicha, B.R., 2010. Magmatic history and
800 evolution of the Central American Land Bridge in Panama since Cretaceous
801 times. *Geol. Soc. Am. Bull.* 123, 703-724.

Werner, R., and Hoernle, K., 2003. New volcanological and volatile data provide strong support for the continuous existence of Galápagos Islands over the past 17 million years. *Int. J. Earth Sci.* 92 (6), 904-911.

White, W. M., Albarede, F., Telouk, P., 2000. High-precision analysis of Pb isotope ratios by multi-collector ICP-MS. *Chem. Geol.* 167, 257-270.

White, W. M., McBirney, A. R., Duncan, R. A., 1993. Petrology and geochemistry of the Galápagos Islands: Portrait of a pathological mantle plume. *J. Geophys. Res.* 98 (B11), 19533-19563.

Zindler, A., Hart, S., 1986. Chemical Geodynamics. *Ann. Rev. Earth Planet. Sci.* 14, 493-571.

FIG. CAPTIONS

Fig. 1. A) Tectonic location of the study area (modified from Denyer and Gazel, 2009). B) Map of the accreted oceanic complexes along the coast of Costa Rica and Panama (modified from Denyer and Gazel, 2009). C) Geological map of the Quepos terrane showing sample locations, structures, and areas comprised by basalts and picrites modified from Duran (2013).

Fig. 2. A) Chemical classification of samples using the total alkali versus silica diagram (TAS). Literature data from Meschede and Frishe (1994), Sinton et al. (1997), and Hauff et al. (2000b). B) CaO-MgO discrimination diagram between peridotite and pyroxenite derived magmas from Herzberg and Asimow (2008). This

model was developed for accumulated fractional melting of peridotite KR4003, and is validated by the fact that no experimental melt compositions from a pure peridotite source plot below the discrimination line. The grey shaded region represents primary magmas of fertile peridotite produced by accumulated fractional melting. Additional data from Stolper et al. (2004) for Mauna Kea and from White et al., 1993; Hauff et al., 1997; Allan and Simkin, 2000; Hauff et al., 2000a,b; Hoernle et al., 2000; Blichert-Toft and White, 2001; Geist et al., 2002; Naumann et al., 2002; Werner et al., 2003; O'Connor et al., 2007; Wegner et al., 2010 for Galápagos related lavas including accreted terranes in Central America and parts of the CLIP including Nicoya and Curacao.

Fig. 3. Multi-element spider diagram normalized to primitive mantle (McDonough and Sun, 1995). Note how the new data from Quepos overlap with the current Galápagos tracks; the Cocos, Carnegie, and Coiba Ridges. Additional data from White et al., 1993; Hauff et al., 1997; Allan and Simkin, 2000; Hauff et al., 2000; Hoernle et al., 2000; Blichert-Toft and White, 2001; Geist et al., 2002; Naumann et al., 2002; Werner et al., 2003; O'Connor et al., 2007; Wegner et al., 2010 for Galápagos Domains.

Fig. 4. Mg-numbers versus Ni and Ca contents and Fe/Mn for calculated olivines and high-precision olivines from ~ 90 Ma Curacao picrites. A Mg-number is defined as $100\text{MgO}/(\text{MgO} + \text{FeO})$ in mole per cent. Black forms = calculated Ni and Ca in olivines that crystallize from all primary melts derived from a fertile peridotite

source having 1960 ppm Ni, 3.45 wt% CaO and 0.13 wt% MnO (Herzberg, 2011a); the grey field in panel A represents $\pm 1\sigma$ uncertainty for Ni (Herzberg et al., 2013). The Curacao primary magma composition was computed from lava sample CR372T92 reported by Klaver (1987). The full range of model olivine Ca for peridotite-source melts far exceeds the 1700-2000 ppm range that is typically observed for MORB, komatiite, and other olivines of a peridotite source provenance; similarly the upper Fe/Mn model bound of 70 exceeds Fe/Mn of observed olivines of a peridotite source provenance. Curacao olivine compositions define trends towards peridotite-source olivines (see text).

Fig. 5. Mg-numbers versus Ni and Ca contents and Fe/Mn for calculated olivines and high-precision olivines from picrites of the ~ 70 Ma Quepos terrane. Examples of olivines from a peridotite source primary magma and its derivatives are given by the green cross and arrow, computed from lava sample CR372T92 reported by Tournon (1984). Quepos olivine compositions are not consistent with derivation from a normal peridotite source. Notice how the new Quepos data is within range of Mauna Kea data from Sobolev et al. (2007) interpreted as olivines that crystallized from a pyroxenite-derived primary magma. The trends can be explained by complex olivine+clinopyroxene fractional crystallization or magma recharge (Supplementary Materials for detailed information). Independent of the explanation, these crystallization trends do not extrapolate to peridotite source primary magmas.

Fig. 6. Source projected Pb and Nd isotopes of picrite and basalt samples from the

CLIP and Quepos terrane. Most Quepos samples fall within the Central Galápagos Domain. Shaded Galápagos Domains for the present day archipelago from Hoernle et al. (2000).

Fig. 7. A) Galápagos plume secular cooling curve modified from Herzberg and Gazel, (2009) using a least squares approximation. Note the drastic decrease in T_p from the time of LIP emplacement to approximately 70-60 Ma. The actual T_p range exhibited for each occurrence like the CLIP at a specific time is typically seen in almost all OIB and LIP occurrences, as reported in Herzberg & Asimow (2008) and Herzberg & Gazel (2009); these authors have attributed the T_p range as a record of plume thermal heterogeneity, higher T_p in the core and lower T_p at the periphery. B) Evolution of the Nb/Nb^* vs. time (Ma) from Galápagos related basalts. Nb/Nb^* is defined as $Nb_N/\sqrt{(Th_N \times La_N)}$ where N means normalized to a primitive mantle composition from McDonough and Sun, 1995. Average Nb/Nb^* for MORB is calculated from the average MORB composition from Gale et al., 2013. Color bars behind symbols represent the age range of Galápagos units. Notice as the plume cools down the Nb/Nb^* ratio increased to a max at 0 Ma (Data from White et al., 1993; Hauff et al., 1997; Allan and Simkin, 2000; Hauff et al., 2000; Hoernle et al., 2000; Blichert-Toft and White, 2001; Geist et al., 2002; Naumann et al., 2002; Werner et al., 2003; O'Connor et al., 2007; Wegner et al., 2010).

Figure 1

FIGURES

Figure 2

Figure 3

Figure 4

Figure 5

Figure 6

Figure 7