



# Origin of $^3\text{He}/^4\text{He}$ ratios in HIMU-type basalts constrained from Canary Island lavas

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## ABSTRACT

New helium isotope and abundance measurements are reported for olivine and clinopyroxene phenocrysts from HIMU-type (high- $\mu$  = elevated  $^{238}\text{U}/^{204}\text{Pb}$ ) lavas and xenoliths spanning the stratigraphies of El Hierro and La Palma, Canary Islands. Some pyroxene phenocrysts have suffered post-eruptive modification, either by less than 1% assimilation of crustal-derived He, or by closed-system ageing of He. Olivine phenocrysts record mantle source  $^3\text{He}/^4\text{He}$  compositions, with the average  $^3\text{He}/^4\text{He}$  for La Palma olivine ( $7.6 \pm 0.8R_A$ , where  $R_A$  is the atmospheric  $^3\text{He}/^4\text{He}$  ratio of  $1.38 \times 10^{-6}$ ) being within uncertainty of those for El Hierro ( $7.7 \pm 0.3R_A$ ), and the canonical mid-ocean ridge basalt range (MORB:  $8 \pm 1R_A$ ). The new helium isotope data for El Hierro and La Palma show no distinct correlations with whole-rock  $^{87}\text{Sr}/^{86}\text{Sr}$ ,  $^{143}\text{Nd}/^{144}\text{Nd}$ ,  $^{187}\text{Os}/^{188}\text{Os}$ , or Pb isotopes, but  $^3\text{He}/^4\text{He}$  ratios for La Palma lavas correlate with  $^{18}\text{O}/^{16}\text{O}$  measured for the same phenocryst populations. Despite limited  $^3\text{He}/^4\text{He}$  variations for El Hierro and La Palma, their He–O isotope systematics are consistent with derivation from mantle sources containing distinct recycled oceanic basaltic crust (El Hierro) and gabbroic lithosphere (La Palma) components that have mixed with depleted mantle, and a high- $^3\text{He}/^4\text{He}$  component ( $>9.7R_A$ ) in the case of La Palma. The new data are consistent with models involving generation of compositionally and lithologically (e.g., pyroxenite, eclogite, peridotite) heterogeneous mantle sources containing recycled oceanic crust and lithosphere entrained within upwelling high- $^3\text{He}/^4\text{He}$  mantle that has been severely diluted by interaction with depleted mantle. We propose that the noble gas systematics of HIMU-type lavas and ocean island basalts (OIB) in general, are most simply interpreted as being controlled by the most gas-rich reservoir involved in mixing to generate their mantle sources. In this scenario, HIMU and enriched mantle (EM) sources are dominated by depleted mantle, or high- $^3\text{He}/^4\text{He}$  mantle, because recycled crust and lithosphere have low He concentrations. Consequently, high- $^3\text{He}/^4\text{He}$  OIB would predominantly reflect derivation from a less depleted mantle source with sub-equal to higher He contents than depleted mantle. The available coupled He–O isotope systematics measured for OIB lavas are consistent with this hypothesis.

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## 1. Introduction

Mantle-derived HIMU-type ocean island basalts (OIB; high- $\mu$  = elevated  $^{238}\text{U}/^{204}\text{Pb}$ ), which possess both radiogenic  $^{206}\text{Pb}/^{204}\text{Pb}$  (19 to 22) and  $^{187}\text{Os}/^{188}\text{Os}$  (0.13 to 0.18) signatures, represent some of the most compelling evidence for the return of ancient (1 to >2 Ga) subducted oceanic crust and lithosphere to Earth's surface (e.g., Day et al., 2009, 2010; Hauri and Hart, 1993; Hofmann and White, 1982). Further corroborating evidence in this respect has been the recent identification of low- $^{18}\text{O}/^{16}\text{O}$  ratios in Canary Islands lavas with radiogenic  $^{206}\text{Pb}/^{204}\text{Pb}$  ratios, indicating the presence of recycled HIMU-type materials that have witnessed low-temperature hydrothermal alteration at Earth's surface (Day et al., 2010). HIMU-type OIB form part of a broader spectrum of elemental and long-lived isotopic compositions recognised in OIB (e.g., Dasgupta et al., 2010; Zindler

and Hart, 1986), which collectively indicate long-term preservation (>1 Ga) of depleted and enriched components, some of which have subducted origins, in a compositionally heterogeneous mantle. Lithological heterogeneity in OIB mantle sources, generated by metasomatism to form pyroxenite ( $\pm$  eclogite)–peridotite mixtures, has been proposed to explain the compositions of OIB olivine populations (Sobolev et al., 2005, 2007), correlations of olivine composition with Sr–Nd–Pb isotopes (Gurenko et al., 2009), and the O–Os isotope systematics of OIB (Day et al., 2009). The mounting evidence for lithological, elemental and isotopic heterogeneity in the mantle, through recycling and/or metasomatic melt reaction processes, represents critical information for understanding mantle dynamics, the efficiency of convective stirring, and for unification of both geochemical and geophysical models of mantle evolution.

Helium isotopes are a proven tool for constraining the origin of mantle heterogeneity, with present-day mantle degassing of  $^3\text{He}$  representing one of the most fundamental observations of terrestrial evolution (e.g., Clarke et al., 1969; Lupton and Craig, 1975; Mamyrin et al., 1969). This is because, unlike  $^4\text{He}$ , which is dominantly a

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radiogenic by-product of U and Th decay,  $^3\text{He}$  is mainly primordial in origin, being trapped during the earliest stages of planetary accretion. Consequently, He isotopes provide direct evidence for the preservation of relatively undegassed reservoirs in the present-day mantle. High- $^3\text{He}/^4\text{He}$  ratios (up to  $50R_A$ ; Stuart et al., 2003), measured for some hotspot-related lavas, implicate the presence of reservoirs in Earth's mantle with high time-integrated  $^3\text{He}/(\text{U} + \text{Th})$ . Despite geophysical complexities (e.g., Ballentine et al., 2002), high- $^3\text{He}/^4\text{He}$  ratios measured in lavas have classically been considered the result of sampling of a partially degassed lower mantle (e.g., Allègre and Moreira, 2004; Allègre et al., 1983; Brandenburg et al., 2008; Gonnermann and Mukhopadhyay, 2009; Hamn et al., 2009; Kaneoka, 1983; Kurz et al., 1982; O'Nions and Oxburgh, 1983) underlying a more degassed upper mantle represented by mid-ocean ridge basalt helium isotope compositions (MORB;  $8 \pm 1R_A$ ; Graham, 2002).

A persistent issue for understanding the distribution, preservation and sampling of helium isotopic variations in OIB reservoirs has been the higher-than-expected  $^3\text{He}/^4\text{He}$  ratios measured for HIMU-type lavas. If such lavas derive from ancient recycled oceanic crust or lithosphere, which is predicted to be relatively rich in U and Th and poor in He (e.g., Hiyagon, 1994), then the  $^3\text{He}/^4\text{He}$  ratios of such a reservoir should be highly radiogenic ( $R_A < 0.1$ ) after transit times in the mantle between 1 to 2 Ga (e.g., Hofmann, 1997). This is not the case, however, as HIMU-type OIB localities have  $^3\text{He}/^4\text{He}$  values higher than predicted, spanning the MORB range – from 4.3 to  $9.5R_A$  (Barfod et al., 1999; Christensen et al., 2001; Graham et al., 1992; Hanyu and Kaneoka, 1997; Hanyu et al., 1999; Hilton et al., 2000; Parai et al., 2009; Vance et al., 1989). Significantly, the range of measured  $^3\text{He}/^4\text{He}$  ratios for OIB samples does not extend to low values ( $< 0.1R_A$ ). To account for this apparent paradox, models involving diffusive equilibration of He between depleted MORB mantle (DMM) and subducted components (Hanyu and Kaneoka, 1997; Hanyu et al., 1999; Hart et al., 2008), closed-system ageing of high- $^3\text{He}/^4\text{He}$  He reservoirs (Barfod et al., 1999), preservation of MORB-like He in recycled oceanic lithosphere (Moreira and Kurz, 2001), and admixtures of recycled material and a relatively undegassed source with DMM (Hilton et al., 2000; Parai et al., 2009), have all been proposed to explain  $^3\text{He}/^4\text{He}$  ratios of HIMU-type OIB.

In this contribution we address the origin of the higher-than-expected  $^3\text{He}/^4\text{He}$  ratios measured in HIMU-type OIB through consideration of the He systematics of a suite of geochemically and petrologically well-characterised lavas and predominantly cumulate xenoliths from the islands of El Hierro and La Palma, Canary Islands. These samples have incompatible element abundances and ratios and O–Sr–Nd–Os–Pb isotope signatures consistent with mixtures of HIMU-type recycled oceanic crust and lithosphere and DMM (Day et al., 2009, 2010; Gurenko et al., 2006; Marcantonio et al., 1995), and which have been interpreted to have developed through interaction between metasomatised mantle and pyroxenite–peridotite mixtures (Day et al., 2009, 2010; Gurenko et al., 2009).

## 2. Samples and methods

Comprehensive petrological and geochemical characterisations of samples analysed in this study are provided elsewhere (Day et al., 2010). Samples were selected for He isotopic analysis based on the abundance of olivine and clinopyroxene phenocrysts, and because the selected samples, in conjunction with prior measurements presented by Hilton et al. (2000), provide access to different series present in the stratigraphic records of both El Hierro and La Palma. Approximately 0.4 to 2 kg of whole-rock per sample was crushed to a maximum pebble diameter of less than 0.3 cm, coned and quartered, and then a  $> 0.2$  kg aliquot was wet sieved to coarse, 500  $\mu\text{m}$  and 200  $\mu\text{m}$  fractions and dried overnight at 110 °C. The sieved fractions were examined under a binocular microscope and fresh olivine and clinopyroxene grains were hand-separated predominantly from the largest size

fraction. Separates were ultrasonicated at 50 °C, sequentially leached for 20 min in 1 M HCl, and then in distilled water, prior to brief treatment in ethanol and drying at 110 °C. After the chemical pre-treatment, mineral separates were further purified to remove any remaining matrix or altered material. Separates underwent final ultrasonic cleaning treatment in ethanol and were dried prior to loading into two on-line crushing devices.

Helium isotope analyses were performed at the Fluid and Volatiles Laboratory, Geosciences Research Division, Scripps Institution of Oceanography. Helium trapped within inclusions of olivine and clinopyroxene phenocrysts were released by crushing under vacuum. Blanks were obtained by operating the crusher (~70 beats per minute) for the same amount of time used for crushing of samples (150 s). Short crush times avoided the release of extraneous He contained in crystal lattice sites (Graham et al., 1998; Hilton et al., 1993; Scarsi, 2000). The average vacuum line blank ( $\leq 0.2 \times 10^{-9} \text{ cm}^3 \text{ STP}^4\text{He}$ ) represented on average 0.26% and 2% of measured sample  $^3\text{He}$  and  $^4\text{He}$ , respectively. In the worst case, the total blank represented 2% of  $^3\text{He}$  and 10% of  $^4\text{He}$  (LP10 olivine). Helium liberated by crushing was expanded into a stainless steel preparation line and sequentially exposed for a total of 30 min to a Ti getter at 750 °C, decreasing to 400 °C, a liquid N-cooled charcoal trap, a SAES® Zr–Al getter operated at room temperature, and a He-cooled charcoal trap, to isolate helium from neon. Following isolation, helium was expanded into a dual collector noble gas mass spectrometer (MAP 215E) for  $^3\text{He}/^4\text{He}$  ratio analysis using a peak-jumping protocol as described by Shaw et al. (2006). Helium-4 was detected using a Faraday cup,  $^3\text{He}$  with a channeltron multiplier operating in ion counting mode at a mass resolution  $> 600$ . Raw helium isotope ratios were normalised using standard aliquots from Murdering Mudpots, Yellowstone National Park ( $16.45R_A$ ) and SIO Pier air ( $1R_A$ ), under the same preparation and measurement procedure used for samples. Neon was monitored during each run to correct  $^3\text{He}/^4\text{He}$  ratios for any air contamination effects ( $< 2\%$  in raw  $R/R_A$  for all samples). Concentration calculations were performed relative to the air standard, which uses a calibrated volume to pipette known amounts into the preparation system.

## 3. Results

A total of 27 lava samples were selected for  $^3\text{He}/^4\text{He}$  and He concentration analyses. Measurements were made on both olivine and clinopyroxene separates for 15 samples: only olivines were used for the remainder (Table 1). Although the majority of olivine–clinopyroxene pairs are broadly in helium isotopic equilibrium ( $n = 11$ ), there are a number of samples that possess clinopyroxene with lower  $^3\text{He}/^4\text{He}$  ratios than co-existing olivine (Fig. 1a). This contrasts with previous He isotopic measurements made for La Palma olivines and clinopyroxenes ( $n = 11$ ) by Hilton et al. (2000), where only one olivine–clinopyroxene pair was found to be in helium isotopic disequilibrium, within the analytical uncertainties. Both olivine and clinopyroxene possess a similar range of He concentrations released during crushing, with abundances typically falling within a factor of five for mineral phases of the same sample. The exception is the pyroxenite xenolith LP13A, which has notably higher olivine He concentrations than the co-existing clinopyroxene (Fig. 1b). Duplicate measurements of olivine and/or clinopyroxene were performed for EH01 and EH13 and show good agreement in  $^3\text{He}/^4\text{He}$ , with values varying by less than 6%, but measured  $^4\text{He}$  concentrations varied by an order of magnitude (e.g., EH01). Differences in He contents for replicate measurements are typical for phenocryst populations analysed by crushing (e.g., Graham, 2002).

The total range of measured  $^3\text{He}/^4\text{He}$  ratios for El Hierro (4.2 to  $8.2R_A$ ; average =  $7.3 \pm 1.0R_A$ ,  $n = 24$ ) and La Palma (4.3 to  $9.7R_A$ ; average =  $7.2 \pm 1.2R_A$ ,  $n = 21$ ) phenocrysts from the new dataset are similar, with the average  $^3\text{He}/^4\text{He}$  composition being identical within uncertainty for the two islands. The new results are in excellent

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