



# Heterogeneous mantle-derived helium isotopes in the Canary Islands and other ocean islands

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## ABSTRACT

Consistent  $^3\text{He}/^4\text{He}$  ratios have been measured for >25 years in geothermal fluids and gases from Cumbre Vieja, La Palma ( $9.4 \pm 0.1 R_A$ , where  $R_A$  is the  $^3\text{He}/^4\text{He}$  of air), and Teide, Tenerife ( $6.8 \pm 0.3 R_A$ ), Canary Islands. Both locations are characterized by similar  $\text{CO}_2/^3\text{He}$  ( $\sim 2$  to  $4 \times 10^9$ ), mantle-like  $\delta^{13}\text{C}$  ( $-3.3\text{\textperthousand}$  to  $-4.4\text{\textperthousand}$ ) and  $\text{CO}_2$  output ( $0.1$ – $0.2 \times 10^{10}$  mol yr $^{-1}$ ). Helium isotopic differences between the islands cannot be explained by differential aging and  $^4\text{He}$  ingrowth in their mantle sources. Instead, distinct He reservoirs exist, with a high- $\mu$  (HIMU)-type mantle source for La Palma and a more enriched mantle, with possible lithospheric mantle influence, for Tenerife. Geothermal samples from the Canary Islands record a present-day He distribution distinct from higher  $^3\text{He}/^4\text{He}$  in olivine from older eastern Canary Island lavas, indicating temporal variability in sources. Comparison of geothermal sample data versus olivine, pyroxene, and glass He isotope data for the Canary Islands, Azores, Cape Verde, Hawaiian islands, and Iceland reveals generally good correspondence, even across >1 m.y. of stratigraphy. However, in addition to the Canary Islands, there are examples of inter-island heterogeneity for He isotopes at Hawaii, the Azores, and within Iceland, preserved in hydrothermal samples, minerals, and glasses. In particular, in northwest Iceland, olivine separates from older lavas preserve higher  $^3\text{He}/^4\text{He}$  than present-day geothermal samples from the same region. This difference likely reflects a reduced mantle-derived  $^3\text{He}$  input to Icelandic magmatism since the Miocene. Temporal variability in  $^3\text{He}/^4\text{He}$ , assessed using geothermal and geological materials in conjunction, offers a powerful tool for examining heterogeneity and temporal evolution of mantle sources at intraplate volcanoes.

## INTRODUCTION

Ocean-island basalts (OIBs) and intraplate basalts are known to sample mantle reservoirs with variable  $^3\text{He}/^4\text{He}$  ratios (<5 to  $50 R_A$ , where  $R_A$  is the  $^3\text{He}/^4\text{He}$  of air,  $1.38 \times 10^{-6}$ ; e.g., Kurz et al., 1982; Stuart et al., 2003). The cause of helium isotopic variability and the ways in which it is related to magmatism are not well understood, but differences relative to mid-oceanic ridge basalts (MORBs;  $8 \pm 1 R_A$ ; Graham, 2002) imply the existence of both primordial  $^3\text{He}$ -rich, less-degassed reservoirs and high  $(\text{U} + \text{Th})/^3\text{He}$  recycled mantle sources, relative to the depleted MORB mantle (DMM). Less-degassed, low  $(\text{U} + \text{Th})/^3\text{He}$  reservoirs likely occur deeper than the mantle transition zone (e.g., Timmerman et al., 2019), although the mechanisms by which

these sources are preserved over Earth history are uncertain. Conversely, high  $(\text{U} + \text{Th})/^3\text{He}$  sources can occur in both oceanic and continental lithosphere, leading to highly heterogeneous  $^3\text{He}/^4\text{He}$  ratios (Moreira and Kurz, 2001; Day et al., 2005, 2015).

Helium isotopic analysis of OIBs occurs both through liberation of gas within olivine and pyroxene grains or glass (hereafter termed minerals and glass), or through direct sampling of geothermal fluids and gases. An advantage of minerals and glass is that they trap He during crystallization of melts and can potentially preserve the He isotope character of their sources over long time periods (>100 m.y.; e.g., Stronci et al., 2017). Disadvantages are the addition of radiogenic and cosmogenic helium over time. Geothermal samples preserve the characteristics of mantle-derived volatiles at the present day, but, as with minerals and glass, they

are susceptible to air contamination and other modification processes. These different media provide complementary information on both the spatial and temporal  $^3\text{He}/^4\text{He}$  variation for individual and global OIB localities. Here, we present new noble gas,  $\text{CO}_2$ , and  $\delta^{13}\text{C}$  values, where  $\delta^{13}\text{C} = ([^{13}\text{C}/^{12}\text{C}_{\text{sample}}]/[^{13}\text{C}/^{12}\text{C}_{\text{standard}}] - 1) \times 1000$ , for geothermal samples from La Palma and Tenerife (Canary Islands) to demonstrate helium isotope heterogeneity between the islands, reflecting different mantle sources. Comparison of hydrothermal samples with minerals and glass from global OIB examples also implies significant temporal and spatial helium isotope heterogeneity in their mantle sources.

## SAMPLES AND METHODS

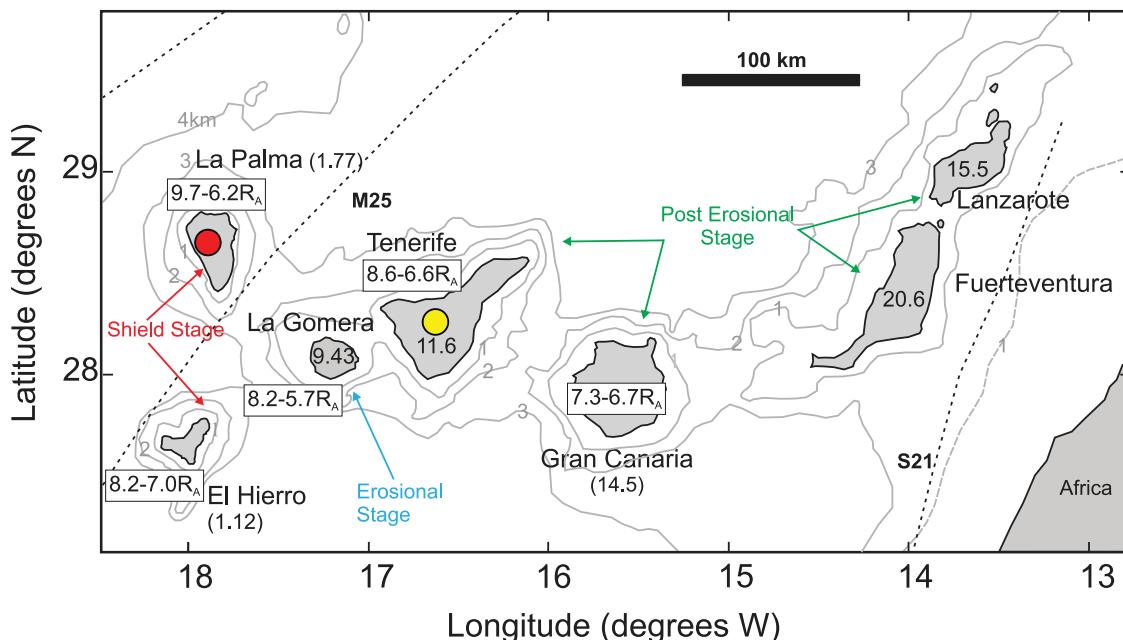
We collected geothermal samples in summer 2006 at the peak of Teide (3718 m,  $28^{\circ}16'21''\text{N}$ ,  $16^{\circ}38'34''\text{W}$ ), Tenerife, and from the Cumbre Vieja ( $28^{\circ}42'06''\text{N}$ ,  $17^{\circ}53'38''\text{W}$ ), La Palma (Fig. 1). Samples were collected in evacuated low-He diffusivity Corning 1720 glass flasks following procedures to minimize atmospheric contamination (Hilton et al., 2002). Methods used for analysis of fluids and gas samples are provided in detail in the Supplementary Material<sup>1</sup>. An ultrahigh-vacuum (UHV) extraction line was used to process all samples (Kulnogoski and Hilton, 2002). Samples were acidified with phosphorus pentoxide for complete release of  $\text{CO}_2$ , with abundances corresponding to total dissolved inorganic carbon (DIC). Water vapor was isolated, after which the condensable gas fraction (dominantly  $\text{CO}_2$ ) was trapped. A non-condensable gas split was collected in a Pyrex glass break-seal flask for transfer to a VG5400 mass spectrometer. The remaining noncondensable gas was purified, and a calibrated aliquot of this fraction was expanded into an AR-glass break-seal flask for transfer to a MAP 215 noble gas mass spectrometer for He isotope analysis.

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<sup>1</sup>Supplemental Material. Detailed methods and supplemental figures. Please visit <https://doi.org/10.1130/GEOL.S.12915671> to access the supplemental material, and contact editing@geosociety.org with any questions.

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**Figure 1.** Map of the Canary Islands with sampling locations for Teide (yellow circle) and Cumbre Vieja (red circle). Maximum and minimum He isotope compositions (as  ${}^3\text{He}/{}^4\text{He}$  [ $\text{R}/\text{R}_A$ ]) are given in boxes for each island and are from Gurenko et al. (2006) and Day and Hilton (2011). Oldest erupted lava ages are given in parentheses (in Ma) or within island boundaries, and are summarized in Day et al. (2010). Current growth stages of the Canary Islands are shown, as are ocean-floor depths (km) and paleomagnetic anomalies M25 and S21. S21 lies close to the juncture of the continental-oceanic lithosphere boundary.

The  $\text{CO}_2$  fraction was condensed into a Pyrex break-seal flask for transfer to a dedicated  $\text{CO}_2$  clean-up line. Helium, Ne, and C isotopes were measured using previously detailed protocols, with identical external reproducibility (Füri et al., 2010; Barry et al., 2014).

## RESULTS

We analyzed six hydrothermal fluid samples for He, Ne, and  $\text{CO}_2$  compositions: two from La Palma, and four from the summit of Teide (Table 1). Contents of  ${}^4\text{He}$  for five of the samples were  $>185 \times 10^9 \text{ cm}^3 \text{ g}^{-1}$ , with only one sample from Teide having lower contents ( $2.4 \times 10^9 \text{ cm}^3 \text{ g}^{-1}$ ). Contents of  ${}^{22}\text{Ne}$  were 2 to 125 times lower ( $1.4\text{--}59 \times 10^9 \text{ cm}^3 \text{ g}^{-1}$ ). Helium isotope compositions between Teide (6.1–6.6  $\text{R}_A$ ) and La Palma (8.8–9.3  $\text{R}_A$ ) were distinct, whereas  $\text{CO}_2/{}^3\text{He}$  ratios ( $2\text{--}7.6 \times 10^9$ ),  $\delta^{13}\text{C}$ , and Ne isotope compositions were similar. The  $\text{CO}_2/{}^3\text{He}$  ratios were similar to MORB, arc, and OIB (Iceland/Hawaii) geothermal compositions with mantle-like  $\delta^{13}\text{C}$  values (Fig. 2). Neon isotope compositions were close to atmospheric composition, reflecting the incorporation of air-like Ne into the geothermal samples, and so Ne isotopes are not discussed further.

## HELIUM ISOTOPE VARIATIONS IN THE CANARY ISLANDS

Hydrothermal samples from Teide (Tenerife) and Cumbre Vieja (La Palma) have been sampled for more than 25 yr (e.g., Pérez et al., 1994; Hilton et al., 1997; Alonso et al., 2016; this study). This time series for La Palma and Tenerife reveals relatively consistent mantle-derived signatures for both locations in terms of  $\text{CO}_2/{}^3\text{He}$  and carbon isotope compositions, and for  ${}^3\text{He}/{}^4\text{He}$ . The helium isotope composi-

tions have remained at  $9.4 \pm 0.1 \text{R}_A$  for La Palma geothermal samples and  $6.8 \pm 0.3 \text{R}_A$  at Teide, implying a consistent mantle He and  $\text{CO}_2$  flux to both islands. The effects of seawater or air contamination, which can be significant at some locations (e.g., Santorini, Greece; Moreira et al., 2019), are negligible, while degassing processes may have a limited effect on  $\text{CO}_2/{}^3\text{He}$  and C isotope compositions ( $\delta^{13}\text{C}$  of  $-3.3\text{--}4.3\text{‰}$  in Teide geothermal samples). Prior workers have estimated  $\text{CO}_2$  ( $\sim 70\text{--}84 \text{ kT yr}^{-1}$ ) with  ${}^3\text{He}$  fluxes from Teide of  $0.4\text{--}0.9 \text{ mol yr}^{-1}$ , and  ${}^3\text{He}$  fluxes for Cumbre Vieja of  $0.6\text{--}0.7 \text{ mol yr}^{-1}$  (Padrón et al., 2012; Alonso et al., 2019), translating to  $0.12\text{--}0.14$  and  $0.16\text{--}0.19 \times 10^{10} \text{ mol yr}^{-1} \text{ CO}_2$  for Cumbre Vieja and Teide, respectively, and representing a minor fraction of total global volcanic degassing (e.g., Kerrick, 2001).

La Palma and Tenerife geothermal samples have  ${}^3\text{He}/{}^4\text{He}$  values outside of analytical uncertainties from each other. These differences are not evident from mineral  ${}^3\text{He}/{}^4\text{He}$  ratios (La Palma = 9.7–6.2  $\text{R}_A$ ; Tenerife and eastern Canary Islands = 8.6–6.6  $\text{R}_A$ ), which show considerable overlap (Fig. 1). A possibility to explain helium isotope variations in geothermal samples is that the mantle source beneath Tenerife has evolved due to aging and ingrowth of  ${}^4\text{He}$ . This would imply that the reservoir remained closed to other He sources for an extended time period. The most logical time of any possible closure would be after the main shield-building phase on Tenerife, when magmatism waned.

To model closed-system  ${}^4\text{He}$  production, we assumed a maximum closure age equal to the oldest age of shield-building lavas on Tenerife (11.6 Ma), and a magmatic Th/U ratio of 3.6 (Day et al., 2010), and U contents similar to mantle peridotites (Day et al., 2015). Start-

ing with a  ${}^3\text{He}/{}^4\text{He}$  ratio of 9.4  $\text{R}_A$  (La Palma), there is negligible change in He isotopic composition over 12 m.y. for reservoirs assumed to have between 0.1 and  $100 \times 10^9 \text{ cm}^3 \text{ g}^{-1} \text{ He}$ , with significant changes to the  ${}^3\text{He}/{}^4\text{He}$  ratio only occurring at very low initial gas contents ( $<0.1 \times 10^9 \text{ cm}^3 \text{ g}^{-1} \text{ He}$ ). While radiogenic  ${}^4\text{He}$  ingrowth has undoubtedly occurred to some extent in the sources of Canary Island magmas, it can be ruled out as the primary cause of He isotope variation, given the higher estimated He contents of Canary Island mantle sources (Day and Hilton, 2011), and the impracticality of closed-system He isotope evolution in magmatically active regions (Day et al., 2015).

An alternate cause for lower  ${}^3\text{He}/{}^4\text{He}$  ratios in Tenerife compared with La Palma geothermal fluids is a difference in mantle source reservoir. Both La Palma and El Hierro have  ${}^3\text{He}/{}^4\text{He}$  consistent with coming from a mantle source similar to, or having lower  $(\text{U} + \text{Th})/{}^3\text{He}$ , than DMM (Day and Hilton, 2011; Gurenko et al., 2006). The host lavas have high- $\mu$  (HIMU)-type compositions, based on Sr-Nd-Pb-Os isotope systematics (Day et al., 2010). Similar HIMU-type compositions are also observed in the eastern Canary Islands (Tenerife, Gran Canaria); however, trends to lower  ${}^{143}\text{Nd}/{}^{144}\text{Nd}$  and  ${}^{206}\text{Pb}/{}^{204}\text{Pb}$  and higher  ${}^{87}\text{Sr}/{}^{86}\text{Sr}$  and  ${}^{207}\text{Pb}/{}^{204}\text{Pb}$  have been attributed to contributions from an enriched mantle component (e.g., Hoernle et al., 1991; Simonsen et al., 2000; Gurenko et al., 2006).

The cause of an increased enriched mantle source contribution in the eastern Canary Islands has been suggested to occur from addition of continental lithospheric mantle (CLM) inherited from thermal erosion and delamination of the West African margin during rifting of

TABLE 1. HELIUM, NEON, AND CARBON ISOTOPIC SYSTEMATICS OF CANARY ISLAND GEOTHERMAL SAMPLES

Sample	Island	Location	${}^4\text{He}$ ( $10^{-9} \text{ cm}^3 \text{ g}^{-1}$ )	${}^3\text{He}/{}^4\text{He}$ $R/R_A$	$X$	${}^3\text{He}/{}^4\text{He}$ $R_c/R_A$	$\pm$	${}^{22}\text{Ne}$ ( $10^{-9} \text{ cm}^3 \text{ g}^{-1}$ )	$\pm$	${}^{20}\text{Ne}/{}^{22}\text{Ne}$	$\pm$	${}^{21}\text{Ne}/{}^{22}\text{Ne}$	$\pm$	${}^{20}\text{Ne}/{}^{22}\text{Ne}$	$\pm$	${}^{13}\text{C}$ ( $\text{\textperthousand}$ )	$\pm$	${}^4\text{He}/{}^{22}\text{Ne}$ ( $\text{\textperthousand}$ )	$\pm$
CAN-11	La Palma	Dos Aguas	186	6	9.27	5322	9.27	0.11	1.49	0.04	9.993	0.040	0.0290	0.0015	1.96	-3.8	125		
CAN-14	La Palma	Dos Aguas	235	7	8.78	519	8.79	0.10	2.25	0.07	9.861	0.039	0.0291	0.0015	5.30	-4.4	104		
CAN-9	Tenerife	Teide Summit 1	282	8	6.20	647	6.21	0.11	37	0.10	9.930	0.040	0.0293	0.0020	3.90	-4.3	8		
CAN-10	Tenerife	Teide Summit 1	241	7	6.63	1064	6.63	0.08	2.35	0.07	9.990	0.040	0.0293	0.0020	7.01	-4.1	103		
CAN-12	Tenerife	Teide Summit 2	2.4	0.07	5.89	21	6.09	0.08	1.44	0.04	10.010	0.040	0.0290	0.0014	6.69	-3.3	2		
CAN-13	Tenerife	Teide Summit 2	211	6	6.04	44	6.14	0.08	59	2	9.613	0.038	0.0284	0.0014	7.62	-3.7	4		

Note: Errors for He isotope ratios are at the 2 $\sigma$  level. Errors on  $\text{CO}_2/{}^3\text{He}$  are  $\pm 5\%$ . X is the air-normalized  $\text{He}/\text{Ne}$  ratio. This value was multiplied by 1.228 (assuming a recharge temperature of 10 °C) when calculating  $R_c/R_A$ , where  $R_A$  is the  $\text{He}/{}^4\text{He}$  of air =  $1.38 \times 10^{-6}$  and  $R_c$  is the air-corrected ratio.

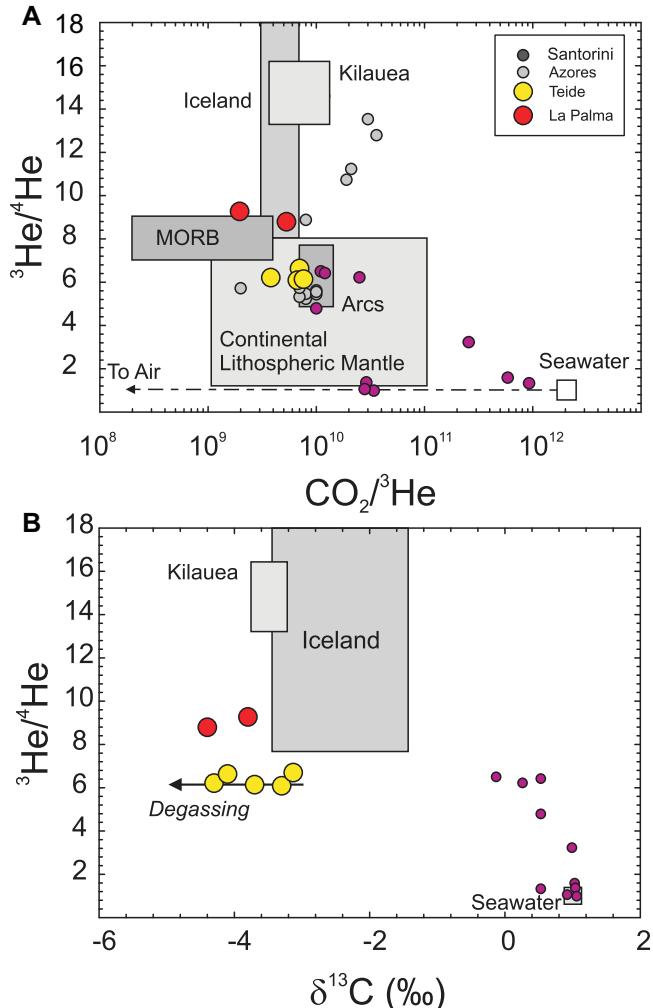


Figure 2. Relationships between helium isotope compositions and  $\text{CO}_2/{}^3\text{He}$  (A) and  $\delta^{13}\text{C}$  (B) in geothermal samples from the Canary Islands compared with other localities, mantle reservoirs, seawater, and air. Data sources for Hawaii, Iceland, Santorini, and mantle reservoirs are Hilton et al. (1997), Jean-Baptiste et al. (2009), Day et al. (2015), Harðardóttir et al. (2018), and Moreira et al. (2019). MORB—mid-oceanic ridge basalt.

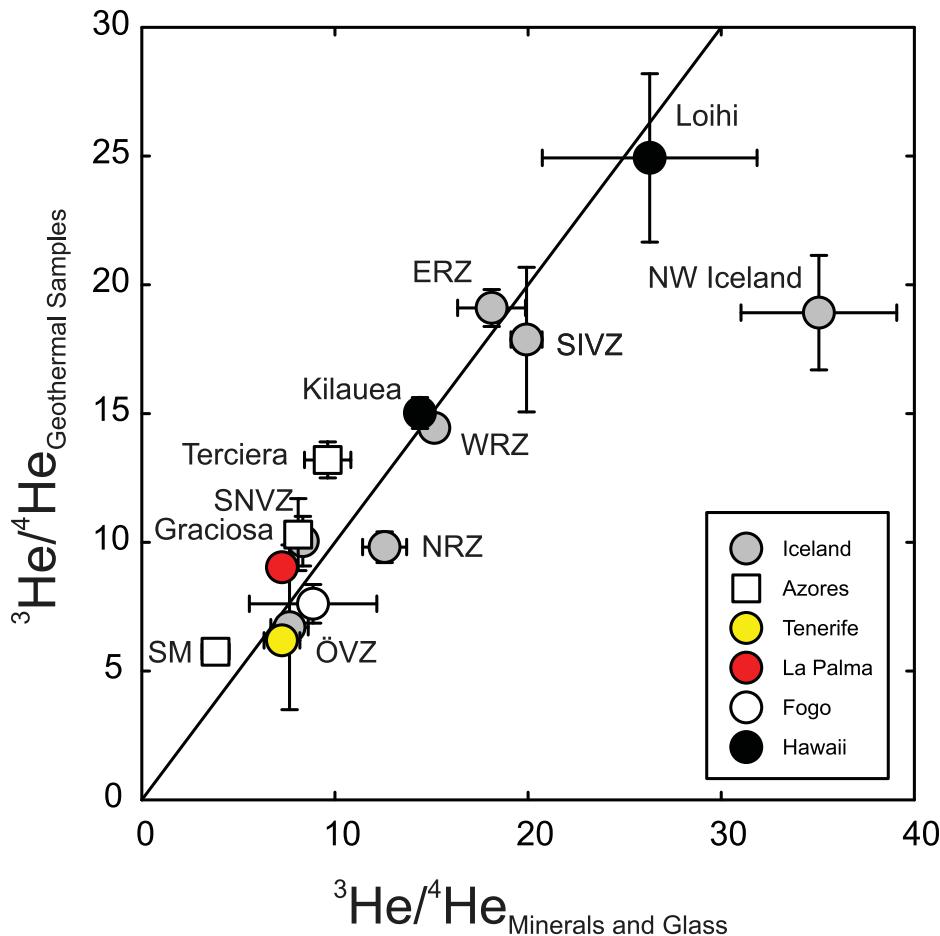
Pangea (Hoernle et al., 1991). Such an origin is consistent with the helium isotope characteristics of Teide, which has  $\text{He}-\text{CO}_2$  characteristics broadly akin to CLM (Fig. 2A; Day et al., 2015). Higher  ${}^3\text{He}/{}^4\text{He}$  ratios than those measured for Teide have been measured in Tenerife and Gran Canaria olivine separates from older lavas, showing a HIMU-type composition akin to the western Canary Islands (e.g., Hoernle et al., 1991). Geothermal samples from La Palma and Tenerife therefore support a changing source composition model for the eastern Canary Islands, indicating that differing mantle sources are being sampled and are persisting beneath the western and eastern Canary Islands to the present day.

#### HELIUM ISOTOPE VARIATIONS IN OIB

For more than 40 years, studies have been systematically performed to obtain the helium isotopic compositions of OIBs, typically by measuring gas trapped within minerals and glass. These studies have shown a wide range in  ${}^3\text{He}/{}^4\text{He}$ , from low ratios ( $<8R_A$ ) in some OIBs, such as São Miguel (e.g., Moreira et al., 2012), to values in excess of 30–47 $R_A$  in Hawaii,

Samoa, and Iceland (e.g., Kurz et al., 1982, 2004; Graham, 2002; Macpherson et al., 2005; Jackson et al., 2007; Harðardóttir et al., 2018).

It has been established that minerals have He contents that are generally not as high as in their host magmas. Olivine separates with high  ${}^3\text{He}/{}^4\text{He}$  from Samoa have gas contents dominated by He and  $\text{CO}_2$ -rich fluid inclusions trapped in only a few grains, suggesting that conditions favoring high He concentrations in olivine are rare (Horton et al., 2019). An alternative is to compare mineral or glass He contents with geothermal sample He contents from the same locality. For example, He contents measured in La Palma olivine grains ( $1.4\text{--}170 \times 10^9 \text{ cm}^3 \text{ g}^{-1}$ ) are typically lower than He contents of geothermal fluids ( $>180 \times 10^9 \text{ cm}^3 \text{ g}^{-1}$ ). The lower He contents in minerals support the concept of decoupling of C and  ${}^3\text{He}$  in olivine through He diffusion (Horton et al., 2019), and they indicate that, even as primary magmatic He contents become diluted during geothermal processes, geothermal samples are a better proxy for  $\text{CO}_2/{}^3\text{He}$  in OIBs. In turn, La Palma and Tenerife geothermal sample compositions are consistent with Canary Island OIBs, having similar  $\text{CO}_2/{}^3\text{He}$  to MORB (Fig. 2).



**Figure 3.** Helium isotope variations for minerals and glass versus hydrothermal samples from the same locations. Shown are average compositions and  $2\sigma$  errors and the 1:1 line. Data for the Canary Islands are from this study, Day and Hilton (2011), and Gurenko et al. (2006). Other data: Azores data (SM—Saô Miguel) are from Moreira et al. (1999, 2012) and Jean-Baptiste et al. (2009); Fogo (Cape Verde Islands) data are from Christensen et al. (2001) and Heilweil et al. (2009); Hawaii data are from Kurz et al. (1982), Sedwick et al. (1994), and Hilton et al. (1997); Iceland localities are from the filtered compilation of Harðardóttir et al. (2018), where NRZ—Northern rift zone; WRZ—Western rift zone; ERZ—Eastern rift zone; SNVZ—Snæfellsnes volcanic zone; SIVZ—South Iceland volcanic zone; ÖVZ—Öræfajökull volcanic zone.

There are several OIB localities where a direct comparison can also be made between He isotopic compositions in geothermal samples and those in minerals and glass. These locations include the Canary Islands of La Palma and Tenerife, three of the Azorean islands (São Miguel, Terceira, Graciosa), Cape Verde (Fogo), Hawaii (Kilauea and Loihi Seamount), and numerous localities in Iceland. We compared the average compositions of the geothermal samples at the localities and the variance of the  ${}^3\text{He}/{}^4\text{He}$  ratios (as  $2\sigma$  errors) with those of minerals and glass considered to be related to the geothermal localities (Fig. 3). Most locations suffer from small number statistics, yet they show that, for the majority, mineral and glass compositions have average compositions that are broadly similar to corresponding geothermal samples.

La Palma and the three Azores locations have helium isotope ratios in geothermal samples generally higher than minerals. This can be explained if concomitant degassing and

assimilation processes preferentially affected mineral He isotope compositions (e.g., Day and Hilton, 2011). In contrast, in northwest Iceland, recently sampled hydrothermal media have lower  ${}^3\text{He}/{}^4\text{He}$  than the average composition of minerals taken from Miocene lavas located there. These differences likely relate to the lavas being fed by magmas from a mantle source with higher  ${}^3\text{He}/{}^4\text{He}$  than that beneath Iceland today (e.g., Harðardóttir et al., 2018).

Analyses of helium isotope variations through  $>2500$  m of lava stratigraphy at Mauna Kea (Hawaiian Islands) have revealed changes from a generally high (up to  $\sim 25\text{R}_\text{A}$ ) to a lower  ${}^3\text{He}/{}^4\text{He}$  composition ( $8\text{R}_\text{A}$ ) with time (Kurz et al., 2004). These variations have been interpreted to reflect heterogeneities in an upwelling mantle plume over short ( $\sim 15$  k.y.) and intermediate ( $\sim 0.38$  m.y.) time scales (Kurz et al., 2004). Comparisons of geothermal fluids with minerals and glass show that helium isotope compositional variability also occurs in Iceland, from the

eruption of the first exposed lavas to the present day. Collectively, these results support “pulsed” high  ${}^3\text{He}/{}^4\text{He}$  in localities such as Iceland, as well as Hawaii, and they also suggest changing He compositions with time, even in low- ${}^3\text{He}/{}^4\text{He}$  OIBs, such as the Canary Islands.

Helium isotope variability on million-year time scales occurs at hotspot localities for which there are ample data: Iceland, Hawaii, and now the Canary Islands. Denser sampling might yield similar temporal trends elsewhere and could shed light on mantle heterogeneity and the geochemical evolution of mantle hotspots in general. For example, ca. 60 Ma lavas from the North Atlantic igneous province (Stuart et al., 2003) and ca. 132 Ma lavas associated with the Parana-Etendeka continental flood basalt (southern Africa and South America; Stronck et al., 2017) have been shown to have high  ${}^3\text{He}/{}^4\text{He}$ . In the absence of geothermal fluids for these localities, it is possible that the He compositions preserved in their minerals reflect distinct periods of high- ${}^3\text{He}/{}^4\text{He}$  magmatism. Where it is possible to do so, combining helium isotopic measurements of geothermal and geological samples is a promising approach for evaluating temporal isotopic trends at hotspot localities.

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