

## Helium isotopes at Rungwe Volcanic Province, Tanzania, and the origin of East African Plateaux

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[1] We report helium isotope ratios ( $^3\text{He}/^4\text{He}$ ) of lavas and tephra of the Rungwe Volcanic Province (RVP) in southern Tanzania. Values as high as  $15R_A$  ( $R_A = \text{air } ^3\text{He}/^4\text{He}$ ) far exceed typical upper mantle values, and are the first observation of plume-like ratios south of the Turkana Depression which separates the topographic highs of the Ethiopia and Kenya domes. The African Superplume - a tilted low-velocity seismic anomaly extending to the core-mantle boundary beneath southern Africa - is the likely source of these high  $^3\text{He}/^4\text{He}$  ratios. High  $^3\text{He}/^4\text{He}$  ratios at RVP together with similarly-high values along the Main Ethiopian Rift and in Afar provide compelling evidence that the African Superplume is a feature that extends through the 670-km seismic discontinuity and provides dynamic support - either as a single plume or via multiple upwellings - for the two main topographic features of the East Africa Rift System as well as heat and mass to drive continuing rift-related magmatism.

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### 1. Introduction

[2] The East African Rift System (EARS) represents the classic example of present-day rifting and nascent stages of continental break-up [*Kampunza and Lubala*, 1991; *Yirgu et al.*, 2006]. Rifting and associated volcanism traverse two prominent and broad (>1000 km wide) uplifted plateaux in the region - the Ethiopia and Kenya domes (Figure 1). The Ethiopia Dome hosts the Afar and Main Ethiopian rifts which bifurcate south of the low-lying Turkana Depression into the Western and Kenyan rifts. These rifts cross the Kenya Dome, partially encircling the Archean Tanzania craton. Gravity [*Ebinger et al.*, 1989a] and topographic [*Nyblade and Robinson*, 1994] constraints together with

seismic imaging of the East African mantle [*Ritsema et al.*, 1999; *Nyblade et al.*, 2000] reveal that dynamic support of the elevated topography of the domes is derived from upwelling convective mantle.

[3] However, the helium isotope geochemistry ( $^3\text{He}/^4\text{He}$ ) of the EARS presents a contrasting picture of the origins and scale of mantle convection. Along the Main Ethiopian Rift and Afar,  $^3\text{He}/^4\text{He}$  ratios are significantly greater than values characteristic of ambient upper mantle [*Marty et al.*, 1996; *Scarsi and Craig*, 1996] - as sampled by mid-ocean ridge basalt (MORB) [*Graham*, 2002], and are consistent with a deep (lower) mantle plume origin for convective uplift of the Ethiopia Dome. On the other hand, Kenya Dome  $^3\text{He}/^4\text{He}$  ratios do not exceed the MORB value [*Darling et al.*, 1995; *Pik et al.*, 2006; *Hopp et al.*, 2007; *Tedesco et al.*, 2010] prompting suggestions of a different scale of mantle convection supporting the Kenya plateau, restricted to the uppermost mantle only [*Pik et al.*, 2006; *Montagner et al.*, 2007]. Here, we target lavas and tephra from Rungwe Volcanic Province (RVP), Tanzania at the southernmost extreme of the Kenya Dome and report the first observation of  $^3\text{He}/^4\text{He}$  ratios >MORB south of the Turkana Depression. The data provide unambiguous evidence for a deep contribution to magma genesis at RVP and likely throughout eastern Africa.

### 2. Samples and Analytical Techniques

[4] The RVP is one of four volcanically-active regions of the Western Rift and represents the southernmost expression of Cenozoic volcanism along the entire EARS (Figure 1). It is located in southwest Tanzania and consists of two volcanic series: Older Extrusives, formed by the earliest eruptions of the Ngozi and Katete central volcanoes at ~7 Ma, and Younger Extrusives formed by Rungwe, Tukuyu, Kiejo and Ngozi volcanoes, starting in the mid-Pliocene and continuing to the present-day [*Harkin*, 1960; *Ebinger et al.*, 1989b; *Furman*, 1995]. We analyzed a total of 31 lava and tephra samples covering both volcanic series for whole rock and olivine (OL) mineral chemistry, and helium isotope ratios ( $^3\text{He}/^4\text{He}$ ) (Tables S1 and S2 of the auxiliary material).<sup>1</sup> All samples are alkalic in composition and include alkali basalts, basanites, nephelinites and a picrite and trachy-basalt. Olivine (OL) and/or clinopyroxene (CPX) crystals from each sample were crushed in vacuo to determine  $^3\text{He}/^4\text{He}$  ratio and He abundance characteristics ([He]). We report a total of 52 He analyses of RVP mineral separates divided between the 4 volcanoes of the Younger Extrusives (n = 45) and

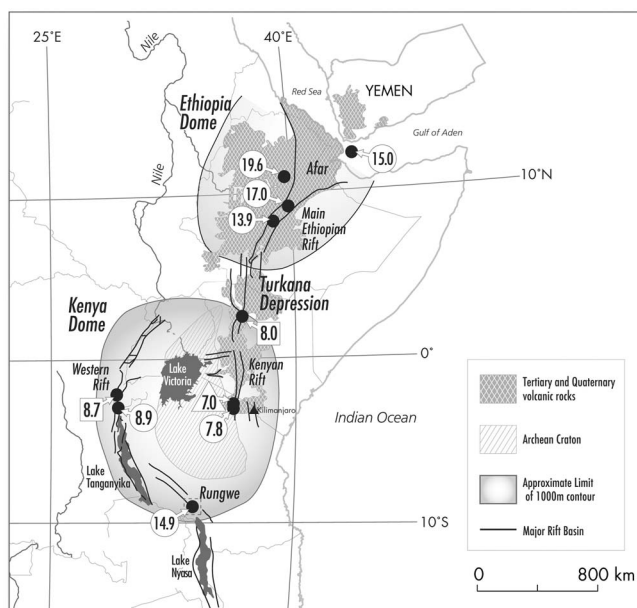
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**Figure 1.** The East African Rift System (EARS) showing the Ethiopia and Kenya domes [modified from Rogers, 2006]. The highest  $^3\text{He}/^4\text{He}$  ratios ( $R/R_A$  notation) are plotted for different segments of the EARS, using circles (lavas), squares (geothermal fluids) and triangles (xenoliths). In the Kenya Dome region,  $^3\text{He}/^4\text{He}$  ratios do not exceed the canonical MORB value ( $8 \pm 1 R_A$ ) with the exception of Rungwe at  $9^\circ\text{S}$  (this work). The most southerly locality in the Ethiopia Dome region to have  $^3\text{He}/^4\text{He} > \text{MORB}$  is Shalla ( $13.9 R_A$ ) located at  $7^\circ\text{N}$  in the Main Ethiopian Rift [Scarsi and Craig, 1996]. High  $^3\text{He}/^4\text{He}$  ratios (up to  $19.6 R_A$ ) extend through the Afar region to the Gulf of Aden, the Red Sea and Yemen [Marty et al., 1996; Moreira et al., 1996].

2 volcanoes of the Older Extrusives ( $n = 7$ ). Nine samples were analyzed for OL only, one sample for CPX only and 21 for co-genetic OL-CPX pairs. We supplement the RVP database with new He isotope data on peridotite xenoliths from 5 localities on or close to the Archean Tanzania craton in northern Tanzania (Table S3 of the auxiliary material).

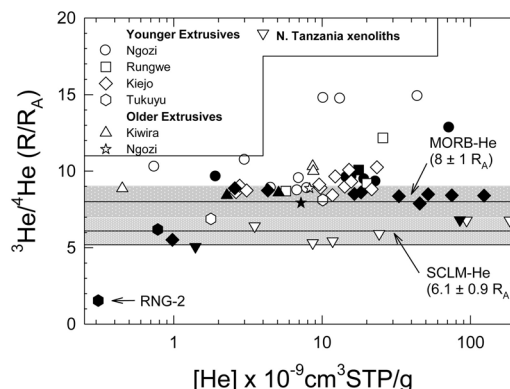
### 3. Results

[5] In Figure 2 we plot the  $^3\text{He}/^4\text{He}$  ratio of each mineral separate (reported in the  $R/R_A$  notation where  $R = \text{sample } ^3\text{He}/^4\text{He}$  corrected for a minor air-derived component and  $R_A = \text{air } ^3\text{He}/^4\text{He}$ ) versus its He concentration ( $[\text{He}]$  in units of  $\text{cm}^3\text{STP/g}$  mineral). There are three noteworthy features of the He results. First, the highest measured  $^3\text{He}/^4\text{He}$  ratios greatly exceed the canonical range of  $8 \pm 1 R_A$ , diagnostic of the upper mantle (MORB-source) [Graham, 2002]. Indeed, a total of 17 (out of 31) RVP samples have an OL and/or CPX  $^3\text{He}/^4\text{He}$  ratio that lies above the MORB range, at the 1-sigma level of analytical uncertainty. Significantly, the high  $^3\text{He}/^4\text{He}$  component is found in 3 out of 4 volcanoes of the Younger Extrusives (Ngozi, Rungwe and Kiejo) and in the Kiwira Series of the Older Extrusives, and thus is widespread in time and space. If the MORB threshold is raised to  $10 R_A$  then 7 samples fall above this value (at the same level of analytical uncertainty) and they cover the same volcanic series. Second, 13 of the RVP samples fall

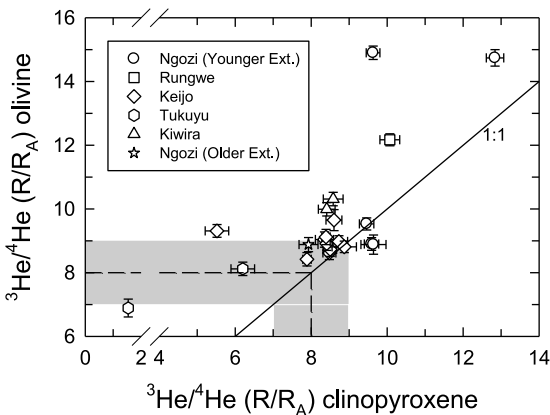
within the range normally associated with MORB (i.e.,  $8 \pm 1 R_A$ ): this He component was sampled in all six of the Rungwe volcanic series. Finally, one sample only (Mafiga, Tukuyu volcano) has a  $^3\text{He}/^4\text{He}$  ratio coincident with sub-continental lithosphere mantle (SCLM =  $6.1 \pm 0.9 R_A$ ) [Gautheron and Moreira, 2002]. Notably, all of the northern Tanzania peridotite xenoliths fall within or overlap the SCLM range. In this respect, results reported here extend the coverage of the SCLM He database to the African continent. Dismissing post-eruptive modification of the He results reported here (see auxiliary material), we conclude that the high  $^3\text{He}/^4\text{He}$  component found throughout the RVP is the first instance of  $^3\text{He}/^4\text{He}$  ratios  $> \text{MORB}$  for the entire Western Rift of the EARS (Figure 1). Indeed, RVP is the only region of Cenozoic rifting/volcanism throughout the entire Kenya Dome region (i.e., the highlands south of the Turkana Depression) where  $^3\text{He}/^4\text{He}$  ratios approach values found along the Main Ethiopian Rift [Marty et al., 1996; Scarsi and Craig, 1996].

### 4. Discussion

[6] The high  $^3\text{He}/^4\text{He}$  ratios ( $> \text{MORB}$ ) at RVP reveal the presence of a high time-integrated  $^3\text{He}/(\text{U}+\text{Th})$  ratio in the mantle source region, commonly viewed as a contribution from a more primitive, or less degassed, portion of the mantle that has experienced long-term isolation from the convecting upper mantle, as sampled by MORB [Graham, 2002]. The lowermost mantle is the obvious candidate for such a source, with transfer to the surface facilitated by upwelling mantle plumes [Courtillot et al., 2003]. Our He results are consistent with the findings of Hopp et al. [2007], who reported plume-like Ne isotope values for 5 xenoliths from Chyulu Hills, southern Kenya. Taken together, the



**Figure 2.** Plot of  $^3\text{He}/^4\text{He}$  ratios ( $R/R_A$  notation) of RVP phenocrysts versus total helium concentration ( $[\text{He}]$  released by crushing ( $\text{cm}^3\text{STP/g}$  mineral phase)). Open and filled symbols are olivines (OL) and clinopyroxenes (CPX), respectively. Analytical errors are smaller than symbols. The MORB  $^3\text{He}/^4\text{He}$  range represents ambient convecting upper mantle, as sampled at oceanic spreading centers [Graham, 2002]. Sub-continental lithospheric mantle (SCLM) represents the isolated and non-convecting mantle reservoir forming the lowermost segment of lithospheric plates. Its average  $^3\text{He}/^4\text{He}$  ratio is derived from a compilation of phenocrysts and xenoliths worldwide [Gautheron and Moreira, 2002]. Note: sample  $\text{RNG-2}_{\text{CPX}}$  is dominated by radiogenic He as a result of crustal contamination (see text).



**Figure 3.** Plot of olivine (OL) versus clinopyroxene (CPX)  $^3\text{He}/^4\text{He}$  ratios ( $R/R_A$  notation) from the same sample. The 1:1 equiline represents helium isotope equilibrium between the two phases (i.e.,  $^3\text{He}/^4\text{He}_{\text{CPX}} = ^3\text{He}/^4\text{He}_{\text{OL}}$ ). For the most part,  $^3\text{He}/^4\text{He}_{\text{CPX}} < ^3\text{He}/^4\text{He}_{\text{OL}}$  (in 14 out of 21 pairs) indicating preferential capture of crustal He ( $^3\text{He}/^4\text{He} \sim 0.05R_A$ ) by CPX at RVP (see text). Shaded area represents the MORB  $^3\text{He}/^4\text{He}$  range ( $8 \pm 1 R_A$ ).

noble gas results are unexpected given the presence of thick and old lithosphere of the Tanzanian craton which must represent a formidable barrier to sampling the high  $^3\text{He}/^4\text{He}$  plume component in the Kenya Dome region, as it both directs rift propagation to the periphery of the plateau (Figure 1), presumably where the plume signal is more dilute, and inhibits formation of a large igneous province as in Ethiopia. We speculate that the high  $^3\text{He}/^4\text{He}$  component is readily apparent at RVP due, at least in part, to the waning influence of the Tanzania craton as it was moved northward by the relative motion of the African plate [Moucha and Forte, 2011].

[7] The other major He component at RVP is also characteristic of asthenospheric mantle, albeit that normally associated with depleted upper mantle [Graham, 2002]. Unless this component is produced by fortuitous mixing between the high plume-like  $^3\text{He}/^4\text{He}$  component and SCLM and/or radiogenic He – and we argue below that these two low  $^3\text{He}/^4\text{He}$  components are either absent or not sampled by olivine crystals at RVP – then the bimodal distribution of  $^3\text{He}/^4\text{He}$  ratios at RVP must indicate a high degree of (He isotope) heterogeneity in the asthenospheric (i.e. sub-continental) mantle source region. We note that both asthenospheric mantle components are sampled over a range of silica compositions and OL forsterite (Fo) contents although the highest  $^3\text{He}/^4\text{He}$  ratios are found in the most under-saturated lavas (<43 wt.%  $\text{SiO}_2$ ) with high (>90%) Fo-contents (auxiliary material).

[8] Xenoliths from northern Tanzania all have  $^3\text{He}/^4\text{He}$  ratios within the range of SCLM (Figure 2) consistent with their origin within the mantle lithosphere of the EARS [Rudnick et al., 1994]. However, with the possible exception of Mafiga, none of the RVP olivines sample this He component. Consequently, we dismiss it both as a main contributor to the He inventory at RVP and as a likely endmember composition fortuitously mixing with a high  $^3\text{He}/^4\text{He}$  component to produce MORB-like  $^3\text{He}/^4\text{He}$  values. On the other hand, low  $^3\text{He}/^4\text{He}$  ratio crustal He ( $\sim 0.05 R_A$ ) is present at RVP, and can be observed in some

CPX phenocrysts. Out of the 21 OL-CPX pairs analyzed for Rungwe volcanics, a total of 14 have  $^3\text{He}/^4\text{He}$  ratios in CPX which are lower than co-genetic OL, with all others (except RNG-21 and TAZ09-16) showing  $^3\text{He}/^4\text{He}$  equilibrium within analytical uncertainty (Figure 3). This observation can be explained by pre-eruptive degassing of magma coupled with contamination by radiogenic He from surrounding wallrock [Hilton et al., 1993]. Crucially, contamination must occur just prior to eruption (presumably at shallow crustal levels) to preserve He disequilibrium conditions [Hilton et al., 1995]. The presence of crustal He in the near-surface environment is supported by observations that geothermal fluids at RVP (hot springs, gas seeps, etc.) have  $^3\text{He}/^4\text{He}$  values between 0.2 and 7.8  $R_A$  (i.e., consistently <MORB) [Pik et al., 2006] indicating that radiogenic He is pervasive and sampled by circulating meteoric fluids. We point out that the relatively low diffusivity of He in OL at magmatic temperatures [Trull and Kurz, 1993] renders OL crystals virtually immune to crustal contamination, so better able to capture and preserve the  $^3\text{He}/^4\text{He}$  signal of the underlying magma source.

[9] The finding that SCLM does not contribute to the He inventory at RVP stands in contrast to conclusions based upon major and trace element data that point to a major role for lithospheric peridotite in the petrogenesis of RVP melts [e.g., Furman, 1995]. The case is somewhat equivocal, however, as RVP and some other volcanics of the Western Rift (e.g., Kivu and Nyiragongo) have Sr and Nd isotope systematics consistent with sub-lithospheric contributions [Rogers, 2006; Chakrabarti et al., 2009]. One possibility that could account for plume-like  $^3\text{He}/^4\text{He}$  ratios in regions where SCLM is dominant is to invoke a large He concentration contrast between plume and lithospheric mantle sources [e.g., Graham et al., 2009], such that plume-derived He is the only tracer not overwhelmed by lithospheric additions, marking He as the tracer of choice in detecting plume contributions [Courtillot et al., 2003]. Another explanation is that prior metasomatic events may have enriched the mantle lithosphere with plume-derived volatiles [Graham et al., 2009]. This hypothesis is difficult to reconcile with recent work arguing that carbonatitic volcanism at Oldoinyo Lengai (northern Tanzania) requires a normal MORB-like asthenospheric mantle source, with enrichment resulting from low-pressure differentiation and liquid immiscibility as opposed to a volatile-rich metasomatic fluid [Fischer et al., 2009].

[10] Although an unequivocal tracer of plume involvement, a notable feature of the He isotope variations at RVP is the disparity between values recorded in mafic crystals (this work) and geothermal fluids [Pik et al., 2006]. This observation has not been made at other rift zones, such as Iceland, where there is close correspondence between  $^3\text{He}/^4\text{He}$  ratios in lavas and fluids [Hilton et al., 1990]. In the case of RVP, geothermal fluid  $^3\text{He}/^4\text{He}$  ratios are clearly susceptible to record additions of radiogenic He which act to mask intrinsic magmatic values. As basement lithologies at RVP are Precambrian and Archean in age [Ebinger et al., 1989b], and presumably rich in radiogenic He, such a finding is hardly surprising. Given the general antiquity of the crustal substrate in eastern Africa [cf. Yirgu et al., 2006], we suggest that this process must occur elsewhere along the EARS so that the He isotope distribution obtained using geothermal fluids is likely skewed to reflect crustal as

opposed to mantle variations. This includes the Main Ethiopian Rift where both high ( $\sim 16 R_A$ ) and low ( $\sim 2 R_A$ ) fluid  $^3\text{He}/^4\text{He}$  ratios are found [Scarsi and Craig, 1996]. In the case of the Kenya Dome, we note that the majority of He isotope data obtained to date use geothermal fluids [Darling et al., 1995; Pik et al., 2006; Tedesco et al., 2010], and that a wide range of  $^3\text{He}/^4\text{He}$  values (between MORB-like values to predominantly crustal ratios) characterize individual segments of the Western and Kenyan rifts (Figure 1). In contrast, mineral  $^3\text{He}/^4\text{He}$  ratios (phenocrysts and xenoliths) fall between 8.9 and 5.6  $R_A$  [Pik et al., 2006; Hopp et al., 2007; Tedesco et al., 2010] but data are limited ( $n = 27$ ). We suggest that other rift segments within the Kenya Dome could also have  $^3\text{He}/^4\text{He} > \text{MORB}$  but that they are unlikely to be identified without extensive sampling and/or specific targeting of primitive alkali volcanics. To date, none of the Kenya Dome rift segments (with the exception of RVP) have been subject to such a sampling strategy.

[11] The absence of a high  $^3\text{He}/^4\text{He}$  component along the Kenyan (Eastern) and Western rifts has hitherto led to suggestions of two distinct scales of mantle convection – deep and from the lower mantle versus shallow involving only the uppermost mantle – supporting the Ethiopia and Kenya domes, respectively [Pik et al., 2006; Montagner et al., 2007]. Our finding of a plume-like  $^3\text{He}/^4\text{He}$  contribution at the southernmost extremity of recent volcanism along the Western Rift removes this constraint, and we suggest that a lower mantle component is pervasive throughout the EARS – from RVP in the south to the Main Ethiopian Rift and Afar in the north – albeit masked at many localities by a veil of crustal He and/or sampling of MORB-like or SCLM He. We view the African Superplume [Nyblade and Robinson, 1994] – a huge low seismic velocity anomaly originating near the core-mantle boundary beneath southern Africa and tilted to north and north-east [Ritsema et al., 1999; Zhao, 2001; Montelli et al., 2006] – as the ultimate source, supplying heat, material and high  $^3\text{He}/^4\text{He}$  ratios to EARS magmatism. Recent seismic wave imaging shows a large-scale ( $>500$  km) connection between the lower mantle Superplume and the upper mantle beneath eastern Africa [Bastow et al., 2008], with ponding of plume material at the base of the transition zone (e.g., at 600–700 km depth beneath Kenya and Tanzania) driving localized thermal upwellings [Huerta et al., 2009]. Although  $^3\text{He}/^4\text{He}$  ratios at RVP are similar to values obtained in Ethiopia, this observation does not necessarily imply that a single plume is responsible for EARS magmatism [cf. Ebinger and Sleep, 1998] given the possibility that a broad thermal anomaly at the base of the transition zone may give rise to multiple thermal/mass upwellings [Huerta et al., 2009]. However, in the case of the Kenya Dome, we emphasize that the presence of high  $^3\text{He}/^4\text{He}$  ratios of RVP is consistent with the presence of a buoyant sub-lithospheric plume-related mantle source which provides geodynamic support for the topographic high. As with the Ethiopia Dome, the key geochemical evidence is provided by He isotope variations.

## 5. Conclusions

[12] Rungwe Volcanic Province, located at the southernmost extremity of the Kenya Dome, has a bimodal distribution of He isotope ratios – involving MORB-like values ( $8 \pm 1 R_A$ ) and plume-like ratios ( $>9 R_A$ ). In contrast to

mantle xenoliths from northern Tanzania, there is no evidence that the SCLM contributes to the He inventory implying that sub-lithospheric sources overwhelm any input from the mantle lithosphere at RVP. These results demonstrate the extreme sensitivity of helium isotopes as an unequivocal tracer of mantle plume and/or asthenospheric (upper) mantle involvement, even at locations such as RVP where major and trace element variations reveal a dominant role for SCLM involvement in petrogenesis. The high (plume-like)  $^3\text{He}/^4\text{He}$  ratios at RVP are consistent with a lower mantle origin, and we suggest that the African Superplume acts to transfer this high  $^3\text{He}/^4\text{He}$  signal to the surface via rift-related volcanism. If high  $^3\text{He}/^4\text{He}$  ratios found along the Main Ethiopian Rift and Afar indicate support for the Ethiopia Dome from dynamic upwelling (plume) mantle, then results presented here suggest a similar mechanism operates for the Kenya Dome.

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