

Heat flow as a catalyst for radiogenic helium release in the East Africa Rift System

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Abstract The Rukwa Rift section of the East Africa Rift System presents a type setting for radiogenic helium accumulation in a petroleum free basin. As a prerequisite for accumulation, a considerable high heat flow anomaly is required from tectonothermal events to drive the release and circulation of radiogenic helium in the continental crust. Here we apply statistical analysis on geochemical data observed in thermal springs and recorded heat flow to account for crustal helium mass balance for each tectonothermal event in the region. Our results demonstrate anomalously high heat flow $\sim 64\text{--}99$ mW/m² with a consistent trend of helium isotopic ratio and fluid chemistry in the Rukwa Rift. Mass balance calculation show that the whole crustal volume underlying the East Africa Helium Pool (EAHP) has a capability of producing radiogenic helium of about 9.9×10^6 mol/yr (22×10^{-6} mol ⁴He/m² yr) while the total radiogenic helium flux ranges between $\sim 2.39 \times 10^6$ mol/yr and $\sim 2.68 \times 10^9$ mol/yr. The Tanzania Craton contributes largely to radiogenic helium releasing up to 50% of the total capacity in the region. The total ⁴He emission in the Rukwa Rift Basin is about $4.45 \times 10^5\text{--}5.01 \times 10^8$ mol/yr which is thus equivalent to 19%–21% of the total production capacity in the region. These results imply that the

helium accumulation in the EAHP would have started as early as Paleoproterozoic (2.349 Ga). These results provide a qualitative and quantitative insight to assess both helium and geothermal potentiality in the region.

Keywords Radiogenic helium · Rukwa Rift · Heat flow · Helium potential · Helium concentration

1 Introduction

Global supplies of helium have to date been produced as a by-product from natural gas fields. Almost all natural gas accumulations contain traces of helium albeit with different origins that is, organic (natural gas) versus radiogenic/primordial sources (helium) (Ballentine and Lollar 2002; Danabalan 2017; Gluyas 2019a, b; Danabalan et al. 2022). The current global helium reserves occur as a serendipitous discovery from giant natural gas provinces such as US Midwest, Canada, Poland, Qatar, Algeria and Australia (Gluyas 2019a, b). However, the Rukwa Rift in the Tanzanian section of the East African Rift System (EARS) presents a rare setting of a petroleum-free prospect rich in radiogenic helium (Gluyas 2019a, b; Danabalan et al. 2022). The radiogenic helium flux in the Rukwa Rift segments is associated with thermal springs along with nitrogen, carbon dioxide and other noble gases (Barry et al. 2013; Danabalan 2017; Danabalan et al. 2022).

Previous work on the isotopic ratios and gas chemistry from the RRB provide important clues that suggest a binary mixing and degassing model from both mantle and crustal origins (e.g., Barry et al. 2013; Danabalan 2017; Mtili et al. 2021; Kimani et al. 2021; Danabalan et al. 2022). However, like many other active extensional tectonic regions, the RRB shows a wide range of flux rate and chemical variability

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associated with potentially high thermal anomalies (Lowenstern et al. 2014; Danabalan et al. 2022; Didas et al. 2022). Although the release of crustal helium depends largely on thermal conditions (e.g., Danabalan et al. 2022), the distribution of the heat flow anomaly in relation to the helium potential in the RRB remains poorly understood.

In this contribution the mass balance between ^4He efflux and production potential of each particular tectonic setting/terrane are quantified in the region which we name the East Africa Helium Pool (EAHP) (Fig. 1A, B). We account for ^4He production capacity of crustal volumes and flux rate through concentrations observed in thermal springs from previous studies in the RRB (e.g., Barry et al. 2013; Lee et al. 2016; Mtili et al. 2021). The calculations assume that each tectonothermal event recorded in the RRB was associated with a heat flow surge (e.g., Torgersen 2010; Lowenstern et al. 2014). The heat flow affects the primary release of radiogenic helium from helium-retentive minerals such as apatite, zircon, and uraninite and initiate diffusive transport (Danabalan et al. 2022). This study provides insights into the timescale of helium production, release and a possible timing of accumulation surge triggered by heat flow anomaly.

2 Geological background

2.1 Geology and tectonic settings

The tectonics setting and terranes that underlie the EAHP comprises the Tanzania Craton, the Bangweulu Craton and the Ubendian mobile belt (Fig. 1A, B). These terranes

surround the RRB and influenced its development (Boniface 2009; Boniface and Schenk 2012; Fig. 1A, B). The Tanzania and Bangweulu cratons comprises mainly granitoids and metamorphic rocks such as granitic gneisses and migmatites (Andersen and Unrug 1984; Many et al. 2007).

The Ubendian mobile belt consists of uplifted Palaeoproterozoic rocks composed of amphiboles-granulite metamorphic facies (Boniface and Schenk 2012; Fritz et al. 2013). The Ubendian mobile belt is divided into eight Palaeoproterozoic terranes which include; Kate-Kipili, Mbozi, Katuma, Ufipa, Wakole, Ubende, Upangwa and Lupa terranes (Daly 1988; Lenoir et al. 1994; Theunissen et al. 1996; Fig. 1A, B).

The EAHP behaves as a comprehensive closed system comprised of rocks which are rich in helium bearing minerals that is, uranium and thorium bearing minerals with average crustal density and thickness of ~ 2.7 g/cc and 42 km respectively (Adams et al 1959; Burwash and Cumming 1976; Chaki et al. 2008; René 2017; Lavayssière et al. 2019). The uranium concentration in both Tanzania and Bangweulu cratons is ~ 18.7 ppm, which is 6–7 times, above the average concentration ~ 2.8 ppm recorded over other Precambrian terranes (Many et al. 2007; Mshiu and Maboko 2012). The thorium concentrations are 5 times higher in the cratons (~ 52.1 ppm) than in other Precambrian terranes (~ 10.7 ppm) (Many et al. 2007; Mshiu and Maboko 2012).

2.2 Tectonothermal events

Several regional tectonothermal events have been previously mapped in the EAHP (e.g. Ebinger et al. 1989; Kilembe and Rosendahl 1992; Mulaya et al. 2022). Notably, the orogenic

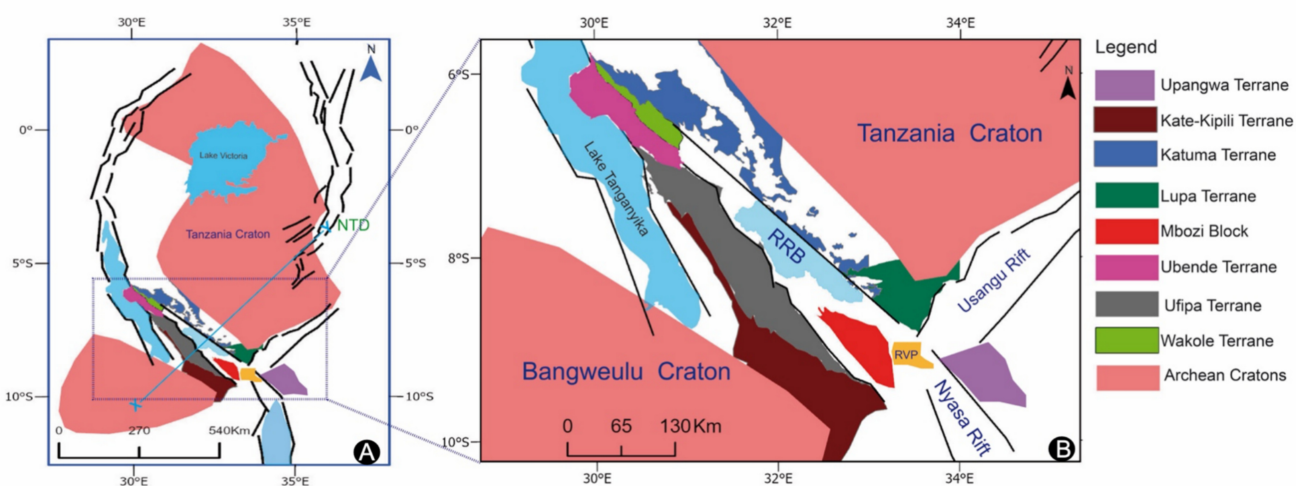


Fig. 1 (A) Map showing tectonic terranes which we refer as East Africa Helium Pool (EAHP). (B) Map showing the tectonic terranes surrounding the Rukwa Rift within the Ubendian mobile belt. RRB. Rukwa Rift Basin, RVP. Rukwa Volcanic Province, NTD. North Tanzanian Divergence

event which led to the Ubendian mobile belt dates back to 500 Ma (Tanganyika Orogeny) which involved a collision between the Tanzania Craton and the Bangweulu Craton (Quennell et al. 1956; Lenoir et al. 1994; Boniface 2009). At least three rifting episodes have been identified in the RRB and Rungwe Volcanic Province (RVP) including Karoo rifting (Late Carboniferous to Triassic), Cretaceous to Oligocene rifting and the Cenozoic rifting (Late Miocene to present) (Ebinger et al. 1989; Kilembe and Rosendahl 1992; Ebinger and Sleep 1998; Delvaux 2001; Boniface 2017; Mulaya et al. 2022). These events as in many other similar tectonic settings (e.g., Lowernstern et al. 2014) are associated with rifting, orogeny and volcanic activity (Danabalan et al. 2022). All these events were likely associated with heat flow surges that can facilitate fluid mobilization and helium degassing in the active tectonic setting of the RRB (Lowernstern et al. 2014; Danabalan et al. 2022).

3 Methodology

3.1 Defining a closed system for helium production and release

Geochemical and isotope data used in this study were compiled by James (1967); Barry et al. (2013); Mtili et al. (2021); Kimani et al. (2021) and the mass balance computation (Tables 1, 2) follows methods described by Lowernstern et al. (2014). These data were used to compute for helium flux and production models by assuming a closed system where fluids interact mainly in rocks within the tectonic terranes surrounding the RRB (e.g., Lowernstern et al. 2014). The closed system defined in this study is named as the EAHP (Figs. 1A, B) whose areas were calculated from ArcGIS Pro shapefiles (Tables 1, 2). The EAHP includes the Ubendian mobile belt ~114,000 km², the Bangweulu Craton ~150,000 km² (Andersen and Unrug 1984) and the Tanzania Craton ~350,000 km² (Danabalan et al. 2022). The EAHP consist of the Ubendian terranes which surround the Rukwa Rift segment of the East Africa Rift System (Fig. 1A, B).

The total calculated helium production rate per year for the whole EAHP is therefore referred as a Whole-Crust (WC) value against which all the helium fluxes in the system were balanced (Table 3). The mass balance computation is based on the fact that ⁴He can accumulate over time within the EAHP whereas tectonic activity can facilitate degassing and circulation in the region (Ingebritsen and Manning 2010; Lowernstern et al. 2014; Danabalan et al. 2022).

3.2 Statistical modeling of ⁴He mass balance and heat flow anomaly

Two different cases for the helium flux model were considered for the selected closed system boundary (Tables 1 and 2). The choice of parameters for the two cases is based on previously reported CO₂ flux estimates of 4.05 and 0.0055 tonnes/day in areas sized 981.5 and 1500 km² respectively (Barry et al. 2013; Lee et al. 2016). Previously, the lowest and highest end member of crustal and mantle Ra signatures have been reported as 0.02 Ra and 8.0 Ra respectively and hence these values were adopted in this study (Barry et al. 2013; Mtili et al. 2021; Danabalan et al. 2022).

Heat flow data were extracted from previous work by Morley et al. (1999), Njinju et al. (2019) and Didas et al. (2022). The few available isotopic ratios, heat flow and geochemical data from previous studies such as CO₂/³He and R/Ra (Morley et al. 1999; Barry et al. 2013; Danabalan 2017; Njinju et al. 2019; Mtili et al. 2021 and Didas et al. 2022) were used for statistical interpolation across the RRB and RVP using ArcGIS Pro.

For the statistical interpolation in ArcGIS Pro, the Inverse Distance Weighting (IDW) method (Geostatistical Analyst Tool) was applied, using the available isotope ratio (Ra), CO₂ flux and heat flow data as inputs (assuming output cell size ~0.01, power of ~2 and number of points up to 12) to produce raster images. From the raster images of Ra and CO₂/³He about 1000 random discrete points were created statistically where a number of random points depends on the resolution required to produce reliable neighbourhoods (Fig. 2). These random points surround various locations where data had been previously sampled/computed in the RRB and RVP (Fig. 2). Finally, the cell values from the raster maps were extracted and statistically assigned to those 1000 random points using the Spatial Analyst Tools ('Extract values to points') to represent unique cell values for interpolated Ra and CO₂/³He. The averages of interpolated cell values bounded by each tectonic terrane were used as inputs during mass balance computation (Tables 1, 2).

4 Results

4.1 Heat flow distribution in the Rukwa Rift

The distribution of heat flow data in the Rukwa Rift shows a consistent trend along strike of the Rukwa Rift Basin (RRB) and the Rungwe Volcanic Province (RVP) (Fig. 3A, B). In the RRB the heat flow ranges between 64 mW/m² and 86 mW/m² above the global average for Mid-Permian Provinces (e.g., Njinju et al. 2019; Fig. 3A, B). The heat flow is higher in the Rungwe Volcanic Province up to 99 mW/m² and drops sharply towards the Rukwa Rift graben (minimum 64 mW/

Table 1 Case-I: CO₂ and He flux for the various tectonic settings of the EAHF. Original flux data e.g., CO₂ were extracted from Lee et al. (2016); Barry et al. (2013); Mtuli et al. (2021); Dana-balan et al. (2022) while other parameters were interpolated using statistical tools in ArcGIS Pro software

Thermal area	Area (km ²)	QCO ₂ (grams/day)	CO ₂ (mol/yr)	CO ₂ ³ He	CO ₂ ⁴ He	Q ⁴ He (mol/yr)	R/Ra Mantle ^{e2}	R/Ra Local	³ He/ ⁴ He	f _{crustal}	q ^C He (mol/yr)	WC	CO ₂ Flux(Mt/yr)	Area of flux
Rukwa Basin	8890	1.01E+11	8.34E+11	5.8E+09	1.6E+03	5.1E+08	8.00E+00	0.2	2.8E-07	9.8E-01	5.01E+08	51	4.05	981.5
RVP	1500	1.70E+10	1.41E+11	8.7E+09	4.0E+04	3.5E+06	8.00E+00	3.3	4.6E-06	5.9E-01	2.07E+06	0		
Ubende Terrane	5275	5.96E+10	4.95E+11	1.0E+10	1.7E+04	2.9E+07	8.00E+00	1.2	1.7E-06	8.5E-01	2.51E+07	3		
Wakole Terrane	2045	2.31E+10	1.92E+11	9.9E+09	1.7E+04	1.2E+07	8.00E+00	1.2	1.7E-06	8.5E-01	9.81E+06	1		
Ufipa Terrane	13,978	1.58E+11	1.31E+12	9.2E+09	1.4E+04	9.2E+07	8.00E+00	1.1	1.5E-06	8.6E-01	7.96E+07	8		
Lupa Terrane	6560	7.42E+10	6.15E+11	7.9E+09	2.1E+04	2.9E+07	8.00E+00	1.9	2.7E-06	7.6E-01	2.22E+07	2		
Nyika Terrane	10,310	1.17E+11	9.67E+11	6.4E+09	3.0E+04	3.2E+07	8.00E+00	3.4	4.8E-06	5.8E-01	1.83E+07	2		
Bangweulu Craton	150,000	1.70E+12	1.41E+13	1.1E+10	1.9E+04	7.2E+08	8.00E+00	1.3	1.8E-06	8.4E-01	6.08E+08	62		
Tanzania Craton	350,000	3.96E+12	3.28E+13	1.0E+10	2.2E+04	1.5E+09	8.00E+00	1.5	2.1E-06	8.1E-01	1.22E+09	124		
Total	614,293.6										2.68E+09	272.06		

Table 2 Case-II: CO₂ and He flux for the various tectonic settings of the EAFP. Original flux data e.g., CO₂ were extracted from Lee et al. (2016); Barry et al. (2013); Mtilli et al. (2021); Dana-balan et al. (2022) while other parameters were interpolated using statistical tools in ArcGIS Pro software

Thermal area	Area (km ²)	QCO ₂ (grams/day)	CO ₂ (mol/yr)	CO ₂ ^β He	CO ₂ ^α He	Q ⁴ He (mol/yr)	R/Ra Mantle ²	R/Ra Local	³ He/ ⁴ He	f _{crustal}	q ⁶ He (mol/yr)	WC	CO ₂ Flux (Mt/yr)	Area of flux
Rukwa Basin	8890	8.93E+07	7.41E+08	5,810,147,238	1.63E+03	4.55E+05	8.00E+00	0.2	0.00000028	9.77E-01	4.45E+05	0.05	0.00055	1500
RVP	1500	1.51E+07	1.25E+08	8,669,586,054	4.01E+04	3.12E+03	8.00E+00	3.3	0.00000462	5.89E-01	1.84E+03	0.00		
Ubende Terrane	5275	5.30E+07	4.39E+08	9,983,714,962	1.68E+04	2.62E+04	8.00E+00	1.2	0.00000168	8.52E-01	2.23E+04	0.00		
Wakole Terrane	2045	2.05E+07	1.70E+08	9,913,632,768	1.67E+04	1.02E+04	8.00E+00	1.2	0.00000168	8.52E-01	8.72E+03	0.00		
Ufipa Terrane	13,978	1.40E+08	1.16E+09	9,247,200,474	1.42E+04	8.18E+04	8.00E+00	1.1	0.00000154	8.65E-01	7.07E+04	0.01		
Lupa Terrane	6560	6.59E+07	5.47E+08	7,947,573,838	2.11E+04	2.59E+04	8.00E+00	1.9	0.00000266	7.64E-01	1.98E+04	0.00		
Katuma Terrane	3883	3.90E+07	3.24E+08	9,577,752,101	1.61E+04	2.01E+04	8.00E+00	1.2	0.00000168	8.52E-01	1.71E+04	0.00		
Upangwa Terrane	11,081	1.11E+08	9.23E+08	2,310,542,513	1.20E+04	7.71E+04	8.00E+00	3.7	0.00000518	5.39E-01	4.16E+04	0.00		
Mbozi Terranes	7887.5	7.92E+07	6.57E+08	21,669,463,904	7.89E+04	8.33E+03	8.00E+00	2.6	0.00000364	6.77E-01	5.64E+03	0.00		
Kate-Kipili Terrane	42,884.1	4.31E+08	3.57E+09	11,611,227,264	2.60E+04	1.37E+05	8.00E+00	1.6	0.00000224	8.02E-01	1.10E+05	0.01		
Nyika Terrane	10,310	1.04E+08	8.59E+08	6,393,470,310	3.04E+04	2.82E+04	8.00E+00	3.4	0.00000476	5.76E-01	1.63E+04	0.00		
Bangweulu Craton	150,000	1.51E+09	1.25E+10	10,672,146,767	1.94E+04	6.43E+05	8.00E+00	1.3	0.00000182	8.40E-01	5.40E+05	0.05		
Tanzania Craton	350,000	3.52E+09	2.92E+10	10,415,137,036	2.19E+04	1.33E+06	8.00E+00	1.5	0.00000021	8.15E-01	1.09E+06	0.11		
Total	114,293.6										2.39E+06	0.24		

Table 3 Radiogenic helium production models. Concentration data sourced from Manyá et al. (2007); Chaki et al. (2008); Mshiu and Maboko. (2012); Lavayssière et al. (2019); Danabalan et al. (2022)

Location	U Conc. (ppm)	Th Conc. (ppm)	$J^4\text{He}$ Prod (atoms/g/yr)	Crustal ρ g/cc	Total Area (cm^2)	Thickness (cm)	Mass (g)	Mass/NA (gmol)	$Q^C4\text{He}$ (molyr^{-1})
Cratons	18.7	52.1	$1.0\text{E}+08$	2.7	$5.0\text{E}+15$	$4.2\text{E}+06$	$5.67\text{E}+22$	$9.4\text{E}-02$	$9.5\text{E}+06$
Other terranes	2.8	10.7	$1.7\text{E}+07$	2.7	$1.1\text{E}+15$	$4.2\text{E}+06$	$1.29609\text{E}+22$	$2.2\text{E}-02$	$3.7\text{E}+05$

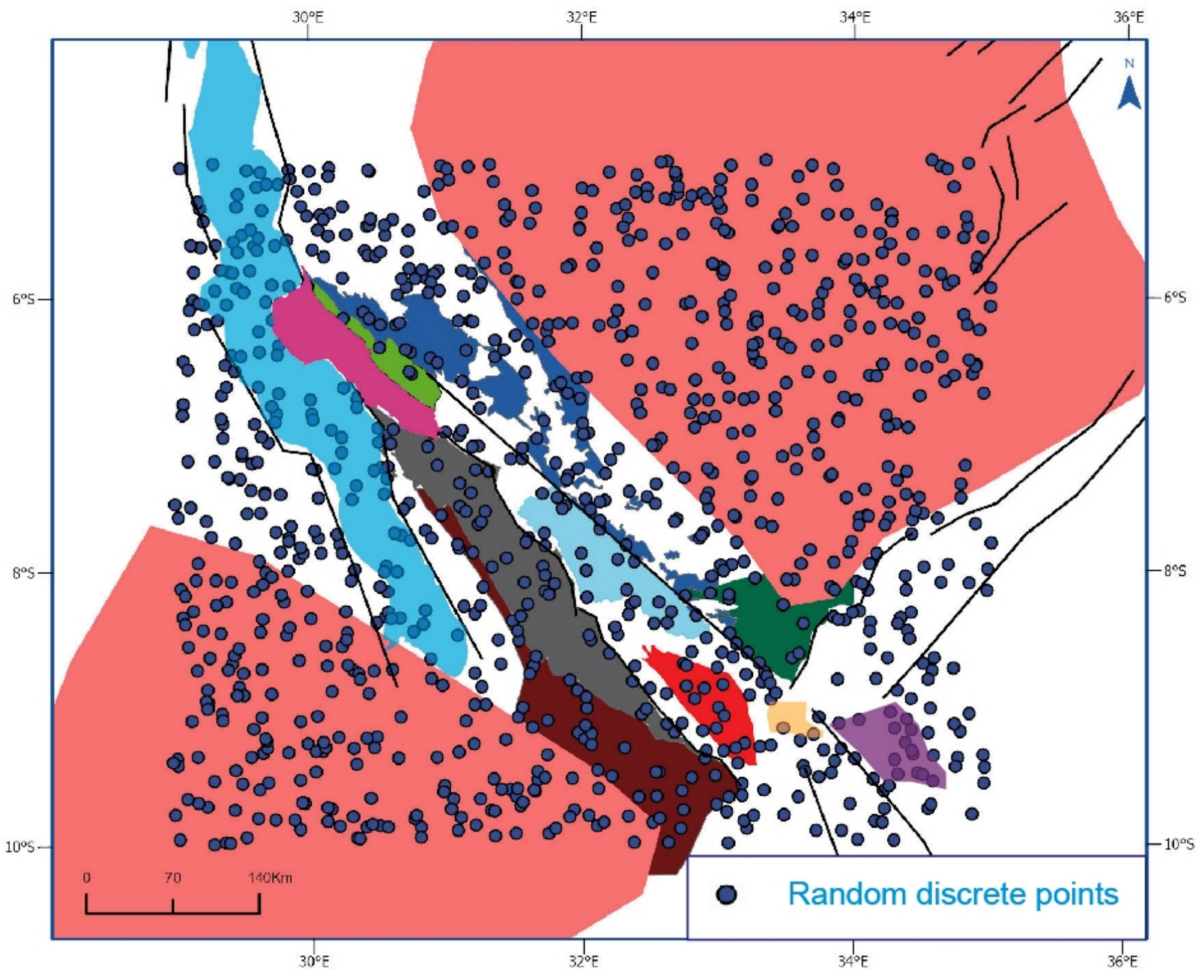
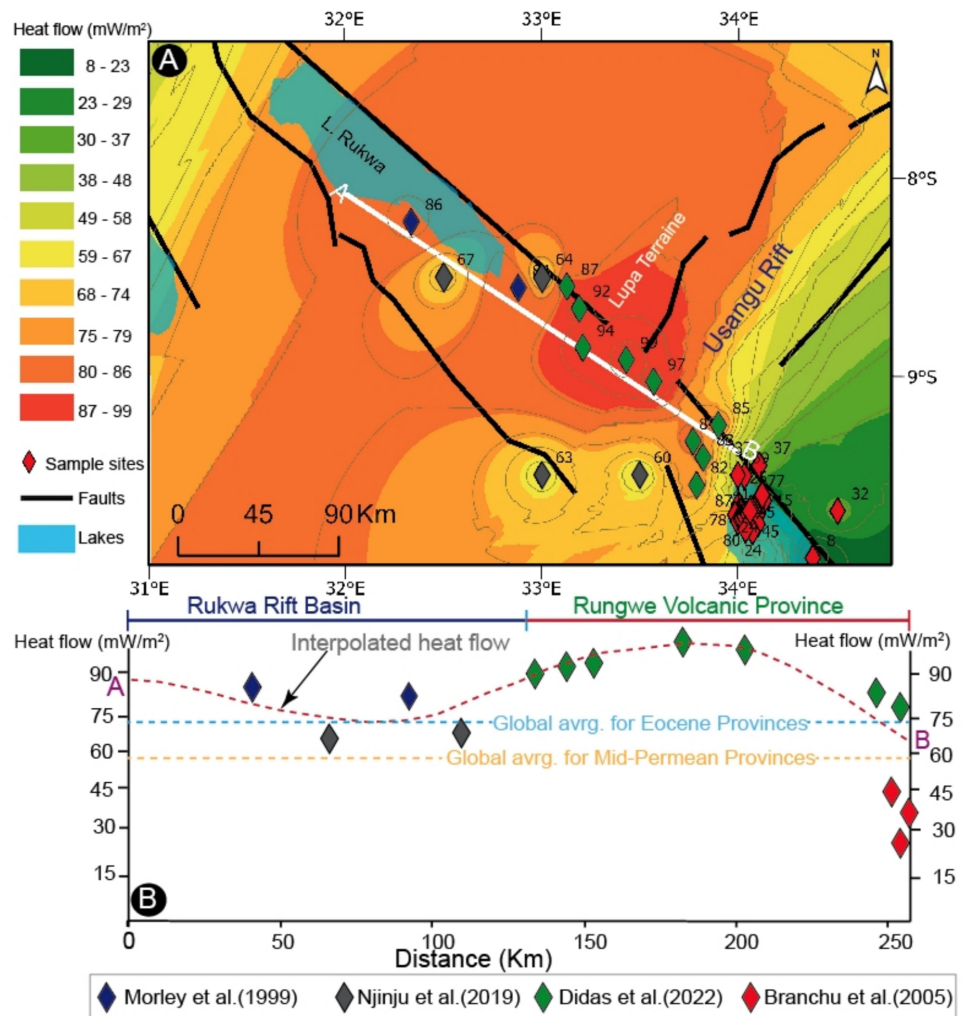


Fig. 2 Random discrete points ($n=1000$) created on which heat flow, $\text{CO}_2/{}^3\text{He}$ and ${}^3\text{He}/{}^4\text{He}$ data were interpolated over various tectonic terranes and averaged for use in mass computation and raster mapping (see Fig. 1 for legend)

m^2) (Fig. 3A, B). Our model reveals that the heat flow anomaly coincides with changes in topography with higher heat flow up to 99 mW/m^2 observed over the RVP highlands compared with the RRB graben with heat flow as low as $\sim 64 \text{ mW/m}^2$ (Fig. 3A, B). Most of the high heat flow values are aligned with faults while others occur in accommodation zones (areas linking rift segments) and volcanically active area such as the RVP (Fig. 3A). The heat flow decreases significantly towards the distal areas from the RVP where the lowest heat flow up

to $\sim 32 \text{ mW/m}^2$ occurs in the Malawi Rift (Fig. 3A). Notably, further to the northeast of the Rukwa Rift, higher heat flow $\sim 94 \text{ mW/m}^2$ is observed over the Lupa Terrane than any other tectonic terrane within the Ubendian mobile belt (Fig. 3A). The high heat flow over the Lupa Terrane is observed near the rift shoulder and horst blocks of both the Rukwa and Usungu rifts and decreases towards the distal areas (Fig. 3A).

Fig. 3 (A) Map showing the distribution of heat flow between RRB and RVP generated from the statistical interpolation at various locations. (B) A profile showing distribution of heat flow values between the RRB and the RVP compared to the global mean heat flow for Mid-Permian and Eocene provinces (after Polyak and Smirnov 1968; Chapman and Pollack 1975; Njinju et al. 2019)



4.2 Relationship between heat flow and helium isotope ratio

The highest heat flow in the RVP (up to 99 mW/m²) corresponds to the highest ³He/⁴He ratio (3.3–7.10 Ra) (Fig. 4A, B). In the RRB the lowest heat flow (64 mW/m²) is associated with the lowest ³He/⁴He ratio (0.18–0.68 Ra) (Fig. 4A, B). The changes in a trend of helium isotopic ratio coincide with the changes in topographic profile (Fig. 4A–D).

4.3 Relationship between heat flow and CO₂/³He ratio

The general trend of heat flow in relation to CO₂/³He shows the highest heat flow (~99 mW/m²) occurring in the RVP characterized by the highest CO₂/³He (1.4×10¹³–2.9×10¹³) (Fig. 5A, B). The RRB reveals the

lowest heat flow (~64 mW/m²) corresponding to the lowest CO₂/³He (0.5×10¹⁰–1.0×10¹¹) (Fig. 5A, B). Similar to helium isotopic ratio, the change in CO₂/³He along strike of the RRB and RVP show corresponding changes with topography (Fig. 5A–D; Fig. 6A–C).

4.4 Heat flow anomaly and frequent seismicity in the region

Our observation shows that seismicity swarms occur in areas of high heat flow (Fig. 7). Notably at the junction of border faults between the Rukwa Rift and Usangu Rift, frequent seismicity most likely revealing active tectonic activity and fault reactivation associated with thermal anomalies (Fig. 7).

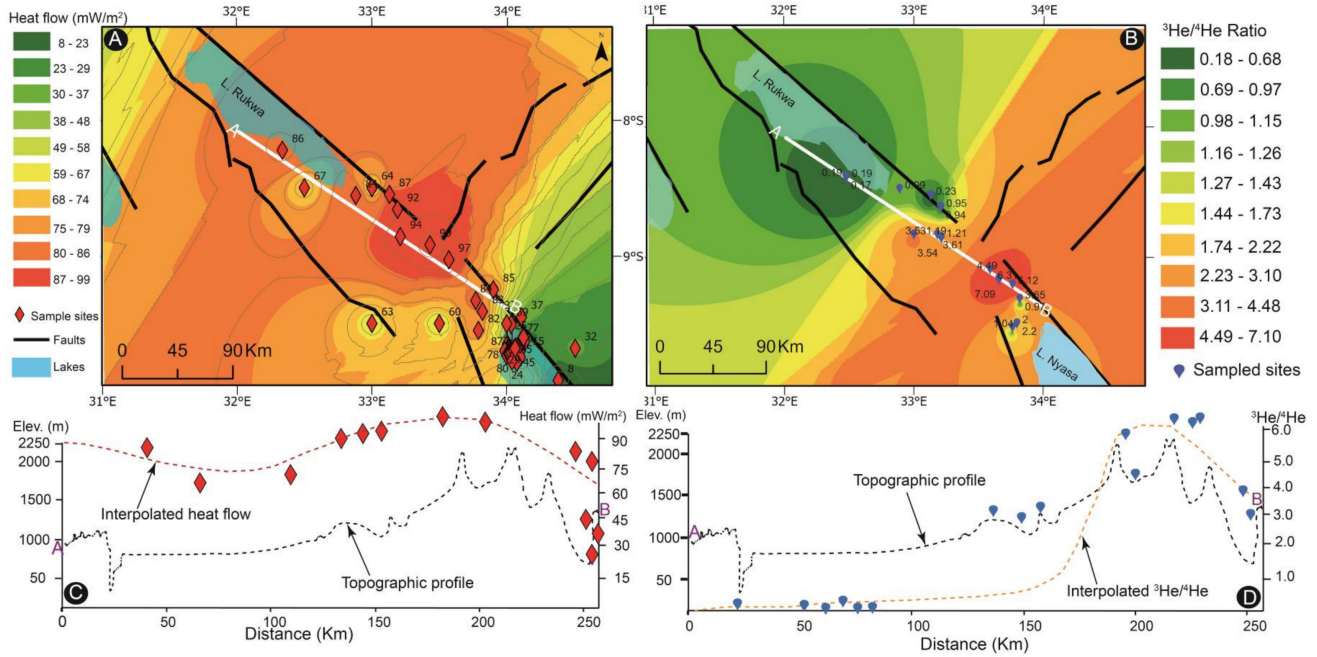


Fig. 4 (A) Interpolated heat flow map for the RVP and RRB. The diamond symbols show heat flow values that have been either tested or computed. (B) Raster map showing the distribution of ³He/⁴He ratios. Areas with available ³He/⁴He data are shown with blue drop symbols. (C) A profile across the southern part of the RRB and RVP showing distribution of interpolated heat flow against the topographic profile. (D) Section across the southern part of the RRB and RVP showing distribution of interpolated ³He/⁴He ratio against the topographic profile

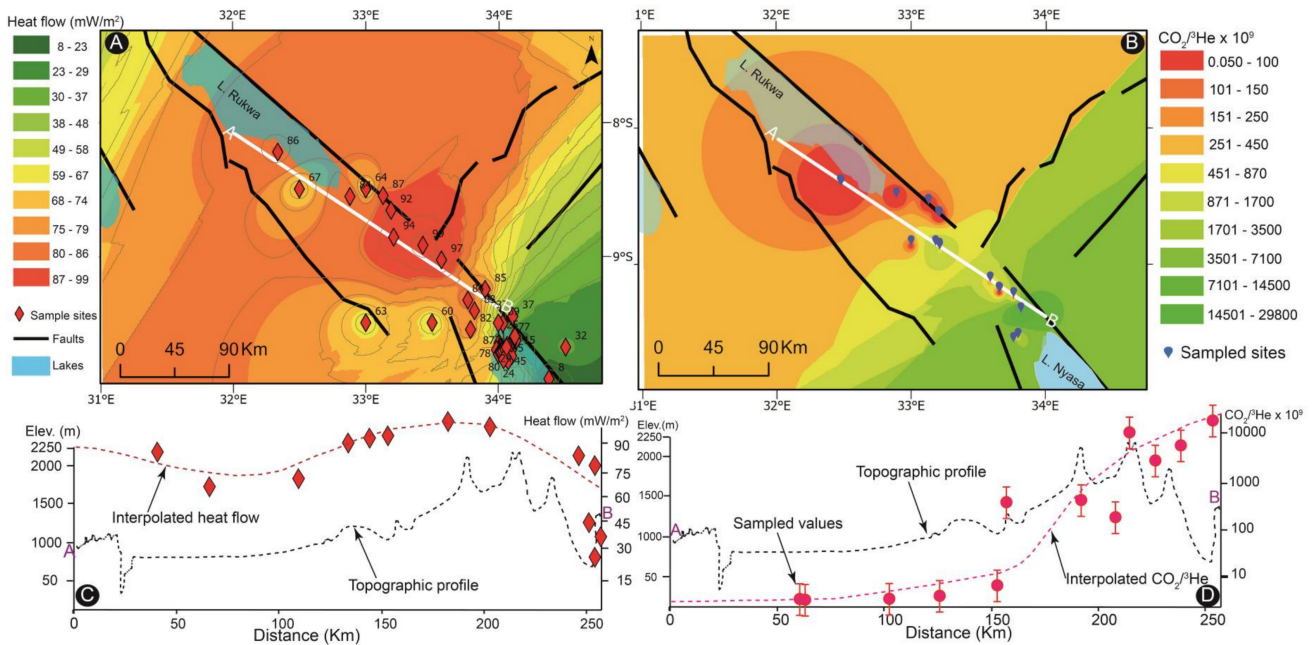


Fig. 5 A Interpolated heat flow map for the RVP and RRB. The diamond symbols show heat flow values where has been either tested or computed. B Raster map created from interpolation of available data to show the distribution of CO₂/³He ratios. Locations with available CO₂/³He data are shown with blue drop symbols. C A profile across the southern part of the RRB and RVP showing distribution of interpolated heat flow against the topographic profile. D A profile across the southern part of the RRB and RVP showing distribution of interpolated CO₂/³He ratio against the topographic profile

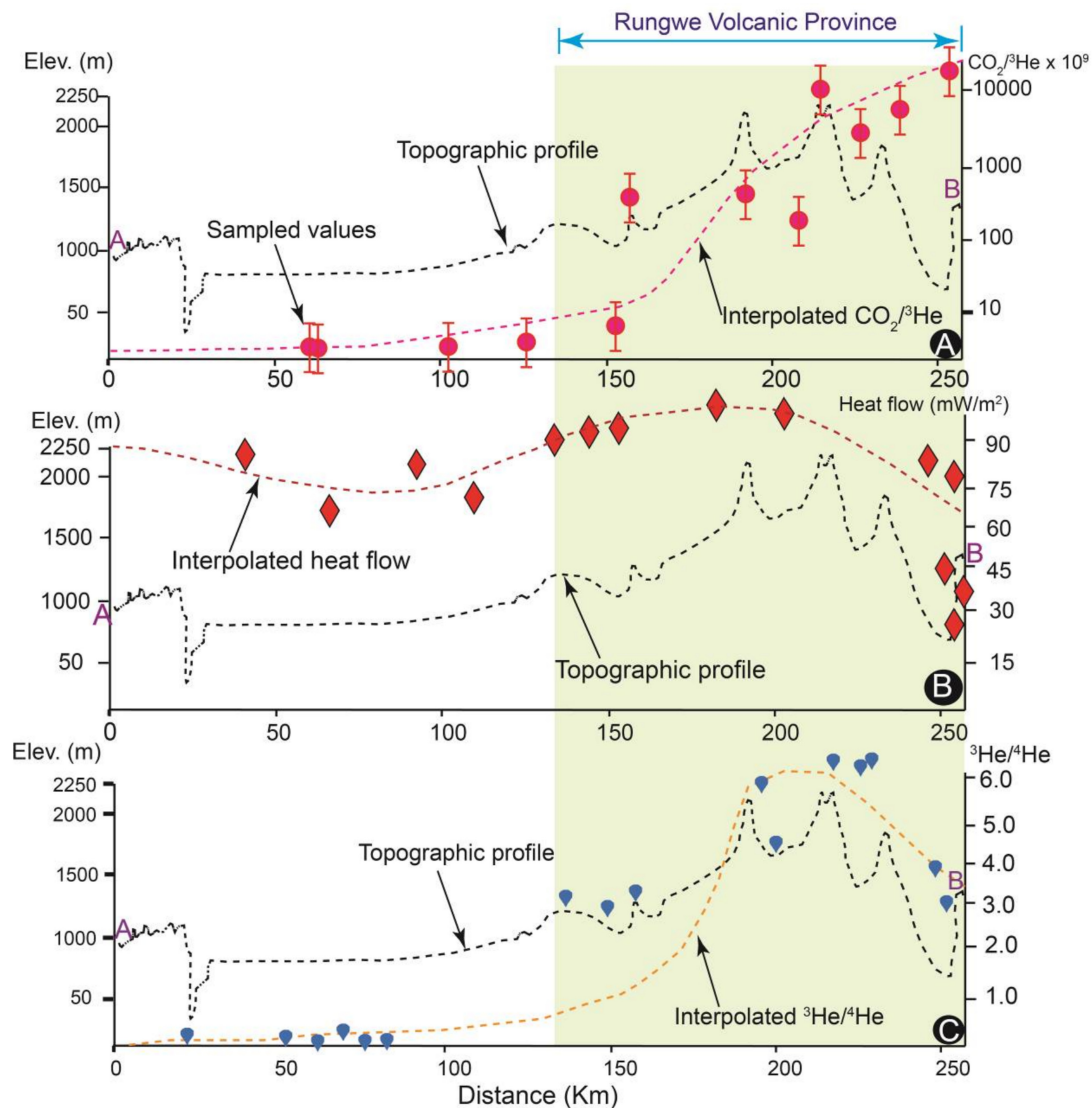


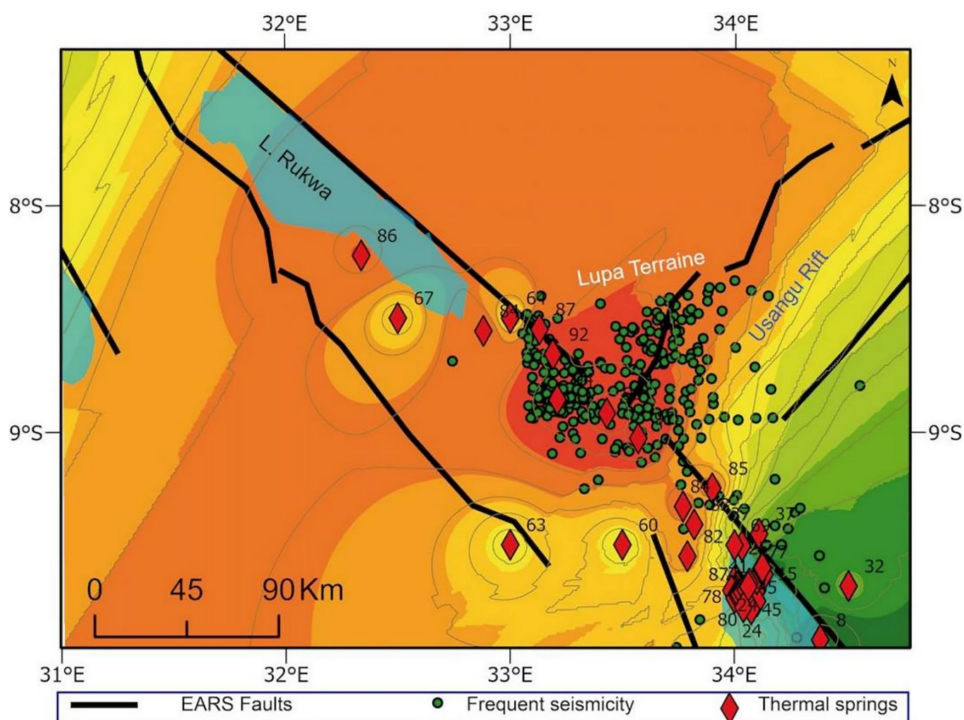
Fig. 6 Profile across the RVP and the southern part of the RRB showing distribution of **A** $\text{CO}_2/{}^3\text{He}$ ratio against the topographic profile. **B** heat flow against the topographic profile. **C** ${}^3\text{He}/{}^4\text{He}$ ratio against the topographic profile. (See Fig. 3A for location)

4.5 Helium potential of the East Africa Helium Pool (EAHP)

Helium isotopic ratios (${}^3\text{He}/{}^4\text{He}$), expressed as Ra were interpolated as essential pre-computational values for use in mass balance calculation. The cratons with ~ 3 to 7 times the average crustal concentration of radioactive elements, have Ra distribution ranging between 1.3 Ra and 1.5 Ra (Tables 1, 2). The RRB presents the lowest Ra ~ 0.2 Ra in the system whereas, the Upangwa Terrane, Nyika Terrane and the RVP have the highest Ra ~ 3.3 – 3.7 Ra. The distribution of Ra in other terranes includes Ufipa ~ 1.1 Ra, Ubende ~ 1.2 Ra, Katuma ~ 1.2 Ra, Wakole ~ 1.2 Ra, Mbozi ~ 2.6 Ra, Lupa ~ 1.9 Ra and Kate Kipili ~ 1.6 Ra (Tables 1–2).

Mass balance computation for degassing and production of radiogenic helium show that the whole EAHP has a crustal mass of 7.0×10^{10} megatonnes which includes the Tanzania Craton, Bangweulu Craton and the Ubendian mobile belt (Table 3). The tectonic terranes and blocks surrounding the RRB are capable of producing at least 3.3×10^{-6} mol ${}^4\text{He}/\text{m}^2$ yr (Table 3). In addition, both the Bangweulu and the Tanzania cratons can potentially produce 6 times more than other terranes in the Ubendian mobile belt ($\sim 1.9 \times 10^{-5}$ mol ${}^4\text{He}/\text{m}^2$ yr) making a total helium production rate of at least 2.2×10^{-5} mol ${}^4\text{He}/\text{m}^2$ yr in an area of $\sim 614,000$ km² for the Whole-Crust (1WC) under the assumption of a closed system EAHP (Table 3).

Fig. 7 Heat flow map overlain by the distribution of frequent seismicity recorded in the region. Note seismicity clusters occur at the junction between the Usangu Rift and Rukwa Rift



4.5.1 Helium degassing: case-I

Under this postulate we assume CO_2 flux of 4.05 tonnes/day over an areal extent $\sim 981.5 \text{ km}^2$ (Lee et al. 2016; Table 1). Feasible models under this postulate suggest a total crustal ^4He emission of $\sim 272\text{WC}$ from the EAHP (Table 1). The contribution of crustal ^4He emissions for each tectonic terrane include 124WC (46%) from the Tanzania Craton, 62WC (22%) from the Bangweulu Craton and 52WC (19%) from the crystalline basement of the RRB. The tectonic terranes in the whole Ubendian mobile belt constitute 36WC (13%) of the helium contribution in the region (EAHP) (Table 1).

Case-I shows total radiogenic helium flux $\sim 2.68 \times 10^9 \text{ mol/yr}$ in the EAHP with areal extent $\sim 614,000 \text{ km}^2$ ($\sim 4.4 \times 10^{-3} \text{ mol } ^4\text{He/m}^2 \text{ yr}$) (Table 1). Notably, in the RRB the calculated ^4He emissions reveal $\sim 5.01 \times 10^8 \text{ mol/yr}$ in an area of $\sim 8890 \text{ km}^2$ ($\sim 5.6 \times 10^{-2} \text{ mol } ^4\text{He/m}^2 \text{ yr}$) (Table 1). The ^4He emission in the RRB is thus equivalent to 19% of the total WC whereas in the RVP $\sim 1500 \text{ km}^2$, the ^4He emissions are lower $\sim 2.07 \times 10^6 \text{ mol/yr}$ ($< 1\%$) than in the RRB (Table 1).

4.5.2 Helium degassing: case-II

This postulate assumes a CO_2 flux of 0.0055 tonnes/day over an areal extent $\sim 1500 \text{ km}^2$ (Barry et al. 2013) which results in a crustal helium flux $\sim 0.24\text{WC}$ (Table 2). Similar to Case-1 above, the Tanzania Craton contributes 46% of

the crustal ^4He emission, Bangweulu Craton (21%), RVP ($< 1\%$), the underlying crystalline basement of the RRB (21%) and the Ubendian mobile belt (12%).

Total radiogenic helium flux in the EAHP under Case II results in up to $\sim 2.39 \times 10^6 \text{ mol/yr}$ ($\sim 3.9 \times 10^{-6} \text{ mol } ^4\text{He/m}^2 \text{ yr}$) (Table 2). Similar to Case-1, the calculated ^4He emissions under this case reveal $\sim 4.45 \times 10^5 \text{ mol/yr}$ from an area of $\sim 8890 \text{ km}^2$ of the RRB ($\sim 5.0 \times 10^{-5} \text{ mol } ^4\text{He/m}^2 \text{ yr}$). The radiogenic helium degassing in the RRB is thus equivalent to 21% of the WC. In the RVP, the helium emission is lower than in the RRB that is, $\sim 1.84 \times 10^3 \text{ mol/yr}$ ($\sim 1.23 \times 10^{-6} \text{ mol } ^4\text{He/m}^2 \text{ yr}$) equivalent to $< 1\%$ (Table 2).

4.6 Radiogenic residence time in the Rukwa rift

Residence time are calculated using geochemical data which include resolved radiogenic isotopes ^4He and ^{40}Ar from uranium and thorium that were quantitatively recovered from fluids by Danabalan et al. (2022), Mtili et al. (2021) and Kimani et al. (2021). The calculations assume that these daughter isotopes were produced in situ and accumulated within the hydrological closed system of the Rukwa Rift which is comprised of the Rukwa Rift Basin and Rungwe Volcanic Province. Therefore, the residence time refers to the period since isolation of a daughter isotope and interaction with a hydrological system within either crystalline basement or overlying sedimentary units.

The radiogenic noble gas concentration of noble gas is calculated per volume of water using Gas–Water ratios (Vg/

Vw). The assumptions made for reference Air Saturated Water (ASW), initial pore fluid composition and salinity are adopted from Mtili et al. (2021). The ASW also assumes that the groundwater has undergone little or no changes that is, phase separation since last equilibration with the atmosphere (Holland et al. 2013; Warr et al. 2018). We further assume that all radiogenic noble gases were produced in situ either within sedimentary settings or from the deeper crust of the Rukwa Rift and accumulated in the stratigraphic sequences controlled dominantly by vertical diffusive migration (cf. Holland et al. 2013; Cheng et al. 2021). For instance, at Ivuna mudpots locality the average V_g/V_w of 0.0075 shown in Eq. (1);

$$\frac{V_g}{V_w}(36_{Ar}) = 0.0075 \quad (1)$$

This can be rewritten in Eq. (2) as;

$$\frac{V_g}{V_w}(36_{Ar}) = \frac{1}{133.3} \quad (2)$$

This means that at the Ivuna mudpots 1 cm^3 at Standard Temperature and Pressure (STP) of ^{36}Ar can be stripped out by 133.3 cm^3 of water in the subsurface. Since $^4\text{He}/^{20}\text{Ne}$ ratio in the Rukwa Rift ranges from 1–15,000 (Mtili et al. 2021), therefore the atmospheric contribution in the hydrologic closed system of the Rukwa Rift is either little or negligible.

Given the average concentrations of the $^{40}\text{Ar}/^{36}\text{Ar}$ ratio = 726 and $^{40}\text{Ar} = 16,175 \times 10^{-6} \text{ cm}^3/\text{cm}^3$ at Ivuna mudpots (Table 6).

The concentration of ^{36}Ar in equivalent volume of water is the concentration of $^{40}\text{Ar} \div ^{40}\text{Ar}/^{36}\text{Ar}$ ratio, thus, $16,175 \times 10^{-6} \text{ cm}^3/\text{cm}^3 \div 726 = 2.2 \times 10^{-5} \text{ cm}^3/\text{cm}^3$ ^{36}Ar .

4.6.1 Noble gases accumulation in the hydrologic system of the Rukwa Rift: Case study of Ivuna mudpots

Since 1 cm^3 STP of ^{36}Ar can be stripped out by 133.3 cm^3 of water in the subsurface (Eq. 2), therefore groundwater ^{36}Ar concentration of $2.2 \times 10^{-5} \text{ cm}^3/\text{cm}^3$ will require $2.97 \times 10^{-3} \text{ cm}^3$ of subsurface water that is, the total volume of water that the noble gases have interacted with beneath the Ivuna mudpots. This amount of water can therefore be used to compute the concentration of other radiogenic noble gases per equivalent volume of water sampled from the same locality.

Using the equivalent volume of water as $2.97 \times 10^{-3} \text{ cm}^3$, the concentration of $^{40}\text{Ar} = 16,175 \times 10^{-6} \text{ cm}^3/\text{cm}^3$ per volume of water will be $5.45 \text{ cm}^3/\text{cm}^3 \text{ H}_2\text{O}$. Similar algorithms can be applied for the resolved ^4He concentration.

In order to calculate the noble gas concentration, the reasonable estimate of the rock matrix density and measured porosity were considered based on assumptions made also by Warr et al. 2018 (Eq. 3);

$$\begin{aligned} \text{Conc. } 40_{Ar} & \left[\frac{\text{cm}^3}{\text{gram}} \text{ of host rock} \right] \\ & = 40_{Ar} \left[\frac{\text{cm}^3}{\text{cm}^3} \text{ of water} \right] \times \text{Porosity} \div \text{Density} \end{aligned} \quad (3)$$

where density = 2.7 g/cc and porosity measurements are 11.8% for the Lake Beds Group, 30% for the Red Sandstone Group, 26% Karoo Supergroup and Precambrian Basement is 0.64% (Chaki et al. 2008).

Substituting Eq. (3) into the radiogenic dating Eq. (4) described by Warr et al. 2018 and references therein.

$$\text{Conc. } 40_{Ar} \left[\frac{\text{cm}^3}{\text{gram}} \text{ of host rock} \right] = 0.105 \times 40_K \times [e^{\lambda_{40}t} - 1] \quad (4)$$

Solving for residence time 't' for K–Ar equation will lead to Eq. (5);

$$t = \frac{\ln \left[\frac{40_{Ar} + (0.105 \times 40_{Ar})}{0.105 \times (40_K)} \right]}{\lambda_{40}} \quad (5)$$

Similarly solving for radiogenic residence time 't' for U–Th/He Eq. (6) will lead to a solution shown in Eq. (7);

$$\begin{aligned} 4_{He} & = 8 \times (238_U) \times [e^{\lambda_{238}t} - 1] + 7 \times (235_U) \times [e^{\lambda_{235}t} - 1] \\ & \quad + 6 \times (232_{Th}) \times [e^{\lambda_{232}t} - 1] \end{aligned} \quad (6)$$

$$t = \left[\frac{\ln \left[\frac{(4_{He}) \times 8 \times (238_U) \times 7 \times (235_U) \times 6 \times (232_{Th})}{8 \times (238_U) \times 7 \times (235_U) \times 6 \times (232_{Th})} \right]}{\lambda_{238} + \lambda_{235} + \lambda_{232}} \right] \quad (7)$$

where ^{238}U , ^{235}U , ^{232}Th and ^{40}K are elemental concentrations in the host rock including; U-238 (2.8 ppm), U-235 (2.8 ppm), U-232 (10.7 ppm) (Manya et al. 2007; Mshiu and Maboko 2012) and K-40 (2%) (Warr et al. 2018) respectively. The decay constants per year with values in parenthesis are as λ_{238} ($0.155125 \times 10^{-9} \text{ yr}^{-1}$), λ_{235} ($0.98485 \times 10^{-9} \text{ yr}^{-1}$), λ_{232} ($0.049475 \times 10^{-9} \text{ yr}^{-1}$) (Davis and Villeneuve 2001; Parsons-Davis et al. 2018; Brevart et al. 1982 and references therein) and λ_{40} ($0.557 \times 10^{-10} \text{ yr}^{-1}$) (Wetherill et al. 1956). The results for these radiogenic ages are as shown in Tables 6 & 7).

5 Discussion

5.1 Crustal radiogenic helium 'kitchen'

Our models reveal that the entire rock volume of the Tanzania Craton contributes to 46% – 50% ($\sim 124\text{WC}$) of ^4He flux in the EAHP (Table 1). The estimated average helium production potential in the whole EAHP sums to $\sim 22 \times 10^{-6}$ mol

$^4\text{He}/\text{m}^2 \text{ yr}$ including the cratons $\sim 19 \times 10^{-6} \text{ mol } ^4\text{He}/\text{m}^2 \text{ yr}$ and the Ubendian mobile belt $\sim 3.3 \times 10^{-6} \text{ mol } ^4\text{He}/\text{m}^2 \text{ yr}$ (Tables 1 and 2). These values are generally greater (up to ~ 9 times) than the global statistical averages calculated for major sedimentary basins which are up to $\sim 1.74 \times 10^{-6} - 2.2 \times 10^{-6} \text{ mol } ^4\text{He}/\text{m}^2 \text{ yr}$ (Torgersen 2010; Cheng et al. 2021). Elsewhere previous studies have reported helium flux from continental crust settings such as the Paris Basin $\sim 0.2 \times 10^{-6} - 7.9 \times 10^{-6} \text{ mol } ^4\text{He}/\text{m}^2 \text{ yr}$ and the Great Artesian Basin $\sim 0.9 \times 10^{-6} - 1.6 \times 10^{-6} \text{ mol } ^4\text{He}/\text{m}^2 \text{ yr}$, Saskatchewan lakes $\sim 0.2 \times 10^{-6} - 52 \times 10^{-6} \text{ mol } ^4\text{He}/\text{m}^2 \text{ yr}$ and Williston Basin $\sim 0.29 \times 10^{-6} - 1.34 \times 10^{-6} \text{ mol } ^4\text{He}/\text{m}^2 \text{ yr}$ which are inline with this study (Torgersen 2010; Cheng et al. 2021).

Such a significant amount of ^4He flux in the EAHP would require massive impingement of high heat flow triggering helium release most likely related to a mantle plume beneath the Tanzania Craton (Pik et al. 2006; Ebinger and Sleep 1998; Danabalan et al. 2022). For the Tanzania Craton, a thermal surge has been previously evidenced from thermal springs, seismic tomography and diamond kimberlitic rocks which serve as unique proxy for metasomatism and heat flow aspects in the region (e.g. Bulanova et al. 2004; Stiefenhofer and Farrow 2004; Park and Nyblade 2006; Brown et al. 2012; Ebinger et al. 2013). Since the Tanzania Craton has the highest contribution to ^4He flux (124 WC), it is likely that cratonic foundering and metasomatic weakening facilitates ^4He release and circulation over large distances

throughout the region (e.g. Shirey et al. 2013; Danabalan 2017; Table 1). Due to the fact that the crustal thickness in the region varies (Kachingwe et al. 2015; Didas et al. 2022), it is possible that the asymmetric geometry of a cratonic ‘keel’ associated with lithospheric-scale faults, may deflect the mantle plume unevenly between the two branches of East Africa Rift System (Koptev et al. 2015; Fig. 8). Similar to suggestions by Koptev et al. (2015), the variation of isotopic ratios and heat flow observed in this study over the RRB and the RVP may also support the asymmetrical magmatic and non-magmatic nature of the East African Rift System.

Since the helium production is a continuous process, several degassing models in this study (e.g., Tables 4 and 5) could suggest that the ^4He mass balance is largely affected by degassing surges triggered by transient tectonothermal regimes. However, since the heat flow decay in the continental crust takes up to ~ 800 Myrs (e.g. Sclater et al. 1980; Hu et al. 2000), each successive tectonothermal event recorded in the Rukwa Rift (Mulaya et al. 2022), implies transient degassing surges followed by a hiatus of crustal stability and tectonic quiescence associated with normal degassing under a steady-state thermal regime. We postulate that heat transfer from the plume magmatism makes a significant contribution to the conductive and convective heat flow recharge, injection and fluid dynamics in the region (e.g., Lysak 1992; Ebinger and Sleep 1998; Pik et al. 2006; Fig. 8). Notably high heat flow up to $99 \text{ mW}/\text{m}^2$ for the Rungwe Volcanic Province reflects a local thermal anomaly associated with

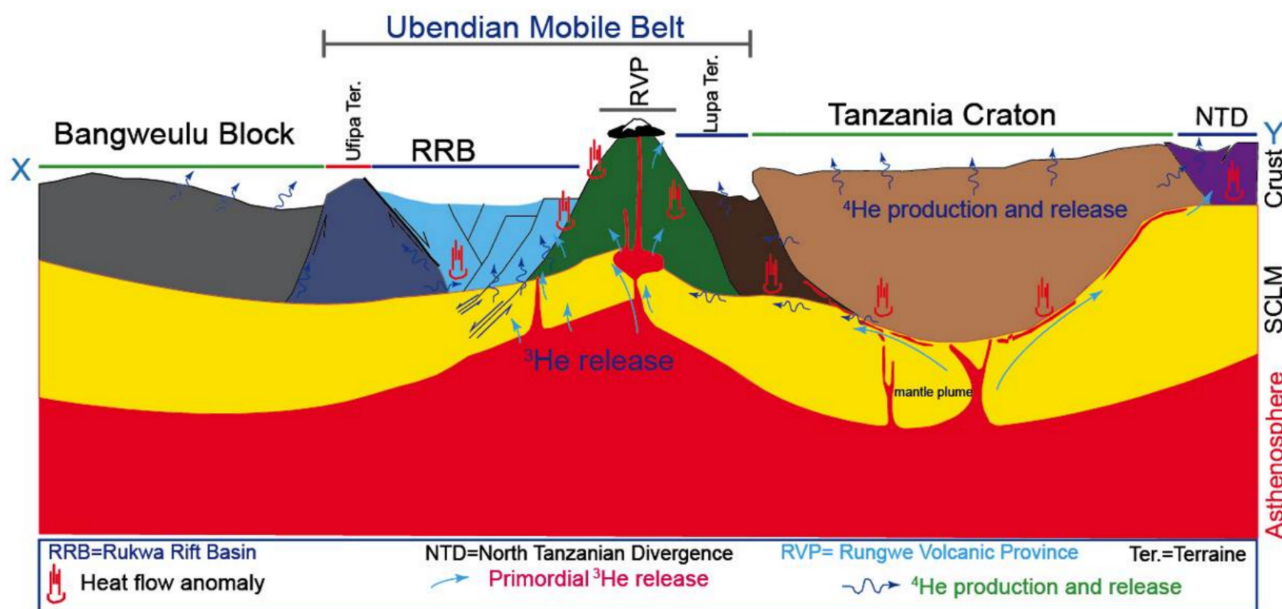


Fig. 8 Conceptual model showing the heat flow distribution for the EAHP related to plume magmatism. The heat flow anomaly triggered by the tectonothermal event is essential for releasing radiogenic helium from the continental crust. NTD. North Tanzanian Divergence; RRB. Rukwa Rift Basin; RVP. Rungwe Volcanic Province; SCLM. Sub-Continental Lithospheric Mantle (SCLM) (refer to Fig. 1A for map location) (after Koptev et al. 2015; Mtili et al. 2021; Kimani et al. 2021; Danabalan et al. 2022)

Table 4 Average radiogenic ages for each sampling locality based on the ^{40}K - ^{40}Ar Equation relating radiogenic isotopes where average residence times are calculated using bulk porosity (Φ) of each lithostratigraphic units in the Rukwa Rift Basin which are Lake Beds Group (LKB), Karoo and Basement. Concentrations of ^{40}Ar isotopes expressed per cm^3 of pore water (degassed water volume) of respective lithology using Vg/Vw after Mtili et al. (2021), Warr et al. (2018), Holland et al. (2013). For each level both individual samples and an average concentration is presented

Sample Location	$[\text{Vg/Vw}]$ ^{36}Ar	^{40}Ar [cm^3 STP/ cm^3]	^{40}Ar [cm^3 / $\text{cm}^3 \text{H}_2\text{O}$]	Φ [LKB]	Φ [RSS]	Φ [Karoo]	Φ [Basement]	Density [g/cc]	^{40}K ppm	λ_{40} Yr-1	Ages [LKB]	Ages [RSS]	Ages [Karoo]	Ages [Basement]
Ivuna mud-pots	7.50E-03	1.62E-02	5.45	0.118	0.3	0.26	0.0064	2.7	2.00E+04	5.57E-11	2.0E+06	5.17E+06	4.48E+06	1.10E+05
Itumbula salt pond	4.83E-02	9.59E-03	38.40	0.118	0.3	0.26	0.0064	2.7	2.00E+04	5.57E-11	1.4E+07	3.64E+07	3.16E+07	7.78E+05
Kajundu spring	4.40E-02	3.99E-03	13.90	0.118	0.3	0.26	0.0064	2.7	2.00E+04	5.57E-11	5.2E+06	1.32E+07	1.14E+07	2.81E+05
Tete spring	6.20E-02	3.10E-03	18.50	0.118	0.3	0.26	0.0064	2.7	2.00E+04	5.57E-11	6.9E+06	1.75E+07	1.52E+07	3.75E+05
Ibaya spring	4.32E+00	8.13E-05	1450.00	0.118	0.3	0.26	0.0064	2.7	2.00E+04	5.57E-11	5.3E+08	1.32E+09	1.15E+09	2.93E+07
Ngwilo spring	2.61E-01	1.37E-03	94.40	0.118	0.3	0.26	0.0064	2.7	2.00E+04	5.57E-11	3.5E+07	8.95E+07	7.76E+07	1.91E+06
Nanyara spring	3.88E+00	7.35E-05	1400.00	0.118	0.3	0.26	0.0064	2.7	2.00E+04	5.57E-11	5.2E+08	1.28E+09	1.12E+09	2.83E+07
Songwe river	2.05E+01	1.69E-05	6420.00	0.118	0.3	0.26	0.0064	2.7	2.00E+04	5.57E-11	2.3E+09	5.25E+09	4.63E+09	1.30E+08

Table 5 Average radiogenic ages for each sampling locality based on equation relating radiogenic uranium, thorium and helium isotopes. The average residence times are calculated using bulk porosity (Φ) of each lithostratigraphic units in the Rukwa Rift Basin which are Lake Beds Group (LKB), Red Sandstone Group (RSS), Karoo and Basement. Generally, the age ranges in Neo-Mesoproterozoic period (See description in text)

Sample Location	^4He [$\text{cm}^3\text{STP}/\text{cm}^3$]	^4He [$\text{cm}^3/\text{cm}^3\text{H}_2\text{O}$]	^4He [cm^3/gm Rock]	$[\text{V}_g/\text{V}_w]_{^{36}\text{Ar}}$	^{238}U [ppm]	^{232}Th [ppm]	$\lambda_{238} \text{Yr}^{-1}$	$\lambda_{235} \text{Yr}^{-1}$	$\lambda_{232} \text{Yr}^{-1}$	Ages [LKB]	Ages [RSS]	Ages [Karoo]	Ages [Basement]
Ivuna mud-pots	4.7E-02	1.58E+01	6.9E-01	0.0075	2.8	10.7	1.55E-10	9.85E-10	4.95E-11	8.1E+08	8.9E+08	8.8E+08	5.7E+08
Itumbula salt pond	3.4E-02	1.36E+02	6.0E+00	0.04825	2.8	10.7	1.55E-10	9.85E-10	4.95E-11	9.9E+08	1.1E+09	1.1E+09	7.5E+08
Kajundu spring	2.0E-04	6.95E-01	3.0E-02	0.044	2.8	10.7	1.55E-10	9.85E-10	4.95E-11	5.5E+08	6.3E+08	6.2E+08	3.1E+08
Tete spring	8.0E-03	4.77E+01	2.1E+00	0.062	2.8	10.7	1.55E-10	9.85E-10	4.95E-11	9.0E+08	9.8E+08	9.7E+08	6.6E+08
Ibaya spring	1.0E-04	1.78E+03	7.8E+01	4.32	2.8	10.7	1.55E-10	9.85E-10	4.95E-11	1.2E+09	1.3E+09	1.3E+09	9.6E+08
Ngwilo spring	1.1E-03	7.23E+01	3.2E+00	0.2605	2.8	10.7	1.55E-10	9.85E-10	4.95E-11	9.4E+08	1.0E+09	1.0E+09	7.0E+08
Nanyara spring	2.0E-05	3.76E+02	1.6E+01	3.8825	2.8	10.7	1.55E-10	9.85E-10	4.95E-11	1.1E+09	1.1E+09	1.1E+09	8.3E+08
Songwe river	7.8E-07	2.97E+02	1.3E+01	20.4935	2.8	10.7	1.55E-10	9.85E-10	4.95E-11	1.1E+09	1.1E+09	1.1E+09	8.1E+08

tectonics and suggests a possible mechanism for continual heat flow recharge in the EAHP (e.g., Njinju et al. 2019; Fig. 8). However, a tectonothermal event related to a mantle plume may not necessarily be the only thermal source as radiogenic heat production and intra-crustal magma chambers may contribute to the heat surge in the region (Sclater et al. 1980; Pollack 1982). The assumption made for degassing and migration of crustal ^4He may equally depend on other factors in place such as the intensity of crustal opening such as fracturing, efficiency of release from the helium bearing minerals and concentration of other carrier fluids (Halford et al. 2022; Danabalan et al. 2022).

5.2 Implication of heat flow anomalies for helium potential

The occurrence of heat flow anomalies in the Rukwa Rift marked by corresponding changes in the topographic profile coincides with notable changes in isotopic ratios (Figs. 4D, 6B). This relationship can be observed on a profile across the RRB and the RVP where the changes in isotopic ratios $\text{CO}_2/{}^3\text{He}$ and ${}^3\text{He}/{}^4\text{He}$ coincide with changes in topography (Fig. 6A–C). The relationship of heat flow to ${}^3\text{He}/{}^4\text{He}$ and $\text{CO}_2/{}^3\text{He}$ is attributable to binary mixing trend between crust-derived gases and magmatic-derived gases whereas proximity to active volcanic center that is, RVP accounts for CO_2 anomaly (Mtili et al. 2021; Danabalan et al. 2022).

Pronounced high topography over the RVP up to ~2250 m is associated with anomalously high heat flow (~99 mW/m^2) and isotopic ratios that is, high $\text{CO}_2/{}^3\text{He}$ $\sim 1.4 \times 10^{13}$ – 2.9×10^{13} and high ${}^3\text{He}/{}^4\text{He}$ ~ 3.3 – 7.0 (Fig. 6A–C). The topographic high in the RVP may be related to crustal uplift due to a thermo-kinematic anomaly contributing to regional stress fields reflecting extension tectonics and doming in the region (Mareschal and Gliko 1991; Negredo et al. 1995).

The anomalies of heat flow and isotopic ratio in some places coincide with tectonically active fault networks in the RRB and RVP most likely due to increased porosity and deep-rooted fracture/fault permeability (Lysak 1992; Mulaya et al. 2022; Halford et al. 2022; Fig. 8). The evidence of the heat flow anomaly occurring along/proximal to faults and accommodation zones in the Rukwa Rift reveal tectonic strain and permeable conduit systems which may facilitate fluid migration in the region (e.g., Lowenstern et al. 2014; Halford et al. 2022; Mulaya et al. 2022). The fact that the seismicity record and geodetic studies show high frequency of earthquakes with significant magnitude (e.g., Stamps et al. 2018; Fig. 7) implies heat flow anomaly is associated with active tectonics which could influence fluid migration. Thermal anomalies in active tectonics have been associated with circulation of hydrothermal fluid and seismicity in the region (e.g., Wilks et al. 2017).

Table 6 Postulate-I: Helium emissions and accumulation models for various tectonothermal events in the Rukwa Rift Basin. *Note* the models highlighted in green show realistic cases

CASE - I	Accumulation Time (Myr)	Initial Production (at least)
MODEL - I: Assuming similar ^4He emissions were discharged since onset of East African rifting coeval with magmatism (8.6 Ma- Harkin 1960; Ebinger 1989) via the whole region	2.34E+03	2349 Palaeoproterozoic
MODEL - II: Assuming similar ^4He emissions were discharged during the onset of East African rifting coeval with magmatism (8.6 Ma- Harkin 1960; Ebinger 1989) via only Rukwa Basin and RVP	4.E+02	409 Ma (Early Devonian)
MODEL - III: Assuming similar ^4He emissions were discharged since onset of Karoo rifting via the whole region (Late Carboniferous 300 Ma-Delvaux 2001)	8.16E+04	
MODEL - IV: Assuming similar ^4He emissions were discharged since onset of Karoo rifting via the Rukwa Rift (Late Carboniferous 300 Ma-Delvaux 2001)	1.52E+04	
MODEL - V: Assuming similar emissions were discharged since Cretaceous Carbonatite magmatism (120 Ma) via whole region	3.26E+04	
MODEL - VI: Assuming g similar emissions were discharged since Cretaceous Carbonatite magmatism (120 Ma) via Rukwa Rift	6.E+03	

More than 50% of the $\text{CO}_2/{}^3\text{He}$ ratios in this study range between $\sim 0.5 \times 10^{10}$ and 2.9×10^{13} (Figs. 5A, B and 6A) that implies a binary source between crust and mantle signatures in the Rukwa Rift as suggested by Barry et al. (2013) and Danabalan. (2017). The observed spatial variation of geochemical anomalies and heat flow along the strike of the Rukwa Rift segments may be attributable to a bimodal crustal thickness distribution ~ 29 and 42 km previously reported in the region (e.g., Last et al. 1997; Plasman et al. 2017; Borrego et al. 2018). This lithospheric thickening and thinning profile may have caused a variable rifting intensity and fluid dynamics (e.g., Lysak 1992; Lemna et al. 2019; Kolawole et al. 2021; Kimani et al. 2021; Mtili et al. 2021; Mulaya et al. 2022). Similarly, the variation of active strain patterns mostly inherited from Karoo fabrics may equally modulate the heat flow through deflection of thermal weakening effects within individual rifts (Bellahsen et al. 2013; Kolawole et al. 2021; Mulaya et al. 2022).

Along the East Africa Rift System, heat flow has been reported to vary considerably depending on the stages of rifting with highest heat flow recorded in the Afar Rift (~ 150 – 250 mW/m^2) while the lowest is in the Malawi Rift ~ 8 – 24 mW/m^2 (Lysak 1992; Jones 2020; Fig. 3a). However, the heat flow values in the RRB and RVP are higher than both the average value for continental provinces ~ 49 mW/m^2 (Sclater 1980) and the global average for Mid-Permian provinces (~ 58 mW/m^2) during deposition of Karoo sediments (Polyak and Smirnov 1968; Chapman and Pollack 1975; Kilembe and Rosendahl 1992; Njinju et al. 2019; Fig. 3B). Most of the heat flow data values are also above the global average for recent Eocene Provinces ~ 72 mW/m^2 and differs by 10%–18% (Polyak and Smirnov 1968; Chapman and Pollack 1975; Fig. 3A, B).

5.3 Reconstructing helium accumulation models

A number of feasible closed system models for various terranes surrounding the RRB provide insights into the mass balance between helium production and flux in the EAHP. These models are considered under two major postulates (Postulate 1 & 2) which correspond to the two cases of helium flux model (Case I & II) through specific area/terranes of conduits. Each postulate reveals fluid dynamics during certain tectonothermal events which acted as a ‘catalyst’ for helium production and release surge by assuming 100% efficiency of helium release in the region. The tectonothermal events which have been previously mapped include; initiation of the Karoo rifting (~ 300) (Kilembe and Rosendahl 1992; Delvaux 2001); carbonatite volcanism (~ 120 Ma) (Boniface 2017) and the recent East African rifting coeval with magmatism (8.6 Ma); (Ebinger et al. 1989; Ebinger and Sleep 1998).

5.3.1 Postulate—1

We illustrate six helium accumulation and degassing models based on helium degassing Case-I and in relation to tectonothermal events recorded in the RRB (e.g., Danabalan et al. 2022; Mulaya et al. 2022). Although six models were considered (Table 1), only two models can explain feasibly the volume of ${}^4\text{He}$ emissions from the EAHP;

- (1) *MODEL-I* Assuming constant ${}^4\text{He}$ emissions were discharged since the onset of East African rifting coeval with magmatism to date (8.6 Ma Harkin 1960; Ebinger et al. 1989) via the whole region (EAHP) (Table 6; Fig. 9a). This model shows unmatched mass balance between helium production and helium flux. In order to account for this unbalanced scenario, the

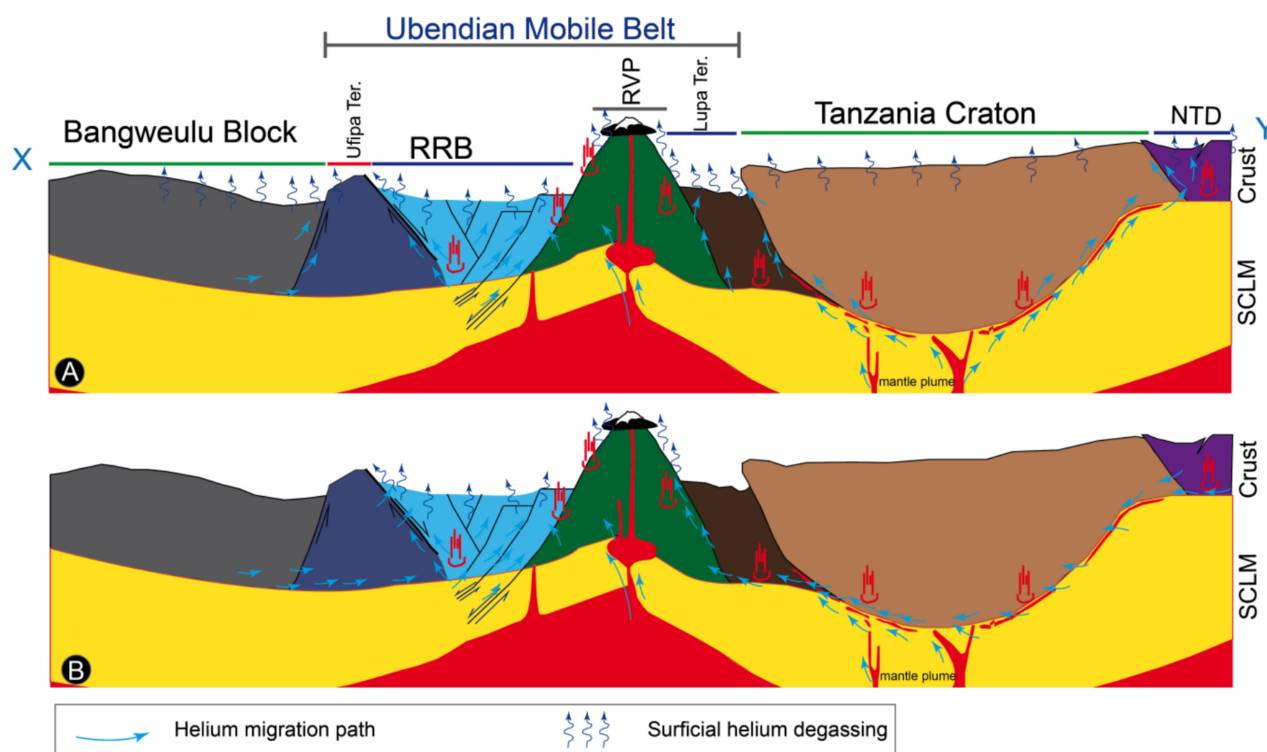


Fig. 9 Conceptual model showing the current helium migration and degassing model for the whole East Africa Helium Pool. **A** radiogenic degassing via the whole EAHP region. **B** degassing via the Rukwa Rift Basin (RRB) and the Rungwe Volcanic Province (RVP). Note the arrow-head indicates a proposed direction of regional migration

whole rock volume under the EAHP must have started ^4He production since at least the Palaeoproterozoic (2.349 Ga) with residence/accumulation time of at least 2340 Ma (Table 6). This timing coincides with the previously reported last phase of metamorphism in the Tanzania Craton at 2.4 Ga which serves as a proxy for high heat flow in the region (Pinna et al. 1994; Weeraratne et al. 2003; Danabalan et al. 2022).

MODEL—II Assuming ^4He emissions similar to those we observe today were discharged during the onset of East African rifting coeval with magmatism (8.6 Ma, Harkin 1960; Ebinger et al. 1989) via the Rukwa Basin and RVP which serve as the main conduits only. This degassing model can only be explained if the ^4He production had started since the Early Devonian (409 Ma) and accumulated for at least 400 Ma (Table 6; Fig. 9b).

Under Postulate-1, the mass balance calculations show that four models (Model-III-VI) out of six do not provide reasonable values since the degassing yields more radiogenic helium (hundred to thousand times) than the production capacity of the underlying crust. The assumption of a closed system model does not apply here since the accumulation time would extend far beyond the age of the Earth (beyond 4.6 Ga). Therefore, the unbalanced mass

computation between ^4He flux and production for these models may otherwise imply an open system involving an exogenous source rather than a closed system within the EAHP only (e.g., Halford et al. 2022).

5.3.2 Postulate—2

Mass balance results based on the assumption of the helium degassing model for Case II illustrated above and six plausible models for the helium production and release. These models assume 100% for both production and release efficiency from the source rocks within the EAHP.

- (1) **MODEL-I** Assuming ^4He emissions similar to those we observe today were discharged during the onset of East Africa rifting and coeval with magmatism (8.6 Ma, Harkin 1960; Ebinger et al. 1989) via the whole region (EAHP). This model suggests that the production capacity of the EAHP crustal rock volume would take 2.08 Myr to accumulate crustal ^4He in the region. The helium production would have therefore started in the Miocene (10.8 Ma) to account for the current flux rate observed in the region (Barry et al. 2013; Lee et al. 2016; Table 7; Fig. 9A).

Table 7 Postulate 2: Helium emissions and accumulation models for various tectonothermal events in the Rukwa Rift Basin

CASE-II	Accumulation time (Myr)	Initial production (at least)
<i>MODEL-I</i> Assuming similar ^4He emissions were discharged during onset of East African rifting coeval with magmatism (8.6 Ma, Harkin 1960; Ebinger et al. 1989) via the whole region	2.08E+00	10.68 Ma (Miocene)
<i>MODEL-II</i> Assuming similar ^4He emissions were discharged since onset of East African rifting coeval with magmatism (8.6 Ma, Harkin 1960; Ebinger et al. 1989) via only Rukwa Basin and RVP	3.89E-01	389 Ka (Quaternary)
<i>MODEL-III</i> Assuming similar ^4He emissions were discharged since onset of Karoo rifting via the whole region (Late Carboniferous 300 Ma, Delvaux 2001)	7.25E+01	372.5 Ma (Late Devonian)
<i>MODEL-IV</i> Assuming similar ^4He emissions were discharged since onset of Karoo rifting via the Rukwa Rift (Late Carboniferous 300 Ma, Delvaux 2001)	1.36E+01	313.6 Ma (Pennsylvanian)
<i>MODEL-V</i> Assuming similar emissions were discharged since Cretaceous Carbonatite magmatism (120 Ma) via whole region	2.90E+01	149 Ma (Late Jurassic)
<i>MODEL-VI</i> Assuming similar emissions were discharged since Cretaceous Carbonatite magmatism (120 Ma) via Rukwa Rift conduits	5.43E+00	125.43 Ma (Early Cretaceous)

- (2) *MODEL-II* This model is similar to Model-I above except that we assume the conduits of fluid flow to include both the Rukwa Basin and the RVP. The combined conduit areas over ^4He emission would therefore be smaller than the volume of rock and helium production capacity in the EAHP. The accumulation minimum time would therefore be reduced to helium released recently since the Quaternary (389 Ka) (Table 7; Fig. 9b).
- (3) *MODEL-III* In this model we postulate that the tectonothermal event during Karoo rifting from late Carboniferous 300 Ma (Delvaux 2001) would have paved a way for crustal instability and break up associated with helium release in the region. We therefore assume similar ^4He emissions observed today were discharged since onset of Karoo rifting where the helium was vented off through the whole region (EAHP). This model suggests that production of helium must have been started since the Late Devonian (372.5 Ma) having potentially accumulated for at least 72.5 Ma (Table 7).
- (4) *MODEL-IV* This model is similar to Model-III except the ^4He was vented through the Rukwa Rift only. To account for this emission, the helium production would have started since the Pennsylvanian time (313.6 Ma) and accumulated for 13.6 Ma (Table 7).
- (5) *MODEL-V* Assuming similar emissions we observe today were discharged since the Cretaceous (120 Ma) during carbonatite magmatism via the whole region (EAHP). Under this model, helium production would have initiated during the Late Jurassic (149 Ma) for the residence period of at least 29 Ma (Table 7).
- (6) *MODEL-VI* We assume similar conditions to Model-V except that the conduits for ^4He emissions were discharged via the Rukwa Rift. The time for helium release and accumulation would be 5.43 Ma which started

during the Early Cretaceous (125.43 Ma). This timing coincides with the previously reported carbonatite volcanism in the Rukwa Rift hence implying the influence of heat flow surge associated with rifting associated with volcanic dike events hence contributing to helium release. During this time the Red Sandstone sequences were simultaneously depositing whereas the Karoo sequences had already deposited with Top Karoo unconformity in place (Mulaya et al. 2022). Therefore, this timing could have favored accumulation of radiogenic helium in the Karoo sedimentary sequences. The petrophysical analysis of the Karoo sequences has been analyzed previously as 660.6 mD permeability and 26% porosity hence making it potential reservoir for helium accumulation.

The estimate of radiogenic ages for noble gases sampled in the Rukwa Rift show a bimodal cluster of noble gas release surges during recent East Africa Rift System and Pan-African orogenic cycles respectively (Fig. 10). The two episodes are known to have been associated with magmatism and metamorphism respectively (Ebinger et al. 1989; Boniface 2009). We infer that although helium has been continually generated from basement since the Precambrian to date, the release surges were during the Pan-African orogenic cycles and the recent East Africa Rift System. The mechanism of migration of released radiogenic noble gases most likely involved mainly vertical diffusion followed by petrophysical entrapment in pore fluids of the lithostratigraphic sequences in the Rukwa Rift (e.g., Cheng et al. 2021). However, for both K–Ar and U–Th/He methods, some of the computed ages from the same locality show a disparity in the radiogenic ages. Age disparity between calculated and known ages of lithostratigraphic units most likely imply an exogenous source for radiogenic noble gases via vertical

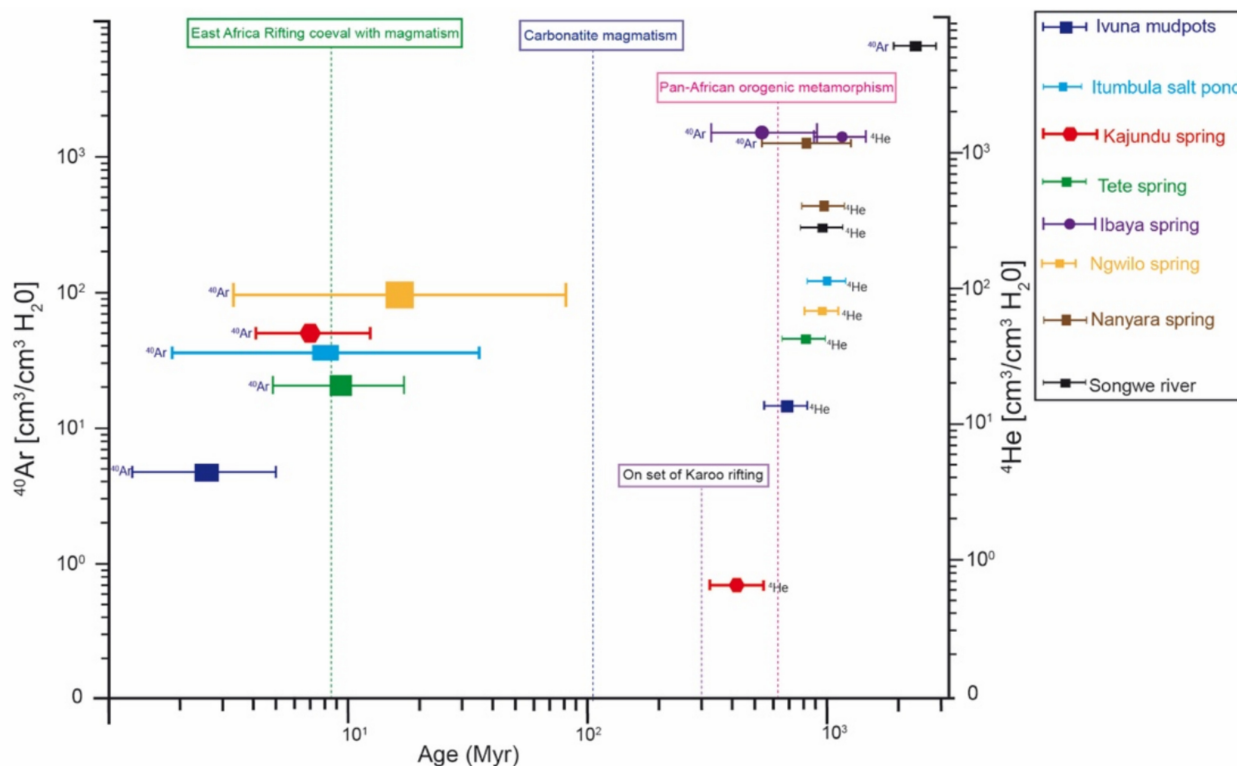


Fig. 10 Distribution of average radiogenic ages given in Ma based on ^{40}Ar and ^4He for eight ($N=8$) sampling locality in the Rukwa Rift (adopted from Warr et al. 2018). The observed uncertainty in ages is due to variation of porosities as discrete inputs in calculation of radiogenic ages which include 11.8% (Lake Beds Group), 30% (Red Sandstone Group), 26% (Karoo Supergroup) and 0.64% (Precambrian Basement) based on petrophysical analysis (Chaki et al. 2008). Note the two clusters of radiogenic helium release during the Precambrian Pan-African orogenic cycles and the recent East Africa Rift System. Major regional geological events are based previous publications (Ebinger et al. 1989; Kilembe and Rosendahl 1992; Delvaux 2001; Boniface 2009)

diffusion of helium within a closed system that is, overlying sedimentary units supplied by Precambrian Basement (e.g., Cheng et al. 2021). Disparities of radiogenic ages may also imply interplay of other factors e.g. assumptions made in each method and history path of radiogenic minerals including, slow cooling of rocks, alteration and recrystallization of clock minerals which can not be ruled out in this study (e.g., Curtis and Reynolds. 1958; Lippolt et al. 1994; Taylor and Aitken. 1997).

6 Conclusions

Mass balance computation in this study concludes that a comprehensive closed system of the EAHP has a crustal mass of $\sim 7.0 \times 10^{10}$ megatonnes and has the capability of producing ^4He of about $\sim 9.9 \times 10^6$ mol per year. The Tanzania Craton contributes largely to radiogenic helium production and release contributing up to 50% in the region. Such helium accumulation in the EAHP would have started since the Paleoproterozoic. However, subsequent release has been

accelerated largely by episodic tectonothermal events in the region. Apart from the helium potentiality in the EAHP, heat flow anomaly presents a proxy for geothermal energy targets.

The changes of heat flow anomalies in the Rukwa Rift that is, between 64 and 99 mW/m^2 are marked by corresponding changes in topographic profile. The RRB is a potential setting for radiogenic helium accumulation, however, the presence of other factors such as trapping and preservation of the accumulated helium are crucial for a potential helium resource.

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Declarations

Conflict of interest The authors declare no conflicts of interest that are relevant to the content of this article.

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