



The principles of helium exploration

Diveena Danabalan¹, Jon G. Gluyas^{1*}, Colin G. Macpherson¹,
Thomas H. Abraham-James^{2,‡}, Josh J. Bluett², Peter H. Barry^{3,4} and Chris J. Ballentine³

¹ Department of Earth Sciences, Durham University, Science Labs, Lower Mountjoy, South Road, Durham DH1 3LE, UK

² Helium One Global Limited, PO Box 957, Offshore Incorporations Centre, Road Town, Tortola, British Virgin Islands

³ Department of Earth Sciences, University of Oxford, South Parks Road, Oxford OX1 3AN, UK

⁴ Marine Chemistry and Geochemistry Department, Woods Hole Oceanographic Institution, 266 Woods Hole Road, Woods Hole, MA 02543, USA

JGG, 0000-0002-9386-7206; CGM, 0000-0001-5302-6405; PHB, 0000-0002-6960-1555; CJB, 0000-0001-9382-070X

Present addresses: THA-J, Conico Ltd, Level 15, 197 St George's Terrace, Perth, WA 6000, Australia

* Correspondence: j.g.gluyas@durham.ac.uk

Abstract: Commercial helium systems have been found to date as a serendipitous by-product of petroleum exploration. There are nevertheless significant differences in the source and migration properties of helium compared with petroleum. An understanding of these differences enables prospects for helium gas accumulations to be identified in regions where petroleum exploration would not be tenable. Here we show how the basic petroleum exploration playbook (source, primary migration from the source rock, secondary longer distance migration, trapping) can be modified to identify helium plays. Plays are the areas occupied by a prospective reservoir and overlying seal associated with a mature helium source. This is the first step in identifying the detail of helium prospects (discrete pools of trapped helium). We show how these principles, adapted for helium, can be applied using the Rukwa Basin in the Tanzanian section of the East African Rift as a case study. A thermal hiatus caused by rifting of the continental basement has resulted in a surface expression of deep crustal gas release in the form of high-nitrogen gas seeps containing up to 10% ⁴He. We calculate the total likely regional source-rock helium generative capacity, identify the role of the Rungwe volcanic province in releasing the accumulated crustal helium and show the spatial control of helium concentration dilution by the associated volcanic CO₂. Nitrogen, both dissolved and as a free-gas phase, plays a key role in the primary and secondary migration of crustal helium and its accumulation into what might become a commercially viable gas pool. This too is examined. We identify and discuss evidence that structures and seals suitable for trapping hydrocarbon and CO₂ gases will likely also be efficient for helium accumulation on the timescale of the Rukwa Basin activity. The Rukwa Basin prospective recoverable P₅₀ resources of helium have been independently estimated to be about 138 BSCF (billion standard cubic ft: 2.78 × 10⁹ m³ at STP). If this volume is confirmed it would represent about 25% of the current global helium reserve. Two exploration wells, Tai 1 and Tai 2, completed by August 2021 have proved the presence of seal and reservoir horizons with the reservoirs containing significant helium shows.

This article is part of the Energy Geoscience Series available at <https://www.lyellcollection.org/cc/energy-geoscience-series>

Received 31 March 2021; revised 22 December 2021; accepted 4 January 2022

The global helium provinces today, the US Mid-Continent, Canada, Algeria, Qatar/Iran, central Europe, Russia and Australia, are all areas of petroleum production. The history of helium exploration (Gluyas 2019a, b) is one of serendipity, helium only being discovered as a small fraction of the produced natural gas (methane). Many petroleum companies vent helium alongside other waste gases either because they do not know there is helium in their gas mixture or they fail to recognize its value (Clarke *et al.* 2012).

The USA held a near monopoly on helium supply throughout the twentieth century. However, new discoveries have not delivered the volume required to meet growing demand, particularly those associated with the use of helium in medical cryogenics. Currently, growth in the helium market is around 11% per annum (Research and Markets 2020). In the last decade both Algeria and Qatar have begun to sell helium extracted from hydrocarbon gas produced for liquified natural gas exports. The concentration of helium in both Algerian and Qatari petroleum gas is typically below 0.1%. Should demand for liquified petroleum gas decline, then extraction and sale of helium from this source would not remain economical.

The failure of petroleum exploration to deliver additional reserve volumes of associated helium has led to three periods of global shortage of helium since 2010. This caused interruption of some

hospital services in the UK and elsewhere, and universities were informed that demand would not be met (Connor 2013; Stokes 2013; Kalin and Finn 2017; Murphy 2019). Given the importance of helium to modern society and its irreplaceable use in medical cryogenics, leak detection, as a lift gas and in breathing mixtures, it came as a surprise to the authors that back in 2011, when we began this work, we could find no exploration strategies specifically developed for helium. A tested and successful helium exploration strategy was clearly needed (American Physical Society *et al.* 2016; Ballentine 2017).

Here, established petroleum exploration questions have been adapted to create a helium exploration strategy to enable regions globally to be assessed for their helium prospectivity. The strategy was developed by addressing the following questions (Table 1):

- (1) What are the source rocks for helium?
- (2) How do the source rocks mature for helium generation?
- (3) What causes helium to escape from its source rock (primary migration)?
- (4) How is helium transported throughout the subsurface (secondary migration)?
- (5) How is helium concentrated in subsurface reservoirs to form a helium accumulation?

Table 1. *Synthesis of components required for a viable helium province*

Stage	Helium system
Source	U^{238} , U^{235} and Th^{232} decay in the crust, producing alpha particles that acquire electrons and become helium
Maturation	Time to accumulate (stability of crust) v. volume of crust
Primary migration	Heat to above the closure temperatures of minerals with respect to helium retention v. diffusion. Release of nitrogen from associated minerals and clays. Tectonism and rock fracturing to release fluids.
Secondary migration	Gas buoyancy and/or solution in water and hydrostatic drive
Accumulation in reservoir	Direct input into trap of a buoyant free-gas phase; or degassing of oversaturated groundwater; or gas stripping via hydrocarbon (CH_4) or magmatic (CO_2) gas phase
Trap integrity and longevity	Risk of: microseepage; capillary failure; fracture failure; tectonic destruction of trap

(6) How long do helium-rich gas accumulations remain in reservoirs?

By answering these questions, it then becomes possible to develop an exploration strategy for helium that is similar to that for petroleum; to develop plays in which a reservoir–seal combination exists and where there is a mature helium source rock. Here we use the historical discovery of nitrogen- and helium-rich natural gas seeps in the Rukwa Basin, Tanzania (James 1967a, b), to provide the geological environment in which to test the concepts required for a successful commercial helium system in a low petroleum prospectivity setting.

Defining the helium system

Helium source rocks and helium generation

The dominant stable isotope of helium, 4He , is sourced radiogenically from the alpha decay of ^{238}U , ^{235}U and ^{232}Th isotopes in Earth's mantle and crust. Thus, a helium source rock will be one that has the combination of U and Th concentrations and age to provide the helium. Radioactive decay is a first-order physical process in which the rate is dependent upon the absolute amount of the isotope and is unaffected by temperature or pressure. It is measured in terms of radioactive 'parent' half-life: that is, the time taken for half of the parent element to 'decay' to the daughter element. It follows that the older the rock is, the more the isotopes will have decayed, with the corresponding production of helium. The half-lives of the three relevant isotopes are: $^{238}U = 4.468$ Gyr (billion years), $^{235}U = 0.7$ Gyr and $^{232}Th = 14.05$ Gyr.

Table 2. *Helium closure temperatures for helium-retentive minerals*

Mineral	Closure temperature range (°C)	References
Apatite	55–100	Lippolt <i>et al.</i> (1994), Wolf <i>et al.</i> (1996), Farley (2000), Farley (2002), Shuster <i>et al.</i> (2006)
Hematite	90–250	Bähr <i>et al.</i> (1994), Farley (2002)
Zircon	180–200	Farley (2002), Reiners (2005), Reich <i>et al.</i> (2007), Cherniak <i>et al.</i> (2009)
Garnet	590–630	Dunai and Roselieb (1996), Farley (2002)
Monazite	182–299	Boyce <i>et al.</i> (2005)
Titanite	150–200	Reiners and Farley (1999), Farley (2002)
Uraninite	c. 200	Martel <i>et al.</i> (1990), Stuart <i>et al.</i> (1994)

Ranges in closure temperature for individual minerals are due to combinations of differing grain sizes and cooling rates.

Earth's cratonic basement rocks are both very old and commonly of granitic composition – rich in thorium at 20–30 ppm (René 2017), but containing only modest quantities of uranium (typically around 4 ppm: Adams *et al.* 1959). For example, basement rocks and cratons of Archean–Proterozoic age (3.8–0.54 Ga) in the Canadian Shield or the Yavapai–Mazatzal province in the southern USA are predominantly metamorphic or granitic, and contain on average 2.8 ppm U and 10.7 ppm Th (Burwash and Cumming 1976). Mafic basement rocks tend to have lower uranium and thorium contents (Rudnick and Fountain 1995; Heikal *et al.* 2013).

Maturation

During tectonically quiescent periods, subsurface concentrations of 4He naturally increase over time as uranium and thorium decay. The helium generated remains in both the mineral matrices and interstitial pore fluids (Reimer 1976; Bottomley *et al.* 1984; Zadnik and Jeffery 1985; Ballentine and Burnard 2002). This has been observed in the Canadian and Fennoscandian shields, Kaapvaal Craton and Greenland Craton (Zadnik and Jeffery 1985; Lippmann-Pipke *et al.* 2011; Holland *et al.* 2013; Neretnieks 2013; Warr *et al.* 2018).

The observation that shallow (<2 km) waters and gases in continental sedimentary systems contain low helium concentrations (ppm level) is mostly due to the ability of helium-retentive minerals, such as apatite, zircon, uraninite or monazite, to retain and accumulate helium below their closure temperatures (Table 2) (e.g. Ballentine and Burnard 2002), as well as the relatively young age of shallow sedimentary systems and associated fluids. For apatite, closure temperature is c. 70°C, indicating that under an average crustal temperature gradient of 30°C km⁻¹, assuming a surface temperature of 10°C, helium would only start being released from these minerals at 2 km or deeper. Since most helium-rich gas reservoirs generally occur at shallower depths (Table 3), helium atoms and thus helium accumulations both require a mechanism for their release from deeper regional geological systems, transport and a focusing mechanism.

Primary migration

The primary migration of helium is a two-stage process that requires initial migration (diffusion) out of helium-producing minerals into the interstitial porosity, and migration out of the source rock.

Radiogenic 4He produced from the alpha decay of ^{238}U , ^{235}U and ^{232}Th will be found within 10–20 µm of the parent radioelement (e.g. Martel *et al.* 1990). This is due to the penetration distance of the original alpha particle (Meunier *et al.* 1990). This distance is within the same length scale as most mineral crystals and grains. Thus, helium can become trapped both within the mineral matrix and on mineral grain boundaries, depending on the distance of the radionuclide from the grain boundary (Martel *et al.* 1990; Ballentine and Burnard 2002; Barry *et al.* 2015).

Table 3. Average depths of a selection of helium-producing reservoirs in the USA

Field and location	Producing reservoir	Helium concentration (%)	Depth of producing reservoir (km)	References
Hugoton–Panhandle, Kansas–Oklahoma–Texas, USA	Chase Group (Permian)/Council Grove Group (Permian)/Brown Dolomite (Permian)	0.60 (average)	0.90	Ballentine and Lollar (2002), Gage and Driskill (2005)
Woodside, Utah, USA	Kaibab (Permian)	1.31	0.95	Morgan and Chidsey (1991), Harris (1993)
Harley Dome, Utah, USA	Entrada (Jurassic)	7.02–7.18	0.26	Dobbin (1968), Danabalan (2017)
Model Dome, Colorado, USA	Entrada (Jurassic)	7.00–8.34	0.31	Dobbin (1968)
Greenwood, Kansas, USA	Topeka (Pennsylvanian) and Wabaunsee (Pennsylvanian)	0.52–0.70	0.94	Wingenter (1968), Gage and Driskill (2005)
McElmo Dome, Colorado, USA	Leadville (Mississippian)	≤0.71	0.50	Gerling (1983), Gilfillan (2006), Gilfillan <i>et al.</i> (2008)
LaBarge, Wyoming, USA	Madison (Mississippian)	0.50–0.73	4.42–5.03	Hamak (1989), Stewart and Street (1992), De Bruin (1995), Martin <i>et al.</i> (2008), Merrill <i>et al.</i> (2014)

The first stage of primary migration, the diffusion of helium from producing minerals into the surrounding pore or fracture space, enables high concentrations of helium to accumulate in tectonically quiescent areas of the crust (Lippmann-Pipke *et al.* 2011; Holland *et al.* 2013; Neretnieks 2013; Warr *et al.* 2018). The bulk diffusion of ^4He in the crust is limited in terms of length scale. Diffusion and buoyancy-driven fluid advection can both contribute to the migration of helium from the crust into the sedimentary cover, with advection likely to be the dominant process in regions of recent tectonic activity and diffusion controlling helium release in more quiescent systems (Torgersen 1989, 2010; Hussain 1997; Ballentine and Burnard 2002; Neretnieks 2013; Cheng *et al.* 2018).

In the case where advection is responsible for helium transport from the deep continental crust to shallow regions, then two conditions are required. First, a thermal event, one which is significant enough to overcome the closure temperature associated with the various minerals within which helium is trapped and to generate, via fluid-overpressure-induced fracturing, an interconnected fracture fluid migration pathway (Warr *et al.* 2018); Secondly, a carrier fluid is required to facilitate advective movement out of the source rock. The carrier phase may be groundwater with the gases held in solution, or a gas phase such as N_2 , CO_2 or CH_4 . Both conditions require a significant increase in the regional thermal gradient, which is likely to be caused by tectonism such as extensional rifting, orogeny or volcanic activity. Moreover, the timing needs to be correct insofar as the source rock needs to be mature and replete with helium.

An example of the primary migration of ^4He is currently occurring within Yellowstone National Park in the USA (Lowenstern *et al.* 2015). Calculations show that the supervolcano, which is heating the Archean-aged Wyoming Craton, is releasing the ^4He accumulated within the craton source alongside its magmatic CO_2 carrier gas.

In subsurface reservoirs, helium is always found with nitrogen, although nitrogen can also occur without significant helium. $^4\text{He}/\text{N}_2$ in natural gas fields typically ranges between 0.02 and 0.20 (Pierce *et al.* 1964; Poreda *et al.* 1986; Gold and Held 1987; Jenden *et al.* 1988a; Jenden and Kaplan 1989; Stilwell 1989; Hiyagon and Kennedy 1992; Hutcheon 1999) and can be a mixture of helium-associated nitrogen and other sources of nitrogen (Ballentine and Sherwood Lollar 2002). The isotopic ($\delta^{15}\text{N}$) composition of the N_2 end member associated with the economic ^4He in the Kansas–Texas Hugoton–Panhandle gas field falls within a very narrow range (–3.00 to +2.45‰). This compares with the range seen from both low-temperature metamorphism of the crust and the release of ammonia from clays (–5.00 to + 4.00‰) (Zhu *et al.* 2000; Ballentine and Lollar 2002; Danabalan 2017). That radiogenically produced helium is associated with non-radiogenic nitrogen suggests they share a process in common, although their source rocks could be different; nitrogen, as the major gas, may be acting as an advective carrier gas phase for the helium or providing the primary mechanism of gas phase formation from solution.

The link between ^4He and N_2 during primary migration is discussed in a study of the Eger Rift by Weinlich *et al.* (1999) with an added caveat: while this region exemplifies primary migration for the crustal-sourced helium system, it is also shows the potential dilution of ^4He - and N_2 -rich gases by the addition of magmatically sourced CO_2 and associated gases in a tectonically active region, also seen in the Yellowstone example discussed earlier (Weinlich *et al.* 1999; Lowenstern *et al.* 2015).

Gases from the Eger Rift in central Europe show a CO_2 dilution trend for N_2 and ^4He concentrations close to current geothermal centres in the area (Fig. 1). This is an indicator that tectonism such as rifting and associated volcanism and high heat flows can facilitate the primary migration of helium out of previously quiescent source rocks. If a trap is already in place when tectonism occurs, there is a

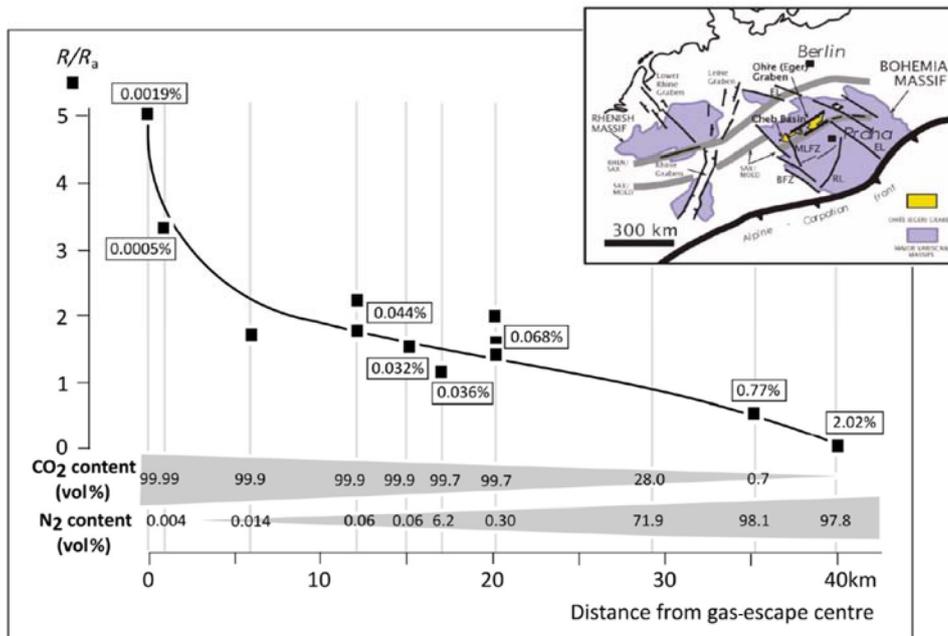


Fig. 1. $^3\text{He}/^4\text{He}$ ratio (R/R_a) in natural gas seeps plotted as a function of distance from the geothermal system in the Egger Graben, Czech Republic (inset) (modified from Weinlich *et al.* 1999). Samples close to the gas-escape centre contain only trace levels of ^4He (percentage shown in boxes), are CO₂ dominated and have $^3\text{He}/^4\text{He}$ ratios $>1R_a$ showing a significant mantle helium contribution (e.g. Ballentine and Burnard 2002). Between 20 and 30 km distance from the geothermal centre the gas composition transitions from volcanic/geothermal CO₂ to dominantly nitrogen and, notably, per-cent-level helium with $^3\text{He}/^4\text{He} < 1R_a$. This is consistent with a significant contribution of crustal-derived gas in the distal fluids. This pattern is observed in other systems (e.g. Barry *et al.* 2013).

possibility that the upward migration of ^4He and N₂ as a free-gas phase may be all that is needed to generate a helium-rich reservoir. If the primary fluid contains no gas phase, or mixing with shallower groundwater causes any gas phase to be fully dissolved into solution, the salinity and pressure–temperature regime controlling the gas saturation point of the groundwater will need to be exceeded for a helium-rich gas reservoir to form.

However, primary vertical migration alone cannot explain the presence of large-volume helium-rich gas in areas that have not experienced any recent major tectonic activity, such as in Kansas, USA (Jenden *et al.* 1988b) or the Williston Basin, USA–Canada (Cheng *et al.* 2018). Such cases instead may require long-distance secondary migration (i.e. lateral migration of helium) or diffusional control.

Secondary migration

Secondary migration of helium is the lateral and vertical movement of helium and other associated gases after primary migration has occurred. Secondary migration can occur as free-gas migration or movement of groundwater containing dissolved gases (Table 4), or, be it very slowly, by diffusion through static fluids (Torgersen 1989, 2010; Cheng *et al.* 2018).

Observed correlations between air-saturated-water (ASW)-derived noble gas isotopes such as ^{20}Ne and ^{36}Ar , and admixtures of ^4He and ^4He -associated N₂ in fields from Kansas, Oklahoma,

Texas and Arizona show strong evidence of groundwater involvement in helium-rich systems (Ballentine and Lollar 2002; Gilfillan *et al.* 2008; Danabalan 2017). In addition to groundwater movement, regional uplift or other mechanisms resulting in a pressure drop could also cause the carrier gas, and hence helium, to exsolve from solution to generate a free-gas phase (e.g. Sorenson 2005).

Accumulation in reservoirs

When groundwater containing dissolved ^4He and N₂ equilibrates with a CO₂ or CH₄ gas phase, insoluble ^4He and N₂ will preferentially exsolve from the groundwater and partition into the free-gas phase (gas stripping). In this way hydrocarbon (or CO₂) generation and buoyant migration as a gas phase through a groundwater containing accumulated but dissolved helium and nitrogen will result in a helium-rich hydrocarbon (CO₂) phase, with the resulting helium concentration dependant on both the amount of helium accumulated in the groundwater and the hydrocarbon gas/water ratio. Similarly, a pre-existing hydrocarbon or CO₂ gas phase within a geological trapping structure promotes the degassing of groundwater containing accumulated ^4He and N₂ (e.g. Barry *et al.* 2016, 2017). This process operated to form most of the helium deposits currently exploited. Gas stripping by its nature dilutes any ^4He , and commercial gas fields containing admixtures of CH₄ or CO₂ contain at most concentrations of up to a few per cent of

Table 4. Possible mechanisms of secondary migration and gas phase formation for the helium system

Secondary migration mechanism	Description
Free-gas phase – gas buoyancy	<ul style="list-style-type: none"> The ^4He–N₂ gas phase released from minerals into proximal fracture porosity migrates surfacewards by gas buoyancy along faults or lithologies with enhanced porosity and permeability
Groundwater flow – hydrostatic	<ul style="list-style-type: none"> The ^4He–N₂ gas phase from primary migration or after limited secondary migration contacts groundwater and at high pressure is dissolved within the groundwater ^4He–N₂-rich groundwater migrates due to hydrostatic drive. The formation of a ^4He–N₂ gas phase may be subsequently caused by changes in water temperature, salinity or pressure that cause ^4He–N₂ saturation within the migrating water
The role of secondary CH ₄ or CO ₂	<ul style="list-style-type: none"> The addition of CO₂ and/or CH₄ to any ^4He–N₂-rich groundwater phase will enable gas saturation to be reached at a deeper level, but will also result in ^4He–N₂ being diluted in any exsolution gas Gas-buoyancy-driven migration to trap Migration of a ^4He–N₂-rich groundwater that contacts an existing gas phase (or vice versa) will result in dissolved ^4He–N₂ partitioning into the gas phase (so called ‘gas stripping’)

helium. There are, however, several gas accumulations that contain as much as 10% helium and 90% nitrogen. Examples include the Permian strata of the USA Kansas Basin, Central Kansas Uplift, as well as in N_2 - 4He -rich wells in Utah, Montana (USA) and Saskatchewan (Canada). In these occurrences, the role of dilutant gases has been minimal and the groundwater must have been oversaturated with 4He -associated N_2 . Depressurization of groundwater through uplift or upward migration of nascent fluids from deeper source rocks are mechanisms that would ensure the bubble point is exceeded for any dissolved gases and thereby the preservation or formation of a primary (generated in the source rock) 4He and 4He -associated N_2 free-gas phase.

Trap efficacy, leakage and destruction

Once 4He and associated gases have migrated into a gas trap, the preservation of 4He in that trap is a function of the rate at which 4He is supplied to the gas pool less the rate at which helium leaks out. The capillary entry pressure required to force helium gas through the seal is similar to that of CO_2 (Wollenweber *et al.* 2009). Thus, seals that are suitable for CO_2 and hydrocarbon gases would similarly trap helium with this mechanism. Diffusive loss, however, is a function of atomic or molecular mass, and helium has a higher diffusion coefficient than other gases.

Low-permeability seals such as evaporite minerals, or a cap rock with similar sealing efficiency, have the physical characteristics that would allow helium to be stored for long periods (>100 Myr) of time without significant diffusive leakage (Broadhead and Gillard 2004; Broadhead 2005; Cheng *et al.* 2018). There is also evidence that less efficient (higher-permeability) cap rocks could suffice. For example, several gas fields in the Pannonian Basin, a Miocene extensional basin with a high thermal gradient, contain helium concentrations at relatively high, although subcommercial, concentrations (Ballentine *et al.* 1991). Dominated by shale and mudrock seals, these gases contain radiogenic $^4He/^{40}Ar^*$ at a similar ratio to that predicted for production in the crust from regional estimates of the basement $(U + Th)/K$ (where $^{40}Ar^*$ is radiogenic ^{40}Ar produced from the decay of ^{40}K). The preservation of the predicted radiogenic $^4He/^{40}Ar^*$ in these gases suggests that 4He has not been preferentially lost, relative to the $^{40}Ar^*$, from the gas phase via diffusion on the timescale of trap filling. However, older reservoirs carry a greater risk of diffusional loss and loss due to tectonic events (similar to hydrocarbon plays).

Tanzania: a helium exploration case study

The Tanzanian section of the East African Rift System (EARS) is an exemplar region where all of the characteristics for development of a helium resource are met. In this sense, Tanzania represents a unique opportunity to develop a 'play fairway' approach for helium exploration. A play fairway is defined as the area occupied by a suitable reservoir rock for helium with an associated overlying seal horizon with access to migrated helium from its mature source rock. Within the area occupied by the play fairway one would expect to find prospects, configurations of seal and reservoir rock that could trap helium and other buoyant gases.

Geological background

The East African Rift (EAR) is a classic example of a continental rift. Crustal extension typically occurred through normal faulting that thinned the crust and resulted in upwelling of dense and hot lithospheric mantle rocks. The lithospheric mantle in turn deformed and was replaced by hotter asthenosphere. The process of heat transfer beneath the rift to the lithosphere reduced rock density and caused time-dependant regional uplift on the timescale of tens of

millions of years (McKenzie 1978; Sengör and Burke 1978). The location of crustal extension was often influenced by pre-existing and large-scale pre-rift tectonic boundaries (Nyblade and Brazier 2002).

The EARS has evolved broadly from north to south and can be divided into several discrete diachronous rift sectors. The northern end of the rift system in Ethiopia and Afar is the most mature where it is transitioning from continental rifting to seafloor spreading. In Kenya the rift is well advanced, whilst the southern end of the eastern branch of the EAR, in northern Tanzania, is still at a juvenile rifting stage. The earliest basaltic volcanism occurred between 45 and 39 Ma in southwestern Ethiopia, with a major phase of flood basalt volcanism by 31–30 Ma (Ebinger *et al.* 1993). Regional heating and widespread mantle metasomatism along the asthenospheric–lithospheric boundary, inferred from kimberlites in the Archean cratons around the rift, is likely to have occurred before any surface expression of rifting across the East African Plateau (Ebinger *et al.* 2013).

The most juvenile rift sector marks the southern termination of the eastern branch and is expressed by a pronounced splay in the Tanzanian Divergence Zone (TDZ). Volcanic activity began at 8–4 Ma in the centre of the TDZ, with volcanism extending to the outer graben after 4.5 Ma. The Manyara Rift has been affected by rifting over a larger area than other young rifting zones, and has retained a thick lithosphere and a lower crust that has not viscously deformed (Uwe 2014). Volcanism migrated southwards, with ages of 4.9–1.5 Ma in the northern Manyara Rift and 1.5–0.7 Ma in the southern branch (Bagdasaryan *et al.* 1973; Foster *et al.* 1997).

The western branch of the EAR extends over 2000 km, from Lake Albert in the north to Lake Malawi. In parts of the western branch, subsidence is at its greatest in the entire rift, while rift-flank uplift is also pronounced (Uwe 2014). In the Albertine Rift of western Uganda, the lithosphere has hardly thinned at all and the lower crust has not viscously deformed; the chemistry of the magmatic rocks is primitive (mantle derived) and the volume of magmatism is limited (Uwe 2014). Volcanism throughout the western branch occurs in four distinct centres (Toro-Ankole, Vrungu, South Kivu and Rungwe), but there is no clear trend in when the volcanism started, with estimates from three systems at *c.* 10 Ma (Rogers 2006; Ebinger *et al.* 2013).

The *c.* 350 000 km² Archean-aged Tanzanian Craton lies in the centre of Tanzania between two branches of the EARS (Fig. 2) (Weeraratne *et al.* 2003). It was emplaced at *c.* 2.7 Ga, and is primarily composed of greenstone belts and granite with ages upwards of 2.4 Ga (Pinna *et al.* 1994; Dawson 2008). The craton experienced several collisional events early in its history, resulting in peripheral mobile belts. The Usagaran (2.0–1.8 Ga) and Ubendian (2.1–1.8 Ga) mobile belts formed via subduction-related accretion, whereas the Mozambique Belt was formed by a later oblique collisional event (<1.3 Ga) and was then reworked by the multistage Pan African Orogeny (950–550 Ma) (Quennell 1956; Lenoir *et al.* 1994; Muhongo and Lenoir 1994; Mruma 1995; Fritz *et al.* 2005; Vogt *et al.* 2006; Dawson 2008; Macheyeke *et al.* 2008; Boniface *et al.* 2012).

Half-graben rift basins occur throughout the EARS; infill occurred from Permian to Recent times with accommodation space generated by episodic tectonism (Delvaux *et al.* 1992; Roberts *et al.* 2004, 2012a, b). In the western rift branch these sedimentary sequences can be up to 11 km thick in some basins (Wheeler and Karson 1994).

In its simplest form, generation of a play fairway requires convergence of four features as previously described: (i) a source rock that has generated a suitable quantity of helium; (ii) a process by which the helium is expelled from the rock that has generated it (primary migration); (iii) fluid migration from depth that might contain helium (secondary migration) and gas-phase formation; and

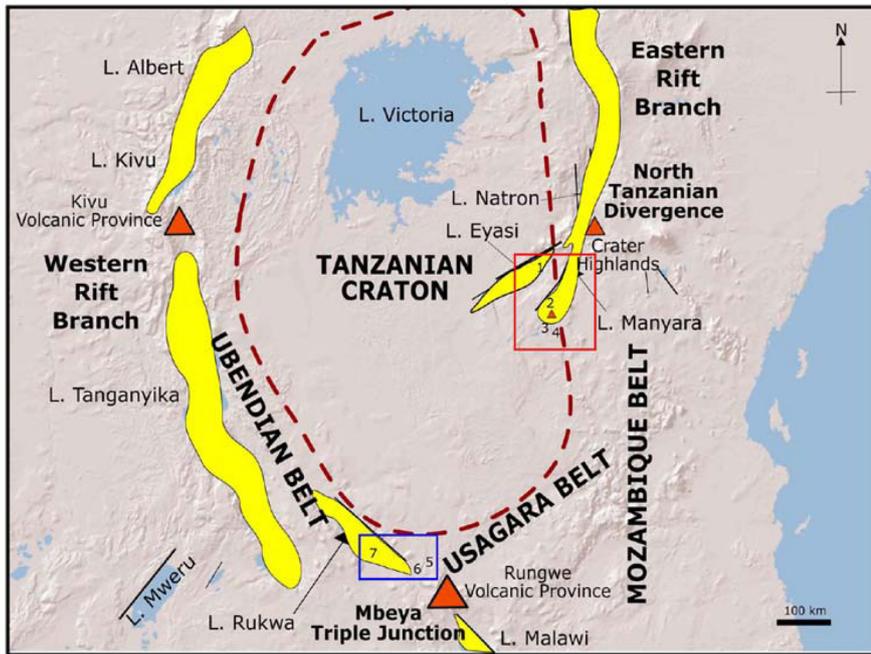


Fig. 2. Construction of a regional ‘play fairway’ for helium exploration in Tanzania. Major active volcanic complexes are shown by red triangles and significant sedimentary deposits in the rift valley are shown in yellow. Two regions with reported helium-rich seeps (James 1967a, b), outlined in blue and red, are underlain with ancient terrain that can provide a helium source rock (see the main text), are proximal to a major geothermal perturbation (the volcanic complex) and have the potential to trap helium-rich gas in local sedimentary structures. Helium concentrations in gas seeps from the literature and this study are shown in detail for the two highlighted regions in Figure 3a and b.

(iv) the presence of trapping structures in proximity to helium migration pathways and which formed prior to helium migration.

Developing a play fairway for Tanzania

The ^4He generation potential is an estimate of the volume of ^4He . We separate the Tanzanian system into five distinct regions of interest: (1) the Tanzanian Craton (350 000 km²); (2) the Ubendian Belt (75 000 km²); (3) the Usagara Belt (57 600 km²); (4) the North Tanzanian Divergence (NTD), which consists of parts of the reworked Usagaran Belt, Mozambique Belt and the Tanzanian Craton (this calculation is confined to the Gregory, eastern, rift arm 42 500 km²); and lastly, on a local scale, (5) the Rukwa Basin asymmetrical half-graben (Roberts *et al.* 2012a, b) (basement and sediment fill) within the Ubendian Belt (12 800 km²) (Fig. 2).

For each region we convert its surface area into a volume by considering helium generation to a depth of 10 km. The crustal thickness in these regions varies from 21 to 27 km (Getachew *et al.* 2011) but radioelement (U + Th) concentrations (2.8 ppm U and 10.7 ppm Th, respectively, values used in the calculations that follow) tend to be higher in shallower, less mafic regions of the crustal section above *c.* 10 km (Rudnick and Fountain 1995). A 10 km depth therefore provides a minimum estimate for the helium generation potential of any individual region. It is further assumed that this crust has an average crustal density of 2.7 cm³ g⁻¹ and an average porosity of 0.64% (Chaki *et al.* 2008). The thickness of the sediment-fill in the Rukwa Basin has a maximum depth of about 11 km (Roberts *et al.* 2012a, b), and the sandstone reservoir (Red Sandstone Group) porosity of up to 28% has been measured (Roberts *et al.* 2012a, b) (Table 5). The Red Sandstone Group occurs below the Lake Beds, which in turn has the potential to act as a regional seal.

In addition, an age is needed before which the helium content of the regional rock volume is assumed to be negligible, and from which subsequent helium generation will have accumulated and/or been released. For the Tanzanian Craton, the ‘resetting age’ is assumed to be the last phase of metamorphism at 2.4 Ga (Pinna *et al.* 1994; Weeraratne *et al.* 2003). For the NTD, this is assumed to be when it was accreted onto the Tanzanian Craton at 2.0 Ga

(Ebinger *et al.* 1997; Dawson 2008). The resetting age for the Ubendian Belt, the southern Usagaran Belt and the Rukwa Basin (basement) is likely to be when they were each reworked during the Pan African Orogeny at 570 Ma (Reddy *et al.* 2004; Boniface and Schenk 2012; Boniface *et al.* 2012). The deposition of the Karoo Supergroup sediments within the Rukwa Basin occurred at 260 Ma and forms the accumulation age for the deeper basin infill (Wescott *et al.* 1991; Delvaux *et al.* 1998; Baiyegunhi *et al.* 2014). The volume of helium produced since the nominal resetting age for each of these regions is shown in Table 5 and ranges from a combined sediment and basement helium generation volume in the Rukwa Basin of $0.16 \times 10^{12} \text{ m}^3 \text{ } ^4\text{He}$ (STP) (5.65×10^3 billion cubic ft (BCF)) to $170 \times 10^{12} \text{ m}^3 \text{ } ^4\text{He}$ (STP) (6×10^6 BCF) within the Tanzanian Craton. For context, the global annual industrial use of helium is *c.* $160 \times 10^6 \text{ m}^3 \text{ } ^4\text{He}$ (STP) (5.65 BCF). The combined efficiency of helium release and gas-phase trapping will determine the recoverable helium volume in any one area.

In the absence of any thermal perturbation there is evidence that rocks forming the crystalline basement dominantly lose helium slowly by diffusion (Torgersen 2010; Neretnieks 2013) and, as a result, retain much of their generated helium, on timescales of up to billions of years (Lippmann-Pipke *et al.* 2011; Holland *et al.* 2013; Warr *et al.* 2018). Helium release from each of the different regions in Tanzania requires identification of a mechanism. The development of the EARS is an obvious source of geologically recent and regional thermal input, with surface exhibition of enhanced heat flow shown by Cenozoic volcanic activity and extant geothermal springs. Over 15 geothermal areas have been identified in Tanzania, and summaries of their water and gas chemistry have been published (James 1967a, b; Walker 1969). Five of these occur on the Archean Tanzanian Craton and its Precambrian surrounds, the remaining geothermal centres are found near Quaternary volcanism both in the Tanzanian divergence zone and within the western branch of the EARS. We cannot at this stage identify whether the region has released helium during earlier thermal events.

In Figure 2 we superimpose the major volcanic provinces, geothermal springs and regions over a base map of Tanzania that shows significant sedimentary overburdens, potentially capable of forming stratigraphic or structural traps. This provides a first ‘play fairway’ for Tanzania.

Table 5. Radiogenic ^4He volumes generated to 10 km depth since the last tectonic event within different parts of the Tanzanian system

Region and age of last thermo-tectonic event prior to current rifting	Event	Radiogenic ^4He volume generated (m^3 STP)	Source area (km^2)	References
Tanzanian Craton (2.4 Ga)	Last phase of metamorphism	1.7×10^{13}	350 000	Pinna <i>et al.</i> (1994), Weeraratne <i>et al.</i> (2003)
Ubandian Belt (570 Ma)	Reworking during the Pan-African Orogeny	7.4×10^{11}	75 000	Boniface <i>et al.</i> (2012), Boniface and Schenk (2012)
Southern Usagaran Belt (570 Ma)	Reworking during the Pan-African Orogeny	5.7×10^{11}	57 600	Reddy <i>et al.</i> (2004), Boniface <i>et al.</i> (2012)
North Tanzanian Divergence (2.0 Ga)	Accretion onto the Tanzanian Craton	1.6×10^{12}	42 500	Ebinger <i>et al.</i> (1997), Dawson (2008)
Rukwa Basin (basement) (570 Ma)	Reworking during the Pan-African Orogeny	1.3×10^{11}	12 800	Wescott <i>et al.</i> (1991), Boniface <i>et al.</i> (2012)
Rukwa Basin (sediments) (260 Ma)	Deposition of the Karoo Supergroup	3.3×10^{10}	12 800	Wescott <i>et al.</i> (1991), Delvaux <i>et al.</i> (1998), Baiyegunhi <i>et al.</i> (2014)

Refining the play fairway

It is clear that in any release of helium from the basement formations proximal to volcanic centres risks being diluted by CO_2 associated with volcanic outgassing. This risk is clearly demonstrated at Yellowstone in the USA where a significant flux of ^4He is liberated from the crust by the extant volcanism, but the helium concentration within the dominant CO_2 gas is far too low to be commercially viable (Lowenstern *et al.* 2014). Determining the size of a thermal aureole around a volcanic system, where helium (and likely nitrogen) is liberated from the basement rocks and yet is suitably distal as to reduce the risk of CO_2 dilution (Fig. 1), is still to be determined and beyond the scope of this paper. It is also important to consider the hydrocarbon generation potential of sedimentary systems, as this subsurface gas source may also act to dilute helium concentrations. Nevertheless, both CO_2 and hydrocarbon (CH_4) subsurface gas sources, while diluting any pristine basement helium, may provide a mechanism that either prevents helium from being dissolved entirely in the groundwater or strips helium dissolved in groundwater into the gas phase to form a subsurface gas field (e.g. Ballentine and Lollar 2002). In practice, therefore, this means that helium-prone areas will have sweet spots or Goldilocks zones in which there is neither too much nor too little of these often-associated gases.

Despite uncertainty as to the precise boundaries of the CO_2 dilution/concentration zone, the Albertine Graben north of the Kivu Volcanic Complex in East Africa can be regarded as a lower-priority helium province because of the proximity of three volcanic complexes (Toro Ankole, Virunga and Kivu), each with potential for CO_2 dilution. These are in addition to multiple hydrocarbon seeps containing CO_2 around Lake Albert, and high concentrations of both CH_4 and CO_2 , as well as mantle-derived helium, reported in Lake Kivu (Schoell *et al.* 1988; Tedesco *et al.* 2010; Abeinomuigisha and Kasande 2012).

The Tanganyika rift area appears to be a good prospect due to both its size and distance to current volcanic activity. Helium-rich geothermal springs to the east of the lake at Uvinza produce highly saline fluids and gases that contain 2.5% ^4He . The $^3\text{He}/^4\text{He}$ ratios of the gas are $0.28R_a$, indicating a crust-dominated helium source (Pflumio *et al.* 1994; Kraml *et al.* 2016). $^3\text{He}/^4\text{He}$ are reported as R_a values, which is $^3\text{He}/^4\text{He}$ of the sample normalized to the air $^3\text{He}/^4\text{He}$ of 1.4×10^{-6} (R_a). At the northern end of Lake Tanganyika at Pemba and Cape Banza, fluids contain predominantly magmatic-origin CO_2 (60–90%) and CH_4 with heavier hydrocarbons (Botz and Stoffers 1993; Tiercelin *et al.* 1993). Lake Tanganyika is an area identified for oil exploration with viable trapping structures in the rift basin (Roberts *et al.* 2016). The southern regions of the Tanganyika rift are more distal to current volcanism and may not be experiencing the same thermal perturbation as the northern section. While these observations do not discount the area as a potential helium-rich province, there is an increased CO_2 risk as the Kivu Volcanic Complex is approached and CH_4 dilution risk within the basin depocentres. Regions where the balance of these additional gas sources provide the mechanism to create or maintain a free-gas phase into which the helium partitions may provide prospectivity, but with lower helium concentrations than a primary ^4He and N_2 gas system.

The North Tanzanian Divergence Zone (NTDZ) provides significant interest for investigating helium occurrence. This, one of the youngest sections of the EARS, is an example of extension within the Tanzanian Craton coupled with significant Quaternary volcanic activity in the form of the Crater Highlands Volcanic Complex (Fig. 2). Numerous geothermal springs are reported in both the Eyasi-Wembere Rift and the Manyara Rift (separated by the Mbulu Plateau), which are variably CO_2 rich and helium poor or nitrogen rich and helium rich. The sediment cover is not well

developed but is reported to be up to 2 km thick (James 1967a, b; Macheyeke *et al.* 2008).

The Rukwa Basin is of similar interest; it has recent rifting and is proximal to the Rungwe Volcanic Complex from which there are reports of nitrogen- and helium-rich gas seeps (James 1967a, b). The area is also now known from interpretation of seismic data to have a substantial thickness of sedimentary rocks (up to 7 km) within which seal and candidate reservoir horizons are known to occur (Mulaya *et al.* 2022).

Helium in the NTDZ and Rukwa

Noble gas abundance and isotopes within natural gases can be used to determine the contribution from mantle and crustal sources (e.g. $^3\text{He}/^4\text{He}$), as well as the degree of interaction with the groundwater system (e.g. ^{20}Ne and ^{36}Ar) (e.g. Ballentine *et al.* 1991; Ballentine and Burnard 2002). Four seeps from the NTDZ (Balangida, Gonga, Eyasi and Mponde) and three seeps from the Rukwa Basin (Idindiro, Rukwa and Ivuna) were sampled for noble gas composition and isotope analysis at the University of Oxford, UK (Fig. 3; Tables 6 and 7). The major gas composition of seeps in both regions was first recorded by James (1967a, b) who found that they were predominantly N_2 and ^4He rich. A mixed mantle–meteoric source for the origin of gases from the seeps was originally proposed. In addition, Barry *et al.* (2013) measured low ^4He concentrations and high CO_2 concentrations in sampled thermal

springs from the Ngozi-Songwe hydrothermal system and the Rungwe Volcanic Province, leading to the observation of an increasing crustal signal distal to the active volcanoes in the region.

Helium, neon and argon determinations

In all samples $^4\text{He}/^{20}\text{Ne}$ values are significantly higher than that of air (0.032), and range between 330 and 8920. Air corrections to the $^3\text{He}/^4\text{He}$ ratio of samples, therefore, have no significant effect on the R_a value (ratio of isotopes relative to that in air, which is defined as $R_a = 1$). This indicates that there are negligible air contributions to ^4He concentrations in samples and that $^3\text{He}/^4\text{He}$ ratios that deviate from purely crustal ratios ($0.02R_a$) are due to a mantle contribution.

The He isotope value of samples varies considerably both within and between study areas. Samples from the NTDZ have consistent $^3\text{He}/^4\text{He}$ ratios that range from 0.039 to $0.053R_a$, whereas samples from Mbeya have a larger range of values (0.18 – $3.45R_a$) (Table 6). From the He isotope values alone it is evident that samples from the NTDZ show predominantly crustal R_a values (where $0.020R_a = \text{crust}$: e.g. Ballentine and Burnard 2002), whereas the Rukwa Basin shows a transition towards a more magmatic signature (where $6.10R_a = \text{subcontinental lithospheric mantle (SCLM)}$): Gautheron and Moreira 2002; Day *et al.* 2015).

Concentrations of ^4He in all gases are orders of magnitude greater than the air concentration of 5.4 ppm. Samples from the NTDZ are consistently high in ^4He , with a narrow concentration range of 2.7×10^{-2} – $10.6 \times 10^{-2} \text{ cm}^3$ (STP) $^4\text{He cm}^{-3}$. In contrast, the Rukwa Basin samples vary in ^4He concentrations from 4.3×10^{-5} to $2.5 \times 10^{-2} \text{ cm}^3$ (STP) $^4\text{He cm}^{-3}$ (Table 7). Magmatic contributions to the ^4He concentrations in the NTDZ samples are between 0.3 and 0.5%, assuming a crustal end member of $0.020R_a$ and a SCLM end member of $6.10R_a$, whereas magmatic contributions to ^4He concentrations in the Mbeya study area are significantly higher at 2.6–56.5%.

^{20}Ne can be used as a proxy for the degree of groundwater contact of a gas phase (e.g. Ballentine *et al.* 1991; Ballentine and Sherwood Lollar 2002; Barry *et al.* 2016). Neon concentrations similarly show two distinct patterns. Within all the helium-rich NTDZ samples, neon shows a small range of concentrations from 1.1×10^{-5} to $1.88 \times 10^{-5} \text{ cm}^3$ (STP) ^{20}Ne . In contrast, samples proximal to the Rungwe volcanic zone in the Rukwa Basin have low helium concentrations, and neon concentrations almost two orders of magnitude lower. Even the helium-rich Ivuna seep has a neon concentration intermediate between the concentrations seen in the NTDZ samples (Rukwa Basin samples range from 0.012×10^{-5} to $0.22 \times 10^{-5} \text{ cm}^3$ (STP) ^{20}Ne). Gas seeps in the Rukwa Basin therefore have a higher gas/water ratio than those in the NTDZ, and those closest to the Rungwe volcanic zone the highest gas/water ratio.

Argon isotopes provide similar information to the helium system since subsurface $^{40}\text{Ar}^*$ is derived in the crust from the radioactive decay of ^{40}K , while ^{36}Ar is dominantly from air dissolved in groundwater similar to ^{20}Ne (Ballentine and Burnard 2002). It is therefore no surprise that ^{36}Ar concentrations show a similar pattern to ^{20}Ne , with only a small variance in concentrations between samples from the NTDZ and significantly lower concentrations in the Rukwa samples. The latter show a similar trend in gas/water ratios recorded in the argon as that seen in neon. The greatest risk to the discovery of a commercial helium reservoir remains whether the gas/water ratio is sufficiently high to exceed the groundwater saturation/bubble point at the depth of the structure being considered.

$^{40}\text{Ar}/^{36}\text{Ar}$ ratios are all in excess of the air ratio ($^{40}\text{Ar}/^{36}\text{Ar} = 295.5$), showing that all samples contain a resolvable radiogenic ^{40}Ar component ($^{40}\text{Ar}^*$). The variance of $^{40}\text{Ar}/^{36}\text{Ar}$ in the NTDZ samples is small, between 410 and 544, whereas the Rukwa Basin

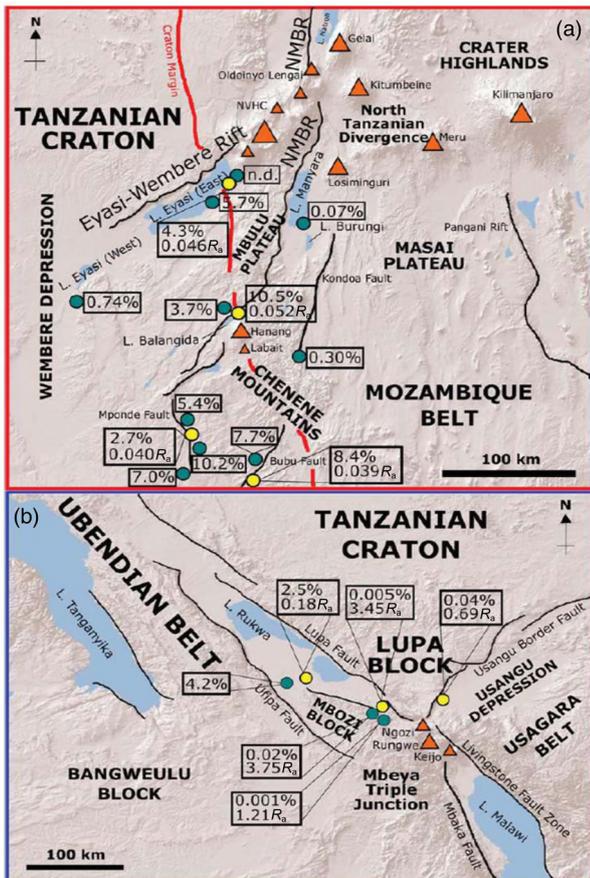


Fig. 3. (a) and (b) The two regions of interest outlined in Figure 2 and the approximate location of the local major tectonic units. Volcanoes are shown by red triangles. Helium concentrations in seeps reported by James (1967a, b) (green circles) are shown as per cent of seep-gas composition. Helium concentrations and $^3\text{He}/^4\text{He}$ ratios (R_a) from this work are also shown (yellow circles). Similar to Figure 1, samples with higher $^3\text{He}/^4\text{He}$ contain only trace levels of helium ($<0.05\%$). Samples with $^3\text{He}/^4\text{He} < 1R_a$ can contain up to 10.5% helium. NMBR = Natron–Manyara–Balangida Rift.

Table 6. Noble gas isotope ratios ($\pm 1\sigma$ errors are shown in brackets)

Sample name	$^3\text{He}/^4\text{He}$ (R_a)	$^{20}\text{Ne}/^{22}\text{Ne}$	$^{21}\text{Ne}/^{22}\text{Ne}$	$^{40}\text{Ar}/^{36}\text{Ar}$	$^{38}\text{Ar}/^{36}\text{Ar}$
Study area 1 (NTD)					
Balangida	0.053 (0.001)	9.74 (0.030)	0.031 (0.0003)	544 (1.2)	0.186 (0.0005)
Balangida	0.052 (0.001)	9.73 (0.030)	0.031 (0.0003)	549 (1.0)	0.186 (0.0005)
Gonga	0.039 (0.001)	9.71 (0.030)	0.029 (0.0003)	432 (1.1)	0.183 (0.0006)
Eyasi	0.046 (0.004)	9.72 (0.030)	0.029 (0.0003)	440 (1.5)	0.187 (0.0006)
Mponde	0.040 (0.002)	9.71 (0.030)	0.030 (0.0004)	410 (0.8)	0.184 (0.0005)
Study area 2 (Mbeya)					
Idindiro	0.69 (0.01)	10.04 (0.033)	0.030 (0.0003)	303 (0.2)	0.187 (0.0003)
Rukwa	3.45 (0.005)	10.04 (0.033)	0.030 (0.0003)	331 (0.9)	0.182 (0.001)
Rukwa	3.45 (0.005)	10.04 (0.033)	0.030 (0.0004)	336 (0.6)	0.184 (0.001)
Ivuna	0.18 (0.01)	9.68 (0.029)	0.032 (0.0004)	787 (0.8)	0.185 (0.0003)
Air (Pepin and Porcelli 2002)	1	9.80 (0.080)	0.029 (0.0003)	295.5 (0.5)	0.188 (0.0004)

samples have a larger range with values of 331–787, with the samples closest to the Rungwe Volcanic Complex showing a $^{40}\text{Ar}/^{36}\text{Ar}$ closest to the air ratio. Qualitatively, these results are consistent with mantle-derived gases in the samples closest to the Rungwe Volcanic Complex, being mostly overprinted by an atmosphere isotopic signature. This is most likely to have been caused by even a small amount of contact between the volcanic gas phase and groundwater containing dissolved atmosphere-derived noble gases.

Binary mixing between crustal and mantle gas end members

It is observed from gas composition analyses that natural gases containing ^4He concentrations, which exceed the economic threshold of 0.1%, also contain high concentrations of N_2 relative to ^4He ($>10:1$) (e.g. Danabalan 2017). In the Rukwa Basin study, gas seeps proximal to the Rungwe Volcanic Province contain the lowest concentration of both N_2 and ^4He , and are dominated by volcanic CO_2 . Higher ^4He and N_2 concentrations, and lower (more radiogenic) $^3\text{He}/^4\text{He}$ (R_a), are observed as the sample localities become more distal from the volcanic system.

Similar spatial patterns of $^3\text{He}/^4\text{He}$ (R_a), N_2 concentrations and CO_2 concentrations as a function of distance from volcanic centres have been observed in other regions (e.g. Fig. 1), and indicate binary mixing between crustal and mantle end members, the latter having likely been derived from the SCLM (Weinlich *et al.* 1999; Barry *et al.* 2013; Karolytè *et al.* 2019).

It is assumed that $^3\text{He}/^4\text{He}$ (R_a) values are primarily controlled by the extent of mixing between crust-derived (N_2 -rich) fluids and associated ^4He , with concentrations of mantle-derived CO_2 and

associated ^3He . In this context the ratio of $^4\text{He}/^{40}\text{Ar}^*$ is correlated with changes in $^3\text{He}/^4\text{He}$. The observed trend is fully consistent with simple two-component mixing between a crustal radiogenic end member with a $^4\text{He}/^{40}\text{Ar}^*$ between 9 and 16, and $^3\text{He}/^4\text{He} = 0.02R_a$, and a mantle end member represented by $^4\text{He}/^{40}\text{Ar}^* = 0.35$ and $^3\text{He}/^4\text{He} = 6.1R_a$ (Fig. 4). While consistent with binary gas mixing between crustal (here dominated by ^4He and N_2) and magmatic gases (dominated by CO_2), we note that the crustal end member resolved in this study is not the ratio at which ^4He and $^{40}\text{Ar}^*$ are produced by radioactivity in the crust (crustal $^4\text{He}/^{40}\text{Ar}^* = 4.9$: Ballentine and Burnard 2002). The data show an enrichment in radiogenic helium relative to argon in the gases associated with the crustal end member. Similar trends have also been observed in the Hugoton–Panhandle giant gas field (Ballentine and Sherwood Lollar 2002). $^4\text{He}/^{40}\text{Ar}^*$ values in crustal fluids that are higher than the crustal production value have been used to argue for evidence of a regional thermal control in which helium is preferentially released from the minerals hosting the radioelements (O’Nions and Ballentine 1993; Ballentine and Burnard 2002; Ballentine and Sherwood Lollar 2002) or the role of regional cross-formational diffusion (Cheng *et al.* 2021).

The Balangida sample from the NTD study area has the highest concentrations of ^4He (at 10.5%) and a low $^3\text{He}/^4\text{He}$ ratio. It also has 89.0% N_2 and negligible CO_2 . It is thus a crustal end member. The mantle end member is taken to be SCLM with $^3\text{He}/^4\text{He} = 6.1R_a$, with CO_2 concentrations of 99.9% and negligible N_2 content. The predominance of the end members is again (e.g. Fig. 1) spatially controlled in the Rukwa region, with the CO_2 -rich samples dominating the fluid composition proximal to the volcanic complex, and N_2 - ^4He dominating the fluid composition distal, up to 135 km, from the volcanic complex.

Table 7. Noble gas and N_2 concentrations ($\pm 1\sigma$ errors are shown in brackets)

Sample name	^4He ($\times 10^{-2}$ cm 3 cm $^{-3}$ STP)	^{20}Ne ($\times 10^{-5}$ cm 3 cm $^{-3}$ STP)	^{40}Ar ($\times 10^{-2}$ cm 3 cm $^{-3}$ STP)	N_2 (cm 3 cm $^{-3}$ STP)
Study area 1 (NTDZ)				
Balangida	10.6 (0.42)	1.19 (0.015)	1.47 (0.042)	0.90
Balangida	10.4 (0.42)	1.17 (0.016)	1.59 (0.021)	0.90
Gonga	8.4 (0.35)	1.88 (0.033)	1.69 (0.043)	0.95
Eyasi	4.3 (0.29)	1.30 (0.016)	1.21 (0.020)	0.95
Mponde	2.7 (0.11)	1.10 (0.019)	1.11 (0.019)	
Study area 2 (Mbeya)				
Idindiro	0.04 (0.002)	0.028 (0.0004)	0.73 (0.017)	
Rukwa	0.0047 (0.0002)	0.014 (0.0004)	0.029 (0.0008)	
Rukwa	0.0043 (0.0002)	0.012 (0.0003)	0.025 (0.0006)	
Ivuna	2.5 (0.04)	0.22 (0.0011)	0.46 (0.002)	0.96
Air (Pepin and Porcelli 2002)	0.000524 (0.000006)	1.65 (0.0036)	0.93 (0.001)	0.78

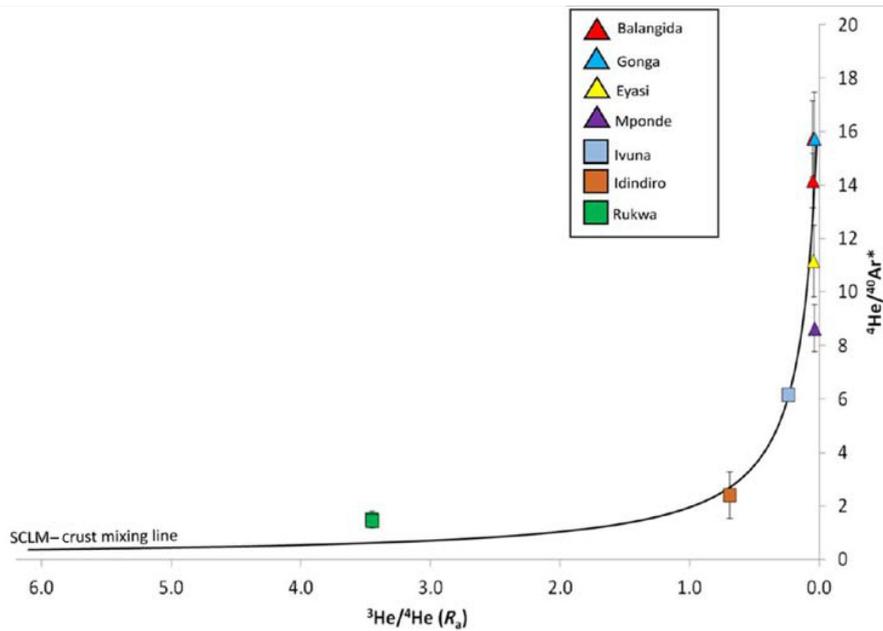


Fig. 4. $^3\text{He}/^4\text{He}$ (R_a) plotted against $^4\text{He}/^{40}\text{Ar}^*$ shows simple binary mixing between two gas sources ($^{40}\text{Ar}^*$ is the resolved radiogenic $^{40}\text{Ar}^*$, see the text). All samples fall on a mixing line between a magmatic gas sourced from the subcontinental lithospheric mantle (SCLM) (with $^3\text{He}/^4\text{He}$ *c.* $6R_a$ and $^4\text{He}/^{40}\text{Ar}^* \leq 1.0$) and a helium/argon source derived from the continental crust (with $^3\text{He}/^4\text{He}$ *c.* 0.02 and $^4\text{He}/^{40}\text{Ar}^* = 9\text{--}16$). Helium-rich ($>1.0\%$) gas samples have $^3\text{He}/^4\text{He} < 0.5R_a$ and $^4\text{He}/^{40}\text{Ar}^* > 8$.

Exploration update

Helium One completed two exploration wells at Rukwa in August of 2021 (Tai 1 and Tai 2). Both demonstrated the presence of thick well-developed seals and reservoir intervals in the Lake Beds with helium shows. The next phase of drilling will target the Red Sandstone reservoir interval (Helium One Global 2021).

Conclusions

We have detailed the principles of helium generation, primary release, secondary migration and considerations of the role of the groundwater system that allow for a $^4\text{He}\text{--}\text{N}_2$ gas phase to develop, with or without volcanic CO_2 or sedimentary CH_4 , and thus form economically viable helium exploration provinces. Geological regions that contain old (billions of years) basement rocks will have accumulated substantial ^4He through the radiogenic decay of U+Th. Within regions of recent tectonism causing geothermal perturbation, stored helium will be released along with associated gases such as nitrogen. Consideration of the migration of either a buoyant $^4\text{He}\text{--}\text{N}_2$ free-gas phase, or $^4\text{He}\text{--}\text{N}_2$ dissolved in the groundwater system and its subsequent exsolution, have been detailed together with the need for the presence of suitable trapping structures. These components enable the formation of a ‘play fairway’ approach to developing an exploration strategy; regions that exhibit all of these characteristics have the prospect of delivering commercially viable helium resources.

We show that sections of the Tanzanian East African Rift contain most, if not all, of the prerequisites for an economic helium province. Noble gas data from gas seeps in the west and east branches of the Tanzanian section of the East African Rift System contain ^4He concentrations of up to 10.5%. This, combined with the potential ^4He generated by the Tanzanian Craton and surrounding mobile belts of *c.* 7.0×10^5 BCF (2×10^{13} m^3 ^4He (STP)), implies that even with inefficient release, migration and trapping, these regions could provide high-helium concentration reservoirs. In the region of the Rukwa Basin independent prospective resource estimates undertaken on behalf of the operator and their financial backers suggest that there is a P_{50} (best estimate) of 138 BCF ^4He (2.78×10^9 m^3 ^4He (STP)) potentially trapped within existing trapping structures (Helium One Global 2020), which, if recoverable, would alone supply the current world consumption for 14 years.

Acknowledgements The authors thank the three reviewers who helped us improve the paper substantially.

Author contributions DD: data curation (lead), formal analysis (lead), investigation (lead), methodology (lead), resources (equal), validation (equal), visualization (equal), writing – original draft (equal), writing – review & editing (supporting); JGG: conceptualization (lead), formal analysis (equal), funding acquisition (lead), investigation (equal), methodology (equal), project administration (equal), resources (equal), supervision (lead), validation (equal), visualization (equal), writing – original draft (equal), writing – review & editing (equal); CGM: conceptualization (supporting), formal analysis (supporting), supervision (equal); THA-J: funding acquisition (supporting), investigation (supporting), methodology (supporting), writing – review & editing (supporting); JJB: funding acquisition (supporting), resources (supporting), validation (supporting), writing – review & editing (supporting); PHB: resources (equal), supervision (supporting), validation (supporting), writing – original draft (equal), writing – review & editing (supporting); CJB: conceptualization (lead), formal analysis (equal), funding acquisition (equal), investigation (equal), methodology (equal), project administration (equal), resources (equal), supervision (lead), validation (equal), visualization (equal), writing – original draft (lead), writing – review & editing (lead).

Funding The PhD study was funded by Statoil (renamed Equinor), Norway. Fieldwork in Tanzania and analyses of the gases sampled in Tanzania were funded by Helium One Global Limited.

Competing interests D. Danabalan, J.G. Gluyas, C.G. Macpherson and C.J. Ballentine received research funding for this work from Statoil (now Equinor). J.J. Bluet and T.H. Abraham-James were founders of Helium One. C.J. Ballentine and P.H. Barry received funds to conduct fieldwork in Tanzania from Helium One.

Data availability All data are included in the published article.

References

- Abeinomugisha, D. and Kasande, R. 2012. Tectonic control on hydrocarbon accumulation in the intra-continental Albertine Graben of the East African Rift System. *AAPG Memoirs*, **100**, 209–228, <https://doi.org/10.1306/13351554M1003539>
- Adams, J.A.S., Osmond, J.K. and Rodgers, J.J.W. 1959. The geochemistry of uranium and thorium. *Physics and Chemistry of the Earth*, **3**, 298–348, [https://doi.org/10.1016/0079-1946\(59\)90008-4](https://doi.org/10.1016/0079-1946(59)90008-4)
- American Physical Society, Materials Research Society and American Chemical Society 2016. *Responding to the U.S. Research Community’s Liquid Helium Crisis: An Action Plan to Preserve U.S. Innovation*. A Science Policy Report issued by American Physical Society, Materials Research Society, American Chemical Society, <https://www.aps.org/policy/reports/popa-reports/upload/HeliumReport.pdf>

- Bagdasaryan, G.P., Gerasimovskiy, V.I., Polyakov, A.I. and Gukasyan, R.K. 1973. Age of volcanic rocks in the rift zones of East Africa. *Geochemistry International*, **10**, 66e71.
- Bähr, R., Lippolt, H.J. and Wemicke, R.S. 1994. Temperature-induced ⁴He degassing of specularite and botryoidal hematite: A ⁴He retentivity study. *Journal of Geophysical Research: Solid Earth*, **99**, 17 695–17 707, <https://doi.org/10.1029/94JB01055>
- Baiyegunhi, C., Oloniniyi, T.L. and Gwavava, O. 2014. The correlation of dry density and porosity of some rocks from the Karoo Supergroup: A case study of selected rock types between Grahamstown and Queenstown in the Eastern Cape Province, South Africa. *IOSR Journal of Engineering*, **4**, 30–40, <https://doi.org/10.9790/3021-041213040>
- Ballentine, C. 2017. Helium in crisis. *Chemistry World*, 25 April, <https://www.chemistryworld.com/opinion/helium-in-crisis/3007152.article>
- Ballentine, C.J. and Burnard, P.G. 2002. Production, release and transport of noble gases in the continental crust. *Reviews in Mineralogy and Geochemistry*, **47**, 481–538, <https://doi.org/10.2138/rmg.2002.47.12>
- Ballentine, C.J. and Sherwood Lollar, B. 2002. Regional groundwater focusing of nitrogen and noble gases into the Hugoton–Panhandle giant gas field, USA. *Geochimica et Cosmochimica Acta*, **66**, 2483–2497, [https://doi.org/10.1016/S0016-7037\(02\)00850-5](https://doi.org/10.1016/S0016-7037(02)00850-5)
- Ballentine, C.J., O’Nions, R.K., Oxburgh, E.R., Horvath, F. and Deak, J. 1991. Rare gas constraints on hydrocarbon accumulation, crustal degassing and groundwater flow in the Pannonian Basin. *Earth and Planetary Science Letters*, **105**, 229–246, [https://doi.org/10.1016/0012-821X\(91\)90133-3](https://doi.org/10.1016/0012-821X(91)90133-3)
- Barry, P.H., Hilton, D.R., Fischer, T.P., De Moor, J.M., Mangasini, F. and Ramirez, C. 2013. Helium and carbon isotope systematics of cold ‘mazuku’ CO₂ vents and hydrothermal gases and fluids from Rungwe Volcanic Province, southern Tanzania. *Chemical Geology*, **339**, 141–156, <https://doi.org/10.1016/j.chemgeo.2012.07.003>
- Barry, P.H., Hilton, D.R. *et al.* 2015. Helium isotopic evidence for modification of the cratonic lithosphere during the Permo-Triassic Siberian flood basalt event. *Lithos*, **216**, 73–80, <https://doi.org/10.1016/j.lithos.2014.12.001>
- Barry, P.H., Lawson, M., Meurer, W.P., Warr, O., Mabry, J.C., Byrne, D.J. and Ballentine, C.J. 2016. Noble gases solubility models of hydrocarbon charge mechanism in the Sleipner Vest gas field. *Geochimica et Cosmochimica Acta*, **194**, 291–309, <https://doi.org/10.1016/j.gca.2016.08.021>
- Barry, P.H., Lawson, M., Meurer, W.P., Danabalan, D., Byrne, D.J., Mabry, J.C. and Ballentine, C.J. 2017. Determining fluid migration and isolation times in multiphase crustal domains using noble gases. *Geology*, **45**, 775–778, <https://doi.org/10.1130/G38900.1>
- Boniface, N. and Schenk, V. 2012. Neoproterozoic eclogites in the Paleoproterozoic Ubendian Belt of Tanzania: Evidence for a Pan-African suture between the Bangweulu Block and the Tanzania Craton. *Precambrian Research*, **208–211**, 72–89, <https://doi.org/10.1016/j.precamres.2012.03.014>
- Boniface, N., Schenk, V. and Appel, P. 2012. Paleoproterozoic eclogites of MORB-type chemistry and three Proterozoic orogenic cycles in the Ubendian Belt (Tanzania): Evidence from monazite and zircon geochronology, and geochemistry. *Precambrian Research*, **192–195**, 16–33, <https://doi.org/10.1016/j.precamres.2011.10.007>
- Bottomley, D.J., Ross, J.D. and Clarke, W.B. 1984. Helium and neon isotope geochemistry of some ground waters from the Canadian Precambrian Shield. *Geochimica et Cosmochimica Acta*, **48**, 1973–1985, [https://doi.org/10.1016/0016-7037\(84\)90379-X](https://doi.org/10.1016/0016-7037(84)90379-X)
- Botz, R.W. and Stoffers, P. 1993. Light hydrocarbon gases in Lake Tanganyika hydrothermal fluids (east-central Africa). *Chemical geology*, **104**, 217–224, [https://doi.org/10.1016/0009-2541\(93\)90152-9](https://doi.org/10.1016/0009-2541(93)90152-9)
- Boyce, J.W., Hodges, K.V., Olszewski, W.J. and Jercinovic, M.J. 2005. He diffusion in monazite: Implications for (U–Th)/He thermochronometry. *Geochemistry, Geophysics, Geosystems*, **6**, Q12004, <https://doi.org/10.1029/2005GC001058>
- Broadhead, R.F. 2005. Helium in New Mexico – geologic distribution, resource demand, and exploration possibilities. *New Mexico Geology*, **27**, 93–101.
- Broadhead, R.F. and Gillard, L. 2004. *Helium in New Mexico: Geologic Distribution and Exploration Possibilities*. New Mexico Bureau of Geology and Mineral Resources Open File Report 483.
- Burwash, R.A. and Cumming, G.L. 1976. Uranium and thorium in the Precambrian basement of western Canada. I. Abundance and distribution. *Canadian Journal of Earth Sciences*, **13**, 284–293, <https://doi.org/10.1139/e76-030>
- Chaki, S., Takarli, M. and Agbodjan, W.P. 2008. Influence of thermal damage on physical properties of a granite rock: porosity, permeability and ultrasonic wave evolutions. *Construction and Building Materials*, **22**, 1456–1461, <https://doi.org/10.1016/j.conbuildmat.2007.04.002>
- Cheng, A., Sherwood Lollar, B., Giunta, T.M., Mundle, S.O.C. and Ballentine, C.J. 2018. Helium distribution in the Williston and the Southwest Ontario basins. *Goldschmidt Abstracts*, **2018**, 403, <https://goldschmidtabstracts.info/abstracts/abstractView?id=2018003772> [accessed 29 December 2020].
- Cheng, A., Sherwood Lollar, B. *et al.* 2021. Determining the role of diffusion and basement flux in controlling ⁴He distribution in sedimentary basin fluids. *Earth and Planetary Science Letters*, **574**, 117175, <https://doi.org/10.1016/j.epsl.2021.117175>
- Chemiai, D.J., Watson, E.B. and Thomas, J.B. 2009. Diffusion of helium in zircon and apatite. *Chemical Geology*, **268**, 155–166, <https://doi.org/10.1016/j.chemgeo.2009.08.011>
- Clarke, R.H., Nuttall, W.J. and Glowacki, B.A. 2012. Introduction. *In*: Nuttall, W.J., Clarke, R.H. and Glowacki, B.A. (eds) *The Future of Helium as a Natural Resource*. Routledge, Abingdon, UK, 1–4.
- Connor, S. 2013. A ballooning problem: the great helium shortage. *The Independent*, 4 January, <https://www.independent.co.uk/news/science/a-ballooning-problem-the-great-helium-shortage-8439108.html>
- Danabalan, D. 2017. *Helium: Exploration Methodology for a Strategic Resource*. PhD thesis, Durham University, Durham, UK.
- Dawson, J.B. 2008. *The Gregory Rift Valley and Neogene–Recent Volcanoes of Northern Tanzania*. Geological Society, London, Memoirs, **33**, <https://doi.org/10.1144/M33.0>
- Day, J.M., Barry, P.H., Hilton, D.R., Burgess, R., Pearson, D.G. and Taylor, L.A. 2015. The helium flux from the continents and ubiquity of low-³He/⁴He recycled crust and lithosphere. *Geochimica et Cosmochimica Acta*, **153**, 116–133, <https://doi.org/10.1016/j.gca.2015.01.008>
- De Bruin, R.H. 1995. Helium resources of Wyoming. *In*: *Resources of Southwest Wyoming: 46th Field Conference Guidebook*. Wyoming Geological Association, Casper, WY, 191–201.
- Delvaux, D., Levi, K., Kajara, R. and Sarota, J. 1992. Cenozoic paleostress and kinematic evolution of the Rukwa–North Malawi rift valley (East African Rift System). *Bulletin des Centres de Recherche Exploration–Production Elf-Aquitaine*, **16**, 383–406.
- Delvaux, D., Kervyn, F., Vittori, E., Kajara, R.S.A. and Kilembe, E. 1998. Late Quaternary tectonic activity and lake level change in the Rukwa Rift Basin. *Journal of African Earth Sciences*, **26**, 397–421, [https://doi.org/10.1016/S0899-5362\(98\)00023-2](https://doi.org/10.1016/S0899-5362(98)00023-2)
- Dobbin, C.E. 1968. Geology of natural gases rich in helium, nitrogen, carbon dioxide, and hydrogen sulfide. *AAPG Memoirs*, **9**, 1957–1969, <https://doi.org/10.1306/M9363C134>
- Dunai, T.J. and Roselieb, K. 1996. Sorption and diffusion of helium in garnet: implications for volatile tracing and dating. *Earth and Planetary Science Letters*, **139**, 411–421, [https://doi.org/10.1016/0012-821X\(96\)00029-5](https://doi.org/10.1016/0012-821X(96)00029-5)
- Ebinger, C.J., Deino, A.L., Tesha, A.L., Becker, T. and Ring, U. 1993. Tectonic controls on rift basin geometry: Evolution of the northern Malawi (Nyasa) Rift. *Journal of Geophysical Research: Solid Earth*, **98**, 17 821–17 836, <https://doi.org/10.1029/93JB01392>
- Ebinger, C., Djomani, Y.P., Mbede, E., Foster, A. and Dawson, J.B. 1997. Rifting Archaean lithosphere: the Eyasi–Manyara–Natron rifts, East Africa. *Journal of the Geological Society, London*, **154**, 947–960, <https://doi.org/10.1144/gsjgs.154.6.0947>
- Ebinger, C.J., van Wijk, J. and Keir, D. 2013. The time scales of continental rifting: Implications for global processes. *Geological Society of America Special Papers*, **500**, 371–396, [https://doi.org/10.1130/2013.2500\(11\)](https://doi.org/10.1130/2013.2500(11))
- Farley, K.A. 2000. Helium diffusion from apatite: General behavior as illustrated by Durango fluorapatite. *Journal of Geophysical Research: Solid Earth*, **105**, 2903–2914, <https://doi.org/10.1029/1999JB900348>
- Farley, K.A. 2002. (U–Th)/He dating: Techniques, calibrations, and applications. *Reviews in Mineralogy and Geochemistry*, **47**, 819–844, <https://doi.org/10.2138/rmg.2002.47.18>
- Foster, A., Ebinger, C., Mbede, E. and Rex, D. 1997. Tectonic development of the northern Tanzanian sector of the East African Rift System. *Journal of the Geological Society, London*, **154**, 689–700, <https://doi.org/10.1144/gsjgs.154.4.0689>
- Fritz, H., Tenczer, V., Hauzenberger, C.A., Wallbrecher, E., Hoinkes, G., Muhongo, S. and Mogessie, A. 2005. Central Tanzanian tectonic map: a step forward to decipher Proterozoic structural events in the East African Orogen. *Tectonics*, **24**, TC6013, <https://doi.org/10.1029/2005TC001796>
- Gage, B.D. and Driskill, D.L. 2005. *Analyses of Natural Gases, 2002–2004*. United States Department of the Interior Bureau of Land Management Technical Note 418, https://www.blm.gov/sites/blm.gov/files/documents/files/Library_BLMTechnicalNote418.pdf
- Gautheron, C. and Moreira, M. 2002. Helium signature of the subcontinental lithospheric mantle. *Earth and Planetary Science Letters*, **199**, 39–47, [https://doi.org/10.1016/S0012-821X\(02\)00563-0](https://doi.org/10.1016/S0012-821X(02)00563-0)
- Gerling, C.R. 1983. McElmo Dome Leadville carbon dioxide field. *In*: *Colorado in Oil and Gas Fields of the Four Corners Area, Volume III*. Four Corners Geological Society, Durango, CO, 735–739.
- Getachew, E., van der Meijde, M., Nyblade, A.A. and van der Meer, F.D. 2011. A crustal thickness map of Africa derived from a global gravity field model using Euler deconvolution. *Geophysical Journal International*, **187**, 1–9, <https://doi.org/10.1111/j.1365-246X.2011.05140.x>
- Gilfillan, S. M. 2006. *Deep Magmatic Degassing and the Colorado Plateau Uplift*. PhD thesis, University of Manchester, Manchester, UK.
- Gilfillan, S.M., Ballentine, C.J. *et al.* 2008. The noble gas geochemistry of natural CO₂ gas reservoirs from the Colorado Plateau and Rocky Mountain provinces, USA. *Geochimica et Cosmochimica Acta*, **72**, 1174–1198, <https://doi.org/10.1016/j.gca.2007.10.009>
- Gluyas, J. 2019a. The emergence of the helium industry: the history of helium exploration, part 1 of 2. *AAPG Explorer*, **2019**, January, 16–17, <https://explorer.aapg.org/story/articleid/50466/the-emergence-of-the-helium-industry>
- Gluyas, J. 2019b. Helium shortages and emerging helium provinces: the history of helium exploration, part 2. *AAPG Explorer*, **2019**, February, 18–22, <https://explorer.aapg.org/story/articleid/51290/helium-shortages-and-emerging-helium-provinces>

- Gold, T. and Held, M. 1987. Helium–nitrogen–methane systematics in natural gases of Texas and Kansas. *Journal of Petroleum Geology*, **10**, 415–424, <https://doi.org/10.1111/j.1747-5457.1987.tb00582.x>
- Hamak, J.E. 1989. Helium resources of Wyoming. In: *Gas Resources of Wyoming: 40th Field Conference Guidebook*. Wyoming Geological Association, Casper, WY, 117–121.
- Harris, J.E. 1993. *Woodside: T. 18-20 S., R. 13-14 E., SLPM Emery County, Utah in Oil and Gas Fields of Utah*. Utah Geological Association, Salt Lake City, UT.
- Heikal, M.T.S., El-Dosuky, B.T., Ghoneim, M.F. and Sherif, M.I. 2013. Natural radioactivity in basement rocks and stream sediments, Sharm El Sheikh Area, South Sinai, Egypt: radiometric levels and their significant contributions. *Arabian Journal of Geosciences*, **6**, 3229–3239, <https://doi.org/10.1007/s12517-012-0622-6>
- Helium One Global 2020. Placing and Subscription of 211 267 597 new Ordinary Shares of no par value at 2.84 pence. Helium One Global Limited, Road Town, Tortola, British Virgin Islands, <http://www.helium-one.com/wp-content/uploads/2020/11/259989-Project-Apollo-CLN-reduced-memory-Final-13.11.20.pdf> [accessed 1 January 2021].
- Helium One Global 2021. Helium One News & Updates. Helium One Global Limited, Road Town, Tortola, British Virgin Islands, <http://www.helium-one.com/media/>
- Hiyagon, H. and Kennedy, B.M. 1992. Noble gases in CH₄-rich gas fields, Alberta, Canada. *Geochimica et Cosmochimica Acta*, **56**, 1569–1589, [https://doi.org/10.1016/0016-7037\(92\)90226-9](https://doi.org/10.1016/0016-7037(92)90226-9)
- Holland, G., Sherwood Lollar, B., Li, L., Lacrampe-Couloume, G., Slater, G.F. and Ballentine, C.J. 2013. Deep fracture fluids isolated in the crust since the Precambrian era. *Nature*, **497**, 357, <https://doi.org/10.1038/nature12127>
- Hussain, N. 1997. Flux of ⁴He from Cammenellis granite: modelling of an HDR geothermal reservoir. *Applied Geochemistry*, **12**, 1–8, [https://doi.org/10.1016/S0883-2927\(96\)00038-8](https://doi.org/10.1016/S0883-2927(96)00038-8)
- Hutcheon, I. 1999. Controls on the distribution of non-hydrocarbon gases in the Alberta Basin. *Bulletin of Canadian Petroleum Geology*, **47**, 573–593.
- James, T.C. 1967a. Thermal springs in Tanzania. *Institution of Mining and Metallurgy, Transactions/Section B (Applied Earth Science)*, **76**, B1–B18.
- James, T.C. 1967b. Thermal springs in Tanzania – discussions and conclusions. *Institution of Mining and Metallurgy, Transactions/Section B (Applied Earth Science)*, **76**, B168–B174.
- Jenden, P.D. and Kaplan, I.R. 1989. Origin of natural-gas in Sacramento Basin, California. *AAPG Bulletin*, **73**, 431–453.
- Jenden, P.D., Kaplan, I.R., Poreda, R.J. and Craig, H. 1988a. Origin of nitrogen rich gases in the Californian Great Valley: Evidence from helium, carbon and nitrogen isotope ratios. *Geochimica et Cosmochimica Acta*, **52**, 851–861, [https://doi.org/10.1016/0016-7037\(88\)90356-0](https://doi.org/10.1016/0016-7037(88)90356-0)
- Jenden, P.D., Newell, K.D., Kaplan, I.R. and Watney, W.L. 1988b. Composition and stable isotope geochemistry of natural gases from Kansas, Midcontinent, USA. *Chemical Geology*, **71**, 117–147, [https://doi.org/10.1016/0009-2541\(88\)90110-6](https://doi.org/10.1016/0009-2541(88)90110-6)
- Kalin, S. and Finn, T. 2017. Qatar closes helium plants amid rift with Arab powers. *Reuters*, 13 June, <https://www.reuters.com/article/us-gulf-qatar-helium-idUSKBN19426X>
- Karolytė, R., Johnson, G. *et al.* 2019. Tracing the migration of mantle CO₂ in gas fields and mineral water springs in south-east Australia using noble gas and stable isotopes. *Geochimica et Cosmochimica Acta*, **259**, 109–128, <https://doi.org/10.1016/j.gca.2019.06.002>
- Kraml, M., Kaudse, T. and Aeschbach, W. and Tanzanian Exploration Team 2016. The search for volcanic heat sources in Tanzania: a helium isotope perspective. Proceedings of the 6th African Rift Geothermal Conference, 2–4 November 2016, Addis Ababa, Ethiopia. 1–13pp.
- Lenoir, J.L., Liégeois, J.P., Theunissen, K. and Klerkx, J. 1994. The Palaeoproterozoic Ubendian shear belt in Tanzania: geochronology and structure. *Journal of African Earth Sciences*, **19**, 169–184, [https://doi.org/10.1016/0899-5362\(94\)90059-0](https://doi.org/10.1016/0899-5362(94)90059-0)
- Lippmann-Pipke, J., Sherwood Lollar, B., Niedermann, S., Stronck, N.A., Naumann, R., van Heerden, E. and Onstott, T.C. 2011. Neon identifies two billion year old fluid component in Kaapvaal Craton. *Chemical Geology*, **283**, 287–296, <https://doi.org/10.1016/j.chemgeo.2011.01.028>
- Lippolt, H.J., Leitz, M., Wernicke, R.S. and Hagedorn, B. 1994. (Uranium +thorium)/helium dating of apatite: experience with samples from different geochemical environments. *Chemical Geology*, **112**, 179–191, [https://doi.org/10.1016/0009-2541\(94\)90113-9](https://doi.org/10.1016/0009-2541(94)90113-9)
- Lowenstern, J.B., Evans, W.C., Bergfeld, D. and Hunt, A.G. 2014. Prodigious degassing of a billion years of accumulated radiogenic helium at Yellowstone. *Nature*, **506**, 355–358, <https://doi.org/10.1038/nature12992>
- Lowenstern, J.B., Bergfeld, D., Evans, W.C. and Hunt, A.G. 2015. Origins of geothermal gases at Yellowstone. *Journal of Volcanology and Geothermal Research*, **302**, 87–101, <https://doi.org/10.1016/j.jvolgeores.2015.06.010>
- Macheyeki, A.S., Delvaux, D., De Batist, M. and Mruma, A. 2008. Fault kinematics and tectonic stress in the seismically active Manyara–Dodoma Rift segment in Central Tanzania – Implications for the East African Rift. *Journal of African Earth Sciences*, **51**, 163–188, <https://doi.org/10.1016/j.jafrearsci.2008.01.007>
- Martel, D.J., O’Nions, R.K., Hilton, D.R. and Oxburgh, E.R. 1990. The role of element distribution in production and release of radiogenic helium: The Cammenellis Granite, southwest England. *Chemical Geology*, **88**, 207–221, [https://doi.org/10.1016/0009-2541\(90\)90090-T](https://doi.org/10.1016/0009-2541(90)90090-T)
- Martin, T.G., Smith, S.N. and Bondos, J. 2008. Materials and corrosion history with Labarge Madison Production: A 20 year story of success. Paper NACE-08634 presented at CORROSION 2008, 16–20 March 2008, New Orleans, Louisiana, USA.
- McKenzie, D. 1978. Some remarks on the development of sedimentary basins. *Earth and Planetary Science Letters*, **40**, 25–32, [https://doi.org/10.1016/0012-821X\(78\)90071-7](https://doi.org/10.1016/0012-821X(78)90071-7)
- Merrill, M.D., Hunt, A.G. and Lohr, C.D. 2014. Noble gas geochemistry investigation of high CO₂ natural gas at the LaBarge Platform, Wyoming, USA. *Energy Procedia*, **63**, 4186–4190, <https://doi.org/10.1016/j.egypro.2014.11.451>
- Meunier, J.D., Sellier, E. and Pagal, M. 1990. Radiation-damage rims in quartz from uranium-bearing sandstones. *Journal of Sedimentary Research*, **60**, 53–58.
- Morgan, C.D. and Chidsey, T.C., Jr 1991. Gordon Creek, Farnham Dome, and Woodside fields, carbon and Emery Counties, Utah. In: Chidsey, T.C., Jr (ed.) *Geology of East-Central Utah*. Utah Geological Association, Salt Lake City, UT, 301–310.
- Mruma, A.H. 1995. Stratigraphy and palaeodepositional environment of the Palaeoproterozoic volcano-sedimentary Konse Group in Tanzania. *Journal of African Earth Sciences*, **21**, 281–290, [https://doi.org/10.1016/0899-5362\(95\)00065-2](https://doi.org/10.1016/0899-5362(95)00065-2)
- Muhongo, S. and Lenoir, J.L. 1994. Pan-African granulite-facies metamorphism in the Mozambique Belt of Tanzania: U–Pb zircon geochronology. *Journal of the Geological Society, London*, **151**, 343–347, <https://doi.org/10.1144/gsjgs.151.2.0343>
- Mulaya, E.S., Gluyas, J.G., McCaffrey, K.J.W., Phillips, T.B. and Ballentine, D.J. 2022. Structural geometry and evolution of the Rukwa Rift Basin, Tanzania: Implications for helium potential. *Basin Research*, <https://doi.org/10.1111/bre.12646>
- Murphy, H. 2019. The global helium shortage is real but don’t blame party balloons. *The New York Times*, 16 May, <https://www.nytimes.com/2019/05/16/science/helium-shortage-party-city.html> [accessed 14 August 2019].
- Neretnieks, I. 2013. Some aspects of release and transport of gases in deep granitic rocks: possible implications for nuclear waste repositories. *Hydrology Journal*, **21**, 1701–1716, <https://doi.org/10.1007/s10040-013-0986-z>
- Nyblade, A.A. and Brazier, R.A. 2002. Precambrian lithospheric controls on the development of the East African rift system. *Geology*, **30**, 755–758, [https://doi.org/10.1130/0091-7613\(2002\)030<0755:PLCOTD>2.0.CO;2](https://doi.org/10.1130/0091-7613(2002)030<0755:PLCOTD>2.0.CO;2)
- O’Nions, R.K. and Ballentine, C.J. 1993. Rare gas studies of basin scale fluid movement. *Philosophical Transactions of the Royal Society A: Mathematical, Physical and Engineering Sciences*, **344**, 141–156, <https://doi.org/10.1098/rsta.1993.0082>
- Pepin, R.O. and Porcelli, D. 2002. Origin of noble gases in the terrestrial planets. *Reviews in Mineralogy and Geochemistry*, **47**, 191–246, <https://doi.org/10.2138/rmg.2002.47.7>
- Pflumio, C., Boulègue, J. and Tiercelin, J.J. 1994. Hydrothermal activity in the northern Tanganyika rift, East Africa. *Chemical Geology*, **116**, 85–109, [https://doi.org/10.1016/0009-2541\(94\)90159-7](https://doi.org/10.1016/0009-2541(94)90159-7)
- Pierce, A.P., Gott, G.B. and Mytton, J.W. 1964. *Uranium and Helium in the Panhandle Gas Field, Texas, and Adjacent Areas*. United States Geological Survey Professional Papers, **454-G**.
- Pinna, P., Calvez, J.Y., Abessolo, A., Angel, J.M., Mekoulou-Mekoulou, T., Mananga, G. and Vernhet, Y. 1994. Neoproterozoic events in the Tcholliré area: Pan-African crustal growth and geodynamics in central-northern Cameroon (Adamawa and North Provinces). *Journal of African Earth Sciences*, **18**, 347–353, [https://doi.org/10.1016/0899-5362\(94\)90074-4](https://doi.org/10.1016/0899-5362(94)90074-4)
- Poreda, R., Jenden, P.D., Kaplan, I.R. and Craig, H. 1986. Mantle helium in Sacramento Basin natural gas wells. *Geochimica et Cosmochimica Acta*, **50**, 9–33, [https://doi.org/10.1016/0016-7037\(86\)90231-0](https://doi.org/10.1016/0016-7037(86)90231-0)
- Quennell, A.M. 1956. The Bukoban System of East Africa. In: *Report of the International Geological Congress, 20th Session, Mexico*, International Union of Geological Sciences, Beijing, China, 281–307.
- Reddy, S.M., Collins, A.S., Buchan, C. and Mruma, A.H. 2004. Heterogeneous excess argon and Neoproterozoic heating in the Usagaran Orogen, Tanzania, revealed by single grain ⁴⁰Ar/³⁹Ar thermochronology. *Journal of African Earth Sciences*, **39**, 165–176, <https://doi.org/10.1016/j.jafrearsci.2004.07.052>
- Reich, M., Ewing, R.C., Ehlers, T.A. and Becker, U. 2007. Low-temperature anisotropic diffusion of helium in zircon: implications for zircon (U–Th)/He thermochronometry. *Geochimica et Cosmochimica Acta*, **71**, 3119–3130, <https://doi.org/10.1016/j.gca.2007.03.033>
- Reimer, G.M. 1976. *Helium Detection as a Guide for Uranium Exploration*. United States Geological Survey Open File Report, **76-240**.
- Reiners, P.W. 2005. Zircon (U–Th)/He thermochronometry. *Reviews in Mineralogy and Geochemistry*, **58**, 151–179, <https://doi.org/10.2138/rmg.2005.58.6>
- Reiners, P.W. and Farley, K.A. 1999. Helium diffusion and (U–Th)/He thermochronometry of titanite. *Geochimica et Cosmochimica Acta*, **63**, 3845–3859, [https://doi.org/10.1016/S0016-7037\(99\)00170-2](https://doi.org/10.1016/S0016-7037(99)00170-2)
- René, M. 2017. Nature, sources, resources and production of thorium. In: Akitsu, T. (ed.) *Descriptive Inorganic Chemistry Researches of Metal Compounds*. IntechOpen, London, <https://doi.org/10.5772/intechopen.68304>
- Research and Markets 2020. The Future of the Helium Industry, 2020–2030; Projected to Reach \$15.73 Billion by 2023. *Global Newswire*, 14 February, <https://www.globenewswire.com/news-release/2020/02/14/19851370/en/The-Future-of-the-Helium-Industry-2020-2030-Projected-to-Rreach-15-73-Billion-by-2023.html> [accessed 29 December 2020].

- Roberts, E.M., O'Connor, P.M., Gottfried, M.D., Stevens, N., Kapalima, S. and Ngasala, S. 2004. Revised stratigraphy and age of the Red Sandstone Group in the Rukwa Rift Basin, Tanzania. *Cretaceous Research*, **25**, 749–759, <https://doi.org/10.1016/j.cretres.2004.06.007>
- Roberts, E.M., O'Connor, P.M. *et al.* 2012a. Sedimentology and depositional environments of the Red Sandstone Group, Rukwa Rift Basin, southwestern Tanzania: New insight into Cretaceous and Paleogene terrestrial ecosystems and tectonics in sub-equatorial Africa. *Journal of African Geosciences*, **57**, 179–212, <https://doi.org/10.1016/j.jafrearsci.2009.09.002>
- Roberts, E.M., Stevens, N.J. *et al.* 2012b. Initiation of the western branch of the East African Rift coeval with the eastern branch. *Nature Geoscience*, **5**, 289–294, <https://doi.org/10.1038/ngeo1432>
- Roberts, D., Chowdhury, P.R., Lowe, S.J. and Christensen, A.N. 2016. Airborne gravity gradiometer surveying of petroleum systems under Lake Tanganyika, Tanzania. *Exploration Geophysics*, **47**, 228–236, <https://doi.org/10.1071/EG15075>
- Rogers, N.W. 2006. Basaltic magmatism and the geodynamics of the East African Rift System. *Geological Society, London, Special Publications*, **259**, 77–93, <https://doi.org/10.1144/GSL.SP.2006.259.01.08>
- Rudnick, R. and Fountain, D.M. 1995. Nature and composition of the continental crust: A lower crustal perspective. *Reviews of Geophysics*, **33**, 267–309, <https://doi.org/10.1029/95RG01302>
- Schoell, M., Tietze, K. and Schobert, S.M. 1988. Origin of methane in Lake Kivu (east-central Africa). *Chemical Geology*, **71**, 257–265, [https://doi.org/10.1016/0009-2541\(88\)90119-2](https://doi.org/10.1016/0009-2541(88)90119-2)
- Sengör, A.M. and Burke, K. 1978. Relative timing of rifting and volcanism on Earth and its tectonic implications. *Geophysical Research Letters*, **5**, 419–421, <https://doi.org/10.1029/GL005i006p00419>
- Shuster, D.L., Flowers, R.M. and Farley, K.A. 2006. The influence of natural radiation damage on helium diffusion kinetics in apatite. *Earth and Planetary Science Letters*, **249**, 148–161, <https://doi.org/10.1016/j.epsl.2006.07.028>
- Sorenson, R. 2005. A dynamic model for the Permian Panhandle and Hugoton fields, western Anadarko basin. *AAPG Bulletin*, **89**, 921–938, <https://doi.org/10.1306/03010504045>
- Stewart, W. W. and Street, B. A. 1992. Labarge Anticline. In: *Wyoming Oil and Gas Fields Symposium, Greater Green River Basin and Overthrust Belt*. Wyoming Geological Association, Casper, WY, 200–205.
- Stilwell D. P. 1989. CO₂ resources of the Moxa Arch and the Madison Reservoir. In: *Gas Resources of Wyoming: 40th Field Conference Guidebook*. Wyoming Geological Association, Casper, WY, 105–115.
- Stokes, M. 2013. Our research is on ice due to shortage of helium. *The Independent*, 4 January, <https://www.independent.co.uk/news/science/our-research-is-on-ice-due-to-shortage-of-helium-8439110.html>
- Stuart, F., Turner, G. and Taylor, R. 1994. He–Ar isotope systematics of fluid inclusions: resolving mantle and crustal contributions to hydrothermal fluids. In: Matsuda, J. (ed.) *Noble Gas Geochemistry and Cosmochemistry*. Terra Scientific Publishing, Tokyo, 261–277.
- Tedesco, D., Tassi, F., Vaselli, O., Poreda, R.J., Darrah, T., Cuoco, E. and Yalire, M.M. 2010. Gas isotopic signatures (He, C, and Ar) in the Lake Kivu region (western branch of the East African rift system): Geodynamic and volcanological implications. *Journal of Geophysical Research: Solid Earth*, **115**, B01205, <https://doi.org/10.1029/2008JB006227>
- Tiercelin, J.J., Pflumio, C. *et al.* 1993. Hydrothermal vents in Lake Tanganyika, East African, Rift system. *Geology*, **21**, 499–502, [https://doi.org/10.1130/0091-7613\(1993\)021<0499:HVILTE>2.3.CO;2](https://doi.org/10.1130/0091-7613(1993)021<0499:HVILTE>2.3.CO;2)
- Torgersen, T. 1989. Terrestrial helium degassing fluxes and the atmospheric helium budget: Implications with respect to the degassing processes of continental crust. *Chemical Geology: Isotope Geoscience Section*, **79**, 1–14, [https://doi.org/10.1016/0168-9622\(89\)90002-X](https://doi.org/10.1016/0168-9622(89)90002-X)
- Torgersen, T. 2010. Continental degassing flux of ⁴He and its variability. *Geochemistry, Geophysics, Geosystems*, **11**, Q06002, <https://doi.org/10.1029/2009GC002930>
- Uwe, R. 2014. The East African Rift system. *Austrian Journal of Earth Sciences*, **107**, 132–146.
- Vogt, M., Kröner, A., Poller, U., Sommer, H., Muhongo, S. and Wingate, M.T.D. 2006. Archaean and Palaeoproterozoic gneisses reworked during a Neoproterozoic (Pan-African) high-grade event in the Mozambique belt of East Africa: Structural relationships and zircon ages from the Kidatu area, central Tanzania. *Journal of African Earth Sciences*, **45**, 139–155, <https://doi.org/10.1016/j.jafrearsci.2006.01.012>
- Walker, B.G. 1969. Springs of deep seated origin in Tanzania. In: Kacura, G. (ed.) *Proceedings of the 23rd International Geological Congress, International Union of Geological Sciences, Beijing, China, Volume 19*, 171–180.
- Warr, O., Sherwood Lollar, B. *et al.* 2018. Tracing ancient hydrogeological fracture network age and compartmentalisation using noble gases. *Geochimica et Cosmochimica Acta*, **222**, 340–362, <https://doi.org/10.1016/j.gca.2017.10.022>
- Weeraratne, D.S., Forsyth, D.W., Fischer, K.M. and Nyblade, A.A. 2003. Evidence for an upper mantle plume beneath the Tanzanian craton from Rayleigh wave tomography. *Journal of Geophysical Research: Solid Earth*, **108**, 2427, <https://doi.org/10.1029/2002JB002273>
- Weinlich, F.H., Bräuer, K., Kämpf, H., Strauch, G., Tesař, J. and Weise, S.M. 1999. An active subcontinental mantle volatile system in the western Eger rift, Central Europe: Gas flux, isotopic (He, C, and N) and compositional fingerprints. *Geochimica et Cosmochimica Acta*, **63**, 3653–3671, [https://doi.org/10.1016/S0016-7037\(99\)00187-8](https://doi.org/10.1016/S0016-7037(99)00187-8)
- Wescott, W.A., Krebs, W.N., Engelhardt, D.W. and Cunningham, S.M. 1991. New biostratigraphic age dates from the Lake Rukwa rift basin in western Tanzania. *AAPG Bulletin*, **75**, 1255–1263.
- Wheeler, W.H. and Karson, J.A. 1994. Extension and subsidence adjacent to a 'weak' continental transform: An example from the Rukwa rift, East Africa. *Geology*, **22**, 625–628, [https://doi.org/10.1130/0091-7613\(1994\)022<0625:EASATA>2.3.CO;2](https://doi.org/10.1130/0091-7613(1994)022<0625:EASATA>2.3.CO;2)
- Wingerter, H. R. 1968. Greenwood gas field, Kansas, Colorado, and Oklahoma: Natural gas in Kansas. *AAPG Memoirs*, **9**, 1557–1566, <https://doi.org/10.1306/M9363C104>
- Wolf, R.A., Farley, K.A. and Silver, L.T. 1996. Helium diffusion and low-temperature thermochronometry of apatite. *Geochimica et Cosmochimica Acta*, **60**, 4231–4240, [https://doi.org/10.1016/S0016-7037\(96\)00192-5](https://doi.org/10.1016/S0016-7037(96)00192-5)
- Wollenweber, J., a Alles, S., Kronimus, A., Busch, A., Stanjek, H. and Krooss, B.M. 2009. Caprock and overburden processes in geological CO₂ storage: An experimental study on sealing efficiency and mineral alterations. *Energy Procedia*, **1**, 3469–3476, <https://doi.org/10.1016/j.egypro.2009.02.138>
- Zadnik, M.G. and Jeffery, P.M. 1985. Radiogenic neon in an Archaean anorthosite. *Chemical Geology: Isotope Geoscience Section*, **52**, 119–125, [https://doi.org/10.1016/0168-9622\(85\)90012-0](https://doi.org/10.1016/0168-9622(85)90012-0)
- Zhu, Y., Shi, B. and Fang, C. 2000. The isotopic compositions of molecular nitrogen: implications on their origins in natural gas accumulations. *Chemical Geology*, **164**, 321–330, [https://doi.org/10.1016/S0009-2541\(99\)00151-5](https://doi.org/10.1016/S0009-2541(99)00151-5)