The relationship among naturally occurring radionuclides, geology, and geography: Tsodilo Hills, Botswana

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ABSTRACT
The area around Tsodilo Hills in the northwestern part of Botswana provided an opportunity to investigate the relationship among naturally occurring radionuclides, geology, and geography. In situ gamma ray surveys were carried out close to the hills in order to assess radionuclide concentrations. The concentrations of potassium, thorium, and uranium were measured and plotted with the help of Geographical Information System (GIS) software. The measurements showed high uranium concentrations and relatively low potassium and thorium concentrations. The highest concentrations were found to be 223 Bq/kg for uranium, 258 Bq/kg for potassium, and 31 Bq/kg for thorium. The measurements demonstrated that high uranium concentrations occurred in low-lying areas close to Tsodilo Hills. It is suggested that uranium concentrations were caused by alluvial deposition, mainly from the hills. Similar results were also found on a road leading to Tsodilo Hills. The article finally draws conclusions as to the distinctive geographical environment at Tsodilo Hills and the resultant unique distribution patterns of naturally occurring radionuclides at the site.

1. Introduction
Tsodilo Hills in the northeastern part of Botswana is a distinctive group of outcrops in the mainly flat landscape of the Kalahari Desert. The highest and most southerly hill is the Male (Nxum Ngxo); the largest hill, situated in the center, is the Female (Naum Di), while the two smaller hills to the north are the Child (Roannin) and the Grandchild. These hills are the only protrusions within a radius of 100 km and also one of the few places in this area of the Kalahari where the bedrock is clearly visible. These hills provided stone tools, iron, water, and shelter to past inhabitants for thousands of years, and this resulted in a collection of 4500 rock paintings that are scattered among the rock formations. Tsodilo Hills has the highest peak in Botswana (1395 m); this, along with its geographical uniqueness as well as its isolation, has led to numerous traditional legends and beliefs about the hills. Consequently, Tsodilo Hills was declared a World Heritage Site in 2001 (ICOMOS, 2001).

The region around Tsodilo Hills is semiarid with no perennial source of water in close proximity, except for the panhandle of the Okavango Delta about 50 km to the west. The distinctive geographical characteristics of the region are a result of geology and ancient meteorological phenomena, and various studies on this have been conducted (Jacobberger & Hooper, 1991; Thomas & Shaw, 2002; Thomas et al., 2003; Wendorff, 2005). Several geophysics methods, which include techniques of remote sensing, were employed to better understand this unique environment. Investigation of the distribution patterns of naturally occurring radionuclides in the area has however never been conducted previously.

Naturally occurring radionuclides are of primordial origin, and the distribution and concentration of these nuclides are directly related to geological and geographical factors. Measurements of these elements are therefore often used for exploration and environmental purposes. Some of the exposed bedrocks in Tsodilo Hills contain high levels of heavy metals that are usually accompanied by naturally occurring radionuclides. The naturally occurring radionuclide concentrations in the area around the Tsodilo Hills will consequently demonstrate characteristics of the geology and geography of the hills. It has also been shown that some archeological sites have unique concentrations of uranium and potassium radionuclides (Bezuidenhout, 2012). An exploratory radionuclide survey was conducted at Tsodilo Hills to investigate these and other relationships.

2. Geology of Tsodilo Hills
Tsodilo Hills withstood denudation over millions of years and is a rare outcrop, within a wide Kalahari sand cover, of old rocks that were deformed and uplifted by plate tectonic forces (Segadika, 2006). The geology of the Tsodilo Hills can be dated back to the Pan-African Orogeny when the Greater Congo craton...
amalgamated with the Kalahari craton and thereby formed parts of the supercontinent of Gondwana (Wendorff, 2005). The Tsodilo Hills group is associated with meta-quartzites, meta-conglomerates, and quartz-mica schists that were all deformed around 530 Ma (Wendorff, 2005). The Pan-African deformation also locally faulted, sheared, and thrusted the strata into the different elements that make up the eroded Tsodilo Hills geological complex today (see Figure 1).

The lithological complexes were initially subdivided into four units by Vermaak (1961), numbered I–IV in stratigraphically ascending order. This stratigraphy was later revised by Wendorff (2005), as illustrated by the most complete lithostratigraphic section through Nxum Ngxo (Male Hill) in Figure 1(B).

Geological elements with high levels of heavy minerals are regularly associated with enhanced concentrations of heavier naturally occurring radionuclides, specifically uranium and thorium (De Meijer, 1998). In the case of the Tsodilo Hills, heavy minerals may have been concentrated within the area’s coarser-grained quartzitic, pebbly to conglomeratic meta-sediments, and thereby of particular importance to this study. Each of the area’s four lithological units contains strata of this nature, but unit I demonstrates the highest density of strata that contains heavy minerals.

3. Geography of Tsodilo Hills

The four hills of the Tsodilo Hills complex stand in a sea of sand in the Kalahari with the Male Hill rising 420 m above the desert floor (see Figure 2). The Female Hill has an overall area of almost three times that of the Male and has several peaks, with the highest being about 300 m above the desert floor. The Female is also covered by denser vegetation and have a few springs with fresh water. The smaller Child and the Grandchild, which are respectively about 2 km and 4 km northwest of the Female, are much less remarkable from the geological and geographical points of view.

The Kalahari Desert around Tsodilo Hills is dominated by ranges of old linear sand dunes, which was

Figure 1. (A) A Geological map of the Tsodilo Hills (de Wit & Main, 2016). (B) A lithostratigraphic column of the Tsodilo Hills group as exposed on the Male Hill (Wendorff, 2005).
the effect of a paleoclimate similar to the current arid Kalahari Sandveld (Jacobberger & Hooper, 1991). The dunes, now degraded in form, were shaped by prevailing easterly winds during a much earlier dryer period. Aeolian deposits from this time resulted in the western slope of Tsodilo Hills being much steeper than the eastern slope. The sediment around Tsodilo Hills mainly consists of sand, duricrust, and pan sediment. There were two ancient lakes just west of the Male and Female Hills, which were supplied by runoff from the hills and the surrounding area (de Wit & Main, 2016). The lake depressions still gather runoff water and alluvial sediment from the hills and the surrounding area during the rainy season.

4. Experimental

Gamma radiations from three naturally occurring radionuclides were measured and interpreted. The nuclides that were studied were uranium (\(^{238}\)U), thorium (\(^{232}\)Th), and potassium (\(^{40}\)K). Potassium is the most abundant and has a one-step decay process, contrary to uranium and thorium, which decay through long chains before reaching stable nuclides. Secular equilibrium was assumed for the uranium and thorium decay chains due to the relatively undisturbed study area.

4.1. Measuring system

A Gamma In Situ Portable Instrument (GISPI) was employed to survey the tracks to and around Tsodilo Hills. The GISPI comprises a NaI(Tl) scintillation detector, a digital Multichannel Analyser (MCA), a tablet PC with an onboard Global Positioning System (GPS), and real-time analyses software that controls the entire system (Bezuidenhout, 2015a). The NaI(Tl) detector (7.62 x 7.62 cm) was attached to the MCA and fitted in a tailor-made steel housing for protection. The scintiSPEC- MCA (http://gs.flir.com/) that was produced by FLIR has a Universal Serial Bus (USB) connection that provides power for operation and transfers data. A Trimble- Yuma rugged tablet PC (http://www.trimble.com/) managed the systems and data acquisition. The detection part of the system was mounted on the front metal bar of a motor vehicle, 20 cm from the ground (see Figure 3). The Yuma rugged tablet PC was fitted in the cabin, and electrical power was supplied from the motor vehicle.

The winTMCA32 code controlled the system settings and spectrum acquisition. The code also continuously acquired the geographical position coordinated from Yuma’s onboard GPS. The code stored all the settings, in situ spectra, and the extracted results in files on the Yuma. The data files were finally analyzed and illustrated by means of QGIS software.

4.2. Calibrations

Energy calibrations were carried out between 0.2 and 2.7 MeV by utilizing cesium (\(^{137}\)Cs), cobalt (\(^{57}\)Co), and environmental spectra. The symmetry assumptions and corrections as described by McCay et al. (2014) were assumed for the calibrations and surveys.

The efficiency calibrations of the GISPI were conducted at standard radiation reference pads of the Nuclear Energy Corporation of South Africa (NECSA) according to a method described previously (Chiozzi, De Felice, Fazio, Pasquale, & Verdoy, 2000; Corner, Toens, Richards, Van As, & Vleggaar, 1979). All the nuclide concentrations were finally calculated and expressed in Becquerel per kilogram (Bq/kg). The efficiency calibration was performed by choosing 1460.8 keV, 1764.5 keV, and 2614.5 keV as the energy counting windows for potassium, uranium, and thorium, respectively. The conversions form counts per energy window to Becquerel per kilogram were conducted according to the method described by Bezuidenhout (2015b).
4.3. Measurements and analyses

Potassium is abundant in nature, and the gamma emission signal of the $^{40}$K nuclide of natural potassium is subsequently significant in environmental spectra. The 1460 keV gamma emission of $^{40}$K was consequently chosen as the centroid for a continuous stabilization function. This stabilization function, which is programmed in winTMCA32 software, conducts energy drift corrections.

A few accessible roads to the west of Tsodilo Hills were surveyed, as well as the access road that enters the area from the Okavango panhandle. These were all dirt roads with no added surface material that were brought in from elsewhere. The activity concentrations of potassium, uranium, and thorium of the various sample points were finally plotted and interpolated with QGIS software and overlaid on Google Earth images of the area. The images are illustrated in Figure 4, 6, 7, 8, and 9.

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**Figure 3.** Photograph of the setup of the measuring system that was utilized during the in situ measurements. The photograph shows the casing with the detector housing mounted on the vehicle. The tablet with an onboard GPS was placed in the cabin of the vehicle for ease of continuous operator monitoring.

**Figure 4.** A Google Earth image with a color-graded overlay indicating the potassium concentrations west from Tsodilo Hills. The elevation profile of the route from A to B is displayed in Figure 5.
5. Results and discussion

The measured potassium, uranium, and thorium concentrations of the survey to the west of the Male and Female Hills are plotted in Figures 4, 6, and 7, respectively. The potassium concentrations that were measured (~130 Bq/kg) are relatively low when compared to the average levels of the potassium in the crust of the Earth at ~500 Bq/kg (Watanabe et al., 2014). There is a slight increase in the potassium concentrations over the route close to the Female Hill. This increase occurs at a depression and can be seen in Figure 4 when combined with the Google Earth elevation profile of the measured route from A to B in Figure 5. Potassium occurs in many types of sediments like feldspar, micas, clays, and salts and is easily transportable by means of alluvial processes (Osmond & Ivanovich, 1992; Rosholt, 1992). The increase of potassium in the depression can therefore be linked to alluvial sediment that is deposited during rainy seasons.

The uranium concentrations on the route are relatively high with substantial variation between A and B. The highest concentrations are mainly confined to two low-lying areas that are close to the Male and Female Hills. These low-lying areas are clearly observable in the Google Earth elevation profile (Figure 5) of the measured route, A to B. The average thorium concentration that was measured (15 Bq/kg) is substantially less than the average values of uranium concentrations (162 Bq/kg). The variation in thorium concentrations is also smaller than that of uranium, but a slight increase in thorium is visible in the depression at the Male Hill (see Figure 7).

Heavy minerals such as zircons, uraninites, and thorites have high concentrations of thorium and
uranium (Hurst, 1990). The lithological units of the Male and Female Hills revealed numerous strata with heavy minerals. Uranium is soluble in neutral aqueous fluids like water and more susceptible to mobilization than thorium. This is particularly true for weathering, where potassium and uranium are easily mobilized but thorium is not (Osmond & Ivanovich, 1992; Rosholt, 1992).

It is therefore likely that uranium might have been transported from the hills and the surrounding area by means of alluvial processes and subsequently deposited in the depressions. This would explain the high concentrations of uranium in the depressions below the Male and Female Hills. These depressions were identified as ancient lakes (de Wit & Main, 2016), which means that the alluvial deposition might have been taking place for thousands of years. The slightly elevated concentrations of thorium at the Male Hill can also be explained by alluvial deposition from the surrounding hills (see Figure 7). The thorium concentrations are however substantially lower than those of uranium, which can be attributed to the higher solubility of uranium when compared to thorium.

Naturally occurring radionuclides were also measured on the access road to Tsodilo Hills leading from the Okavango panhandle. The potassium and
uranium concentrations of this survey are, respectively, plotted in Figures 8 and 9. It is evident that the potassium and uranium concentrations on the road are well below the levels measured at Tsodilo Hills. This may be due to the difference in origin of the sediment on the road and that around Tsodilo Hills. The road to Tsodilo Hills crosses paliodunes that were formed due to Aeolian deposition, compared to the sediment in the depressions at Tsodilo Hills that originated from alluvial deposition. This low-lying area west of Tsodilo Hills is visible in the Google Earth elevation profile in Figure 9. It is also noticeable that the potassium and uranium concentrations show a general increase when nearing Tsodilo Hills, which further supports the theory that the hills may have been the consistent source of radionuclides in this area.

Elevated uranium concentrations are visible nearly halfway between the Okavango panhandle and Tsodilo Hills (Figure 9). The Google Earth elevation profile in Figure 9 shows that this part of the road is relatively even with hills both toward the Okavango panhandle and Tsodilo Hills. This may indicate that this area is part of a flood plain that gathers alluvial deposits during the rainy season, consequently accumulating uranium from the surrounding area. The sharp increase in the concentration of uranium is also evident when crossing the Tsodilo Hills ridge and dropping into the low-lying area to the west of the hills.

This study consistently demonstrated elevated uranium concentrations on the lower-lying Kalahari plain to the west of Tsodilo Hills. High levels of naturally occurring radionuclides would occur in the lithology of Tsodilo Hills due to its geological origin and composition. This is in contrast to the radionuclide levels in the sands of the paliodunes of the surrounding Kalahari Desert, which were formed by winds during an earlier period with a dryer climate. The Aeolian sand also gathered on the earthen slope of Tsodilo Hills resulting in an elevated and gradually sloping Kalahari plain on that side. The localized and elevated uranium concentrations to the west of Tsodilo Hills are thus a geographical exception in the area, and the heavy minerals in the strata of the Tsodilo Hills formation appear to be the source. Further studies to investigate the larger area around Tsodilo Hills, as well as all four outcrops in the complex, are required to better understand this.

6. Conclusions

Tsodilo Hills in the Botswana section of the Kalahari Desert has a unique geographical environment. A pilot survey of naturally occurring radionuclide concentrations in this area was conducted with the GISPI. Results of this survey indicate high concentrations of uranium in two ancient lake depressions next to the Male and Female Hills of the complex. The average concentration of uranium in the area to the west of the hills was significantly higher than the concentrations of potassium and thorium. Marginally elevated concentrations of potassium and thorium were however also measured in some of the depressions. Elevated concentrations of uranium were also found in a flood plain that lies between the Okavango panhandle and Tsodilo Hills.
This study demonstrated that the geology and geography of Tsodilo Hills influenced the distribution patterns of natural occurring radionuclides in that area. It is evident that the geographical uniqueness of the site also influences the composition and distribution of radionuclides, especially that of uranium. The study suggests that long-term alluvial deposition directly relates to high uranium concentrations and to a lesser extent to potassium and thorium concentrations. Additional measurements are planned to survey a larger area around Tsodilo Hills in order to determine the extent of these findings.

Disclosure statement

No potential conflict of interest was reported by the author.

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