

Uranium natural levels in soil, rock and water: assessment of the quality of drinking water in Singida Urban District, Tanzania

S. J. Kaishwa, E. M. Marwa, J. J. Msaky and W. N. Mwakalasya

ABSTRACT

An analytical study was carried out to determine the level of uranium in the soil and rock and associate it with the levels in surface and underground water, which are the main sources of drinking water in Singida Urban District, Tanzania. Fifteen water samples, 12 soil samples and nine rock samples were collected in Singida Urban District in February 2016. Water samples were analysed by the Government Chemist Laboratory Agency using inductively coupled plasma optical emission spectrometry (ICP-OES) (2007) and analyses for soil and rock samples were carried out through total X-ray fluorescence (XL3 ANALYSER) at Sokoine University of Agriculture in the Department of Soil and Geological Sciences. Uranium levels in the soils and rocks samples found ranged from 3.744 to 8.754 mg kg⁻¹, SE ± 0.849 and *P*-value <0.001 for soil and 20.01–31.57 mg kg⁻¹, SE ± 2.474 and *P*-value 0.077 for rocks respectively. The levels in soil and rock influences the levels in surface and underground water which were between 0.087 and 1.097 mg L⁻¹ for surface water and <0.01–0.46 mg L⁻¹ for underground water, respectively. Singida Urban District has higher uranium levels in the soil and rock with a consequence of high levels in drinking water sources.

Key words | drinking water, rock, soil, surface water, underground water, uranium

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INTRODUCTION

Uranium (U) is naturally abundant, occurring in granites and various other naturally occurring mineral deposits. It occurs as three natural isotopes: ²³⁸U, ²³⁵U and ²³⁴U, all of which are radionuclides with low specific activity. U is present in the environment as a result of leaching from natural deposits, release in mill tailings, emissions from the nuclear industry, the combustion of coal and other fuels and the use of phosphate fertilisers that contain U (WHO 2014). Its chemical toxicity is of greater concern than its radiotoxicity. U (IV) is the dominant species of uranium present in primary minerals (Smedley *et al.* 2006). This species is associated with hydroxides, phosphates and fluorides (Handley-Sidhu *et al.* 2010) and is relatively insoluble but can readily oxidise to U (VI), which is stable. Uranium can be enriched in soil via natural paedogenesis processes with

U being released from parent materials to soil and soil solution, and via human activities. Mining phosphate ores can carry U to the soil and river water resources surrounding the mines. Phosphate rocks may contain considerable amounts of U (Romero Guzmán *et al.* 1995).

In Tanzania, U deposits have been found as far back as the 1950s in the Bahi swamp (Cygan & Tazaki 2014). In the early post-independence era (1961–1970s) the country was subjected to a systematic airborne survey for radioactive occurrences. In this period, it was evident that four geological areas in Tanzania contain potentially significant U deposits viable for commercial mining activities (Knivslund 2012). These deposits occur in the lowland areas surrounding the isolated hills of Precambrian granites. Water is the main transport vector for U. In confined aquifers, U occurs in the

tetravalent state, whereas in unconfined or surface water its state is hexavalent (Canfield *et al.* 2006). The soluble products of rock weathering are transported into the groundwater and eventually into the rivers and then to the ocean. The actual content of sea water is specified at a level of $3 \text{ gL}^{-1} \text{ U}$ (Coward & Burnett 1994; Pais & Jones 1997). Reductive precipitation of U (IV) in anoxic marine sediments is globally the most significant sink for dissolved U.

Uranium in surface water can disperse over large distances to ponds, rivers, and oceans. The transport and dispersion of U in surface water and groundwater are affected by adsorption and desorption of U on aquatic sediments. As with soil, factors that control mobility of U in water include oxidation-reduction potential, pH, and sorbing characteristics of sediments and the suspended solids in water (Swanson 1985; Brunskill & Wilkinson 1987). In groundwater, the weathering of U-bearing rocks and minerals is the source of dissolved U. The concentration of U in groundwater is usually in the range $0.1\text{--}50 \mu\text{g L}^{-1}$ (Rivas 2006), and in contaminated groundwater plumes, concentration of uranium is much more, i.e. U concentration of groundwater in some U mill tailing in United States varied from 0.07 to 3.05 mg L^{-1} (Baumgartner *et al.* 2000).

The presence of U in drilled water wells in Singida required studying for the health and safety of people because U and its compounds are carcinogenic and highly toxic (Sasmaz & Yaman 2008). This study aimed to determine the level of U in the soil and rock and associate it with the levels in surface and underground water which are the main sources of drinking water in Singida Urban District, Tanzania.

METHODS

Study area

Soil, water, rock and plant samples were collected in Singida Urban District. Singida Urban District is located between $4^{\circ} 40'$ east and $4^{\circ} 53'$ south of the Equator and longitude $34^{\circ} 30'$ and $34^{\circ} 53'$ east of GMT. It covers an area of 754 km^2 . The district is bordered to the south and west by the Ikungi District and to the north and east by the Singida Rural District. It is situated at an elevation of $1,547 \text{ m}$ above sea level. The sites where the studied samples were collected are as indicated in Figure 1.

Water sampling

Underground and surface water samples were obtained from 15 different drawing points of drinking water in Singida Urban District (Figure 1). Underground water sources were predominant among the samples (13 samples) as it is the major source used as compared to only two sources of surface water. Underground water samples were collected from tap water, whereby tap water was left to run for about 3 minutes to empty standing water before collection. Surface water samples were collected from the two lakes in Singida Urban District. Each water sample had a volume of 0.5 L and was sampled by using clean and sterile bottles.

Soil and rock sampling

Along the dominant slope, the study site was partitioned into three, designated as the upper, middle and lower slopes. In each segment, soil sampling was carried out by using an auger at $0\text{--}30$, $30\text{--}60 \text{ cm}$ and $60\text{--}100 \text{ cm}$ depth, making a total of 12 samples. The main objective of this sampling was to determine vertical and horizontal U distribution along the slopes. In each of the three partitioned sites (Darajani, Misuna and Burudani), three samples of exposed rock around the area were taken to make a total of nine samples in order to analyse and determine levels of uranium in those rocks. There were no rock outcrops at Irao and thus, no rock sample was collected from this area for analysis.

Analysis of water samples

Water samples were analysed by the Government Chemist Laboratory Agency, which is the legal referral laboratory in Tanzania. Eight mL of water samples were mixed with 1 mL of H_2NO_5 . The volume was put into vials and U concentration was directly determined using inductively coupled plasma optical emission spectrometry (ICP-OES) (2007).

Soil and rock processing and analysis

Rock samples were crushed and sieved to pass through $<2 \text{ mm}$ holes for laboratory analysis. Similarly, the soil samples were sieved through the same mesh. Analyses of rock and soil samples were carried out at Sokoine University

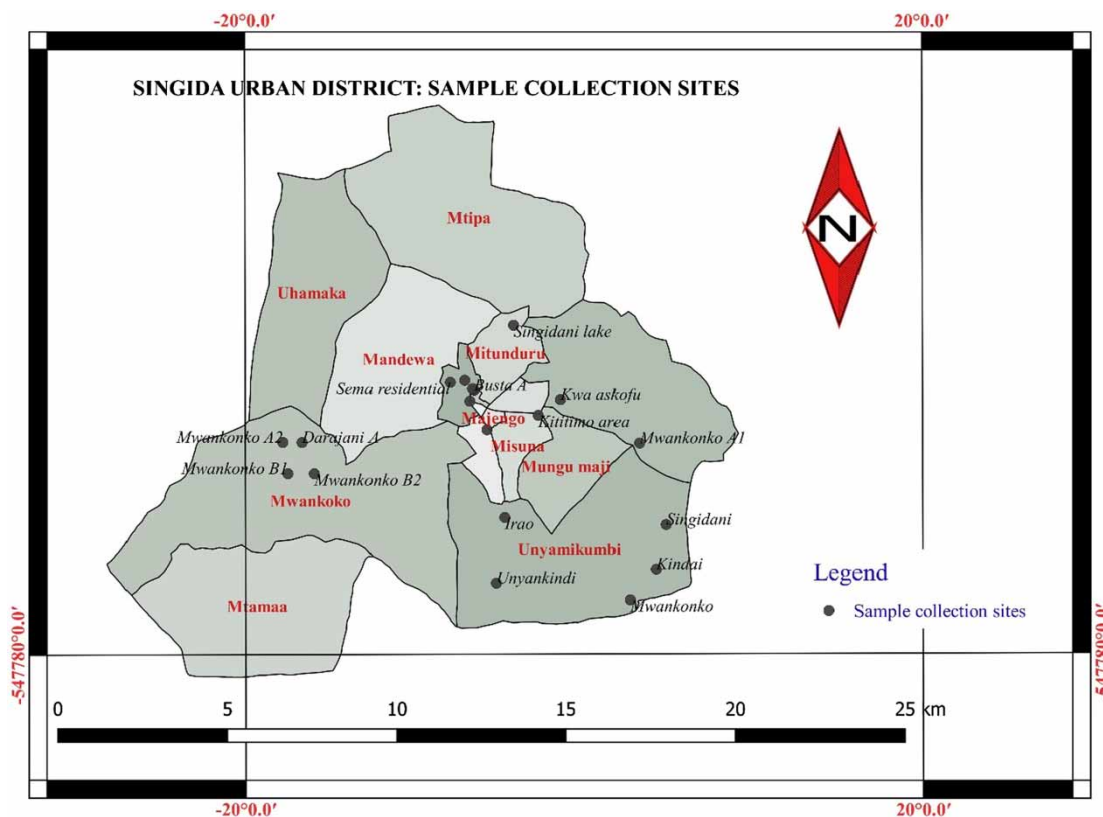


Figure 1 | Sample collection sites.

of Agriculture in the Department of Soil and Geological Sciences. Total X-ray fluorescence (XL3 ANALYSER) was used to determine the levels of uranium in soils and rocks as described by Hunter & Bertsch (1998). Fifty grams of soils and rocks samples were weighed in a test tube by using analytical balance (EN ISO 20483) and then placed in the transparent plastic bags. After that the plastic bags were placed in the screen of an XL3 ANALYSER machine. Uranium concentration in soils and rocks samples was read directly using an XL3 ANALYSER.

RESULTS

Concentration and distribution of uranium in soil

U levels in the soils of Darajani, Irao, Misuna, and Burudani areas are shown in Table 1. These results indicated that, ignoring other possible attributes, the largest amount of U

(8.754 mg kg⁻¹) was obtained in the soil collected from Burudani site. This was followed by the U in soils of Misuna (5.424 mg kg⁻¹) and Irao (5.354 mg kg⁻¹) which did not differ significantly between them. The significantly lower U level (3.744 mg kg⁻¹) was obtained at Darajani.

Table 1 | Mean values of uranium levels in soils

Site	Mean uranium (mg kg ⁻¹)
Darajani	3.744a ^a
Irao	5.354b
Misuna	5.424b
Burudani	8.754c
SE ±	0.849
LSD0.05	1.723
CV (%)	25.3
P-value	<.001

^aMeans along the same column bearing similar letter(s) do not differ significantly at 5% level of probability based on Tukey's honest test.

The mean U elevation value in all areas indicated that U level increases with decreasing elevation down slope, as shown in Table 2. The high level of U at lower elevations could be due to U concentrated from parent material (granite) by weathering and erosion and thus being washed down the slope.

The results indicated that the level of U at Irao and Darajani areas decreases with depth along the soil as presented

in Table 2. The higher amount of U in the upper surface indicates that U is coming from the surface and not from underneath. Exposed granites are weathering and releasing U which is washed down the slope by runoff which then accumulates on the surface. Some are leached down the soil profile.

A high amount of finer particles in the form of clays and low sand content in the upper surface can also cause high U levels in the upper surface. This occurs in Irao and Darajani, as shown in Table 2, where the U level decreases as the clay content decreases. This is because clay minerals are negatively charged and hence they have the ability to retain U cations which are positive.

Table 2 | Variation of uranium with elevation and depth of sampling

Site	Elevation	Depth (cm)	Uranium level (mg kg ⁻¹)	Mean U elevation value (mg kg ⁻¹)	Textural class	
Irao	Lower	0–30	8.15	7.06	CL	
		30–60	7.43		SCL	
		60–100	5.61		SC	
	Middle	0–30	7.61		4.97	SCL
		30–60	3.65			SL
		60–100	3.65			LS
	Top	0–30	5.42		4.03	SC
		30–60	4.00			SCL
		60–100	2.68			LS
Darajani	Lower	0–30	5.24	4.23	SCL	
		30–60	2.55		LS	
		60–100	2.33		S	
	Middle	0–30	4.05		3.76	CL
		30–60	3.89			SL
		60–100	3.35			SC
	Top	0–30	4.82		4.23	SCL
		30–60	4.68			CL
		60–100	3.19			SL
Burdani	Lower	0–30	7.83	9.95	S	
		30–60	8.66		SL	
		60–100	12.65		SCL	
	Middle	0–30	7.86		8.72	SL
		30–60	10.44			SCL
		60–100	11.06			SC
	Top	0–30	5.55		7.59	LS
		30–60	8.55			SL
		60–100	9.38			SC
Misuna	Lower	0–30	2.55	6.93	S	
		30–60	7.52		SL	
		60–100	7.72		SCL	
	Middle	0–30	4.00		5.72	SL
		30–60	4.60			SCL
		60–100	5.23			SG
	Top	0–30	5.52		6.18	LS
		30–60	6.14			SCL
		60–100	6.88			SC

Textural class: SL, Sandy Loamy; CL, Clay Loamy; SCL, Sandy Clay Loamy; SC, Sandy Clay; S, Sandy; LS, Loamy Sandy.

Concentration and distribution of uranium in rocks

The highest U level (31.57 ± 2.474 mg kg⁻¹) was recorded in Burudani followed by Misuna rocks (28.98 ± 2.474 mg kg⁻¹) (Table 3). On the other hand, the lowest U amount (20.01 mg kg⁻¹) was recorded from Darajani rocks.

Uranium levels in surface waters

U concentration from surface water sources of Kindai and Singidani lakes taken at an elevation of 1,481 and 1,484 metres was found to be at 0.087 and 1.097 mg L⁻¹ respectively (Table 4). U levels were statistically higher in high H₂O pH values ($t = 25.863$, $P = 0.025$). However, the outcome is not statistically plausible due to the small sample size.

Table 3 | Levels of uranium in granite rocks

Area	Mean uranium (mg kg ⁻¹)
Darajani	20.01a ^a
Misuna	28.98ab
Burdani	31.57b
LSD _(0.05)	10.64
CV (%)	9.2
SE ±	2.474
P-value	0.077

^aMeans along the same column sharing similar letter(s) do not differ significantly at 5% based on Duncan's' New Multiple Range Test (DNMRT) (Duncan 1995).

Table 4 | Uranium levels in surface waters

Site name	Geographical coordinates	Elevation (m)	Source	Uranium levels (mg L ⁻¹)	H ₂ O pH value
Kindai lake	4°50'4.6"E and 34°44'9.1"S	1,481	Surface	1.097	°7.81
Singidani lake	4°46'51.3"E and 34°46'0.9"S	1,484	Surface	0.087	7.34

Uranium levels in underground waters

The concentration of U from 13 underground water samples taken at an elevation difference of up to 52 metres (minimum 1,464 and maximum 1,516 metres) ranged from 0.00 to 0.46 mg L⁻¹ with a mean value of 0.09 mg L⁻¹ (Table 5). Correlation coefficient ($r = 0.422$, $P = 0.151$) suggests a weak non-significant positive linear relationship between U levels (mg L⁻¹) and H₂O pH value. The pH determines U complexes that are formed in water and hence its accumulation. Where there are carbonate compounds such as soda ash, which is locally produced in Singida from Kindai and Singidani lakes, at high pH U forms soluble uranyl carbonate complexes (Duff & Amrhein 1996).

DISCUSSION

Based on the recommendation given by the Canadian Conference on Medical Education (CCME) from the Canadian Soil

Quality Guidelines for the protection of environmental and human health, the levels of U in the soil do not affect agricultural use of the soil because the acceptable levels of U are 23 mg kg⁻¹ for agricultural land use and for residential/parkland land use, 33 mg kg⁻¹ for commercial land use, and 300 mg kg⁻¹ for industrial land use (CCME 1997). Therefore, the levels of U in agricultural soil in the study areas ranging from 3.76 to 9.95 mg kg⁻¹ were low and more likely not to cause environmental health hazards to plants, livestock and human beings. However, these levels affected the quality of underground water which is the main source of drinking water.

According to De Craen *et al.* (2004), U content increases with a decrease in soil particle size. Mostly, clay minerals are negatively charged hence they have the ability to retain U cations which are positive. This could be the case because clay content in the Misuna and Burudani increases with soil depth. Soil with higher cation-exchange-capacity will retain more U, while carbonate in the soil increases the mobility of U through the formation of anionic U and CO₃²⁺ complexes (Sheppard & Evenden 1987). U does not

Table 5 | Uranium levels in underground and surface waters

Site name	Coordinate	Elevation (m)	U levels (mg L ⁻¹)	H ₂ O pH value
Mwankonko A1	4°50'36.3"E and 34°50'1.9"S	1,493	0.059	7.70
Mwankonko A2	4°50'35.3"E and 34°38'38.6"S	1,479	0.058	5.99
Darajani A	4°50'35.5"E and 34°39'15.6"S	1,464	0.062	5.43
Mwankonko B1	4°51'36.4"E and 34°38'48.4"S	1,480	0.201	7.25
Mwankonko B2	4°51'36.8"E and 34°39'2"S	1,478	0.193	6.62
Sido area	4°49'7.4"E and 34°44'27.0"S	1,508	0.464	7.49
Kititimo area	4°49'43.2"E and 34°46'48.4"S	1,516	0.018	6.81
Busta A	4°48'54.8"E and 34°44'45.1"S	1,500	0.011	6.25
Busta B	4°48'51.6"E and 34°44'42.1"S	1,491	0.023	6.70
Sema office	4°49'16.2"E and 34°44'36.9"S	1,516	<0.01	6.73
Sema residential	4°48'39.6"E and 34°43'59.2"S	1,506	<0.01	6.67
Irao	4°52'59.4"E and 34°45'44"S	1,503	<0.01	6.64
Kibaoni	4°50'10.8"E and 34°45'9.7"S	1,503	0.24	6.70

migrate substantially in loamy soils as compared to sandy soils (Sheppard *et al.* 1984). U migration in soil occurs over the period of a few months, depending on sorption, and may be upwards when there is a net water deficit or downwards as a result of net leaching (Barescut *et al.* 2005).

The trend in U levels in the soil was the same as that in the rocks. Higher levels of uranium in soils matched well with higher levels of U in granite. For instance, levels of U at Burudani in soil were 7.59–9.95 mg kg⁻¹ while in granite it was 31.57 mg kg⁻¹. A similar pattern can also be seen at Darajani where U levels in both soil and rocks were low (Tables 2 and 3). Weathering of granites may have released equivalent quantities as those found in the soil. It is well known that granite rocks are a primary source of U and release U during weathering (Sasmaz & Yaman 2008). Therefore, an area with U rich granite will automatically have high levels of U in soil and water bodies.

The U concentration from the two surface water samples in this study was found to be very high as compared to the WHO drinking water provisional guidelines value of 0.03 mg L⁻¹ for a maximum consumption of 2 L/day (WHO 2014). It is fortunate that the two lakes are not used for human activities. Drinking water supplied in the study area comes from underground sources. High U levels in underground water is a result of the dissolution of uranium bearing minerals that have been in contact with groundwater for long periods of time (South and West Devon Health Authority 2001). This is an indication of potential health hazards associated with drinking water contaminated with high U levels.

Furthermore, the variation of U in water sources among the study areas could be caused by two factors; pH of water where the sample was taken and concentration of U in the surrounding rocks. Water passing through U rich materials is likely to have a high level of U compared to the water from low U rich material (Singh 1997; Anke *et al.* 2009). High U levels were detected in areas where water pH was high because at high pH, U forms soluble U carbonate complexes (Waite *et al.* 1994). U-carbonate complexes are predominant at neutral to alkaline pH values.

CONCLUSIONS AND RECOMMENDATION

Singida Urban District has higher U levels in the soil and rock with the consequence of high levels in surface and

underground water. Therefore, water supplied to the community from a number of boreholes and lakes is not safe for human consumption as the only treatment method used is chlorination. The study recommends the use of ion exchange with anion resin and reverse osmosis (RO) techniques in order to reduce the amount of U in water before supplying for consumption.

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COMPETING INTERESTS

There were no competing interests in undertaking this study.

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