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RESEARCH ARTICLE

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RADON LEVELS IN GROUNDWATER FROM A URANIUM RICH DISTRICT IN SOUTHERN TANZANIA

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ABSTRACT

Water samples were collected from springs, rivers and wells from Namtumbo District Southern Tanzania, an area with proven commercially viable uranium deposits. Samples were collected from 39 water sources located in eight wards within Namtumbo District. Radon in water was quantified using an active radon monitor (AlphaGUARD) in conjunction with the AquaKIT accessory. Radon concentrations in water samples ranged from 0.2 to 86BqL^{-1}; with a mean value of 28.1BqL^{-1}; 11.3BqL^{-1} and 6.8BqL^{-1} for rivers, springs and wells, respectively. The measured radon concentrations in all collected samples were below the limit of 100 BqL^{-1}as set by the World Health Organization (WHO). However, 42% of the samples exceeded the USEPA action level of 11.1BqL^{-1}. The mean values of the annual effective dose due to ingestion of radon and due to the inhalation of radon released from water are 26.4 and 26.1 µSvy^{-1}, respectively. In addition, the mean values of estimated total annual effective doses are found to be 6.3 µSvy^{-1}. The mean value of total annual effective doses is found to be lower than the reference dose level of 100 µSvy^{-1} recommended by the WHO and the United Nations Scientific Committee on the Effect of Atomic Radiation (UNSCEAR). The mean values of effective doses per annum to the lungs and stomach are 3.1 and 3.2µSv, respectively.

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INTRODUCTION

Radon (^{222}Rn) is a naturally occurring radioactive gas found in the uranium-238 (^{238}U) decay chain (Fig.1). ^{222}Rn is produced continuously in the pore spaces of rocks and soils. A fraction of ^{222}Rn produced can diffuse and enter the atmosphere where it decays forming short-lived daughters and part of it can be dissolved in groundwater hence contributing largely to radioactivity content of groundwater (Alharbi *et al*. 2022).According to Moreno *et al*. (2014), the amount of radon dissolved in groundwater depends on factors such as radium (^{226}Ra) concentration, radon emanation coefficient, air-water partitioning coefficient and aquifer characteristics. Humans obtain water for daily uses from both surface and groundwater sources. ^{222}Rn dissolved in drinking water can lead to irradiation of stomach tissues and small intestine through ingestion and secondly, irradiation of lung tissues through inhalation. Over 90% of organ dose resulting from ingested ^{222}Rn is to the stomach (Kendall and Smith 2002), though there is no definitive link between ingestion of drinking water with radon with increased risk of stomach cancer has been established (US NAS 1999; Auvinen *et al*. 2005; WHO 2022). However, based on the assumption of Linear No threshold (LNT) hypothesis in radiation risk assessment, worldwide efforts are ongoing to study the linear relationship between radon exposure and health risk (WHO 2017). Based on the estimated risk to human health from the LNT model forradiation protection purposes different levels of radon in drinking water have been proposed, see for example: 100 – 1,000 BqL^{-1} for European Union (EURATOM 2013), 500 BqL^{-1} for Spain (Gonzalez *et al*. 2018), 11.1 BqL^{-1} for the United States (US EPA 1999), and 100 BqL^{-1} for the World Health Organization- WHO (WHO 2008). ^{222}Rn and its daughters are the major source of radiation exposure to humans. According to UNSCEAR (2000), radiation dose received by the human population due to the inhalation of ^{222}Rn and ^{220}Rn and their progenies contribute more than 50% of the total dose from natural sources. Therefore, studies on ^{222}Rn levels in drinking water have become of great interest globally. This is evident in studies byAuvinen *et al*. 2005 in Finland, Duggal *et al*. 2017 in India, Kaur *et al*. 2019 in India, Nandakumaran *et al*. 2016 in India, Otwoma and Mustapha 1998 in Kenya, Marques, A. L., *et al*. 2004 in Brazil; Yalim *et al*. 2007 in Turkey; Villalba, *et al* 2005 in Mexico; and El-Taher 2012 in Saudi Arabia, among others. Results from these studies revealed that radon concentrations ranged from below to above the recommended limits though authors compared their results with different references e.g. WHO limit of 100 BqL^{-1}, USEPA limit of 11.1 BqL^{-1}, etc. The presence of proven uranium deposits in Namtumbo District created the need for assessing radon levels in domestic water sources.To the best of the author's knowledge, this is the first study in Tanzania investigating radon levels in water.

MATERIALS AND METHODS

Description of study area: Namtumbo District is one of the five districts of the Ruvuma Region of Tanzania. It is bordered to the north by the Morogoro Region, to the east by the Tunduru District, to the south by Mozambique and to the west by the Songea Urban

District and Songea Rural District. According to the Population and Housing census of 2022, the population of the Namtumbo District was 271,368 (URT 2022). The inhabitants of Namtumbo district rely on water from various sources for domestic use. This study focused on springs, rivers and wells which according to the locals, these are the main sources of water supply for their daily domestic use.

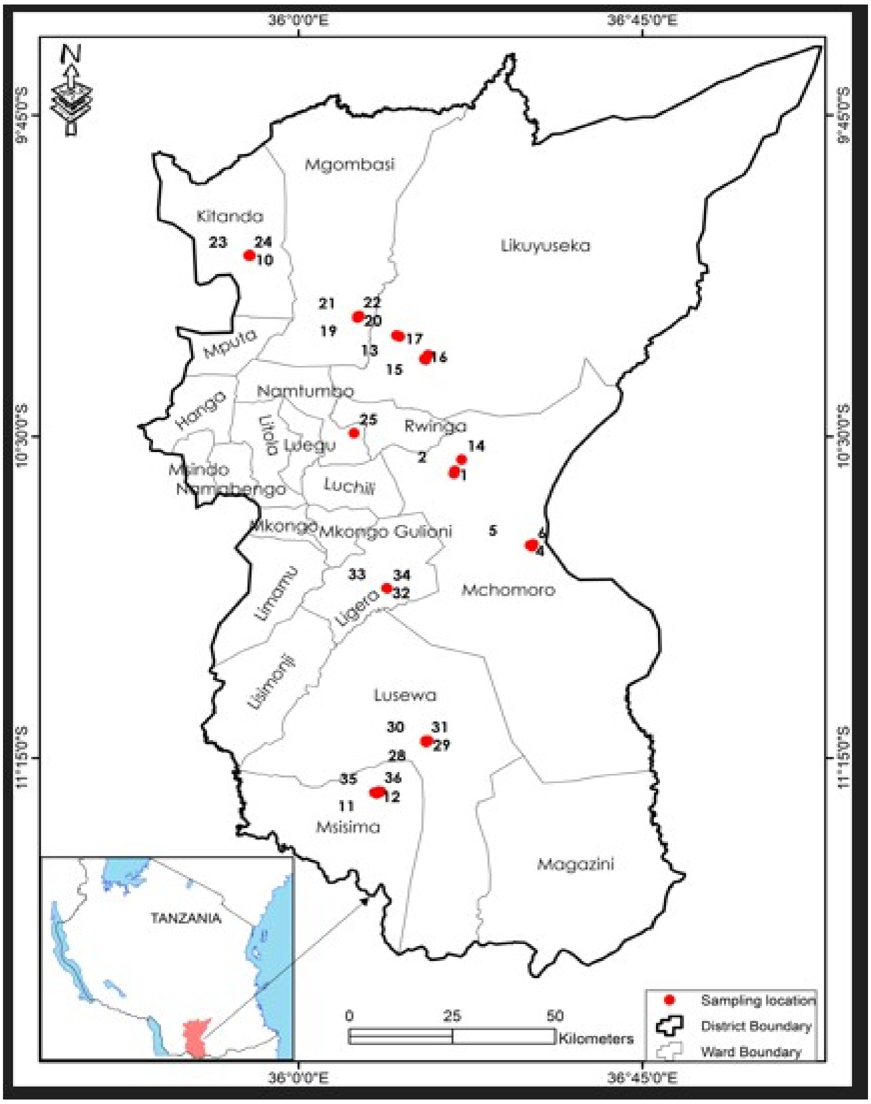

Figure 2. Map of Tanzania showing the location of Namtumbo District and sampling locations

Collection of water samples: 39 samples were collected from water sources that are used by the surrounding communities for different purposes including drinking and cooking. Water samples include twenty two (22) wells/boreholes, thirteen (13) springs and four (4) rivers. Water samples were packed in plastic bottles and sealed tightly to avoid escape of radon gas then transported to the temporary location within Namtumbo where instruments were set up for measuring radon in water. Sampling containers were fully filled so that no air bubbles were present after closing the container air-tight with the container cap.

Instrumental set up and measurements: The AlphaGUARD in combination with AquaKIT were used to determine radon directly in water samples. 100 ml of the sample to be measured were taken up with the supplied 100 ml plastic syringe and afterwards directly injected into the upper nozzle of the degassing unit. Afterwards the measurement operation was started. In a closed gas cycle, radon was expelled from the water sample by means of the pump commonly known as the AlphaPUMP. Measuring of the sample was continued for about 20 min. The pumped air is routed through the ionization chamber of the AlphaGUARD where the radon concentration is determined and stored in the memory (Saphymo 2017). Presented in Figure 3 is the set-up of the measuring system. The setup was rinsed with radon-poor air after measurements to prevent contamination. Detailed measuring steps are provided in the AquaKIT User Manual page 18.

Calculation of radon concentration in water samples: Determination of radon concentration in water samples is based on the radon concentration indicated on the radon monitor (Fig 3).

However, the displayed value is not yet the radon concentration in the measured sample because the radon driven out has been diluted by the air within the measurement set-up and a small part of the radon remains diluted in the watery phase. Taking into consideration the dilution effect of the interior volume in the measurement set-up, Equation 1 which is applicable to AlphaGUARD PQ2000Pro was used to quantify radon in water (Saphymol 2017).

$$RC_{water} = \frac{C_{air}(Bqm^{-3}) \times 1028}{1000} \quad (1)$$

Where C_{air} is the radon concentration indicated on the radon monitor

Estimation of annual effective dose from ingestion and inhalation of radon: Radon gas is the largest contributor to the collective exposition to natural radiation of the population in the world (Bonotto 2014, Jantsikene *et al.*2014, Ravikumar and Somashekar 2014) . The annual effective dose to an individual consumer due to intake of radon from drinking water was evaluated using the Equation 2 (Alam *et al*. 1999, Alharbi *et al*. 2015).

Table 1. Sources of water, radon concentrations in water (RC_{water}), annual effective dose due to ingestion (AED_{ing}), annual effective dose due to inhalation (AED_{inh}), annual effective dose to stomach (AED_{sto}), and annual effective dose to the lungs (AED_{lungs}). RC_{water} is reported in BqL^{-1} and the AEDs are reported in μSvy^{-1}

Sample No.	Source	RC_{water}	AED_{ing}	AED_{inh}	AED_{sto}	AED_{lungs}
1.	Spring	15.4	39.4	38.9	4.7	4.7
2.	Spring	15.7	40.2	39.6	4.8	4.8
3.	Spring	12.0	30.7	30.3	3.7	3.6
4.	Spring	11.7	29.9	29.5	3.6	3.5
5.	Spring	23.8	60.9	60.1	7.3	7.2
6.	Spring	16.8	42.8	42.2	5.1	5.1
7.	Spring	28.1	71.7	70.7	8.6	8.5
8.	Spring	86.0	219.8	216.8	26.4	26.0
9.	Spring	0.5	1.2	1.1	0.1	0.1
10.	Spring	13.5	34.4	33.9	4.1	4.1
11.	Spring	13.7	34.9	34.5	4.2	4.1
12.	Spring	0.6	1.5	1.5	0.2	0.2
13.	Spring	0.5	1.2	1.1	0.1	0.1
14.	River	0.2	0.6	0.6	0.1	0.1
15.	River	11.0	28.1	27.7	3.4	3.3
16.	River	0.5	1.4	1.4	0.2	0.2
17.	River	1.5	3.8	3.7	0.5	0.4
18.	Well	1.7	4.4	4.3	0.5	0.5
19.	Well	9.8	25.0	24.6	3.0	3.0
20.	Well	2.1	5.3	5.2	0.6	0.6
21.	Well	1.0	2.7	2.6	0.3	0.3
22.	Well	0.8	2.2	2.1	0.3	0.3
23.	Well	15.5	39.7	39.1	4.8	4.7
24.	Well	11.8	30.2	29.8	3.6	3.6
25.	Well	5.3	13.4	13.3	1.6	1.6
26.	Well	8.1	20.8	20.5	2.5	2.5
27.	Well	11.5	29.4	29.0	3.5	3.5
28.	Well	1.0	2.7	2.6	0.3	0.3
29.	Well	13.3	33.9	33.4	4.1	4.0
30.	Well	10.6	27.1	26.7	3.2	3.2
31.	Well	14.6	37.3	36.8	4.5	4.4
32.	Well	8.5	21.6	21.3	2.6	2.6
33.	Well	18.1	46.2	45.6	5.5	5.5
34.	Well	7.6	19.4	19.1	2.3	2.3
35.	Well	1.7	4.4	4.3	0.5	0.5
36.	Well	1.8	4.5	4.5	0.5	0.5
37.	Well	5.0	12.8	12.6	1.5	1.5
38.	Well	1.7	4.4	4.4	0.5	0.5
39.	Well	0.6	1.4	1.4	0.2	0.2

$$D_{ing}(\mu Svy^{-1}) = RC_{water} \times AWI \times CF \times 10^{-3} \quad \text{……… (2)}$$

where D_{ing} is the annual effective dose (μSvy^{-1}) due to ingestion of radio-nuclides from the consumption of water, RC_{water} is the concentration of ^{222}Rn in the ingested drinking water (BqL^{-1}), AWI is the annual intake of drinking water for an adult (Age >17 year) equals to $730Ly^{-1}$, and CF is the ingested dose conversion factor for ^{222}Rn equals to $3.5nSvBq^{-1}$ (Kumar *et al.* 2022, UNSCEAR 2000). The annual effective dose (AED_{inh}) following inbreathing of waterborne radon was estimated from Equation 03 (ICRP 1993, UNSCEAR 2000).

$$AED_{inh}(\mu Sv) = RC_{water} \times 10^{-4} \times 0.4 \times 7000 \times 9 \quad \text{……..(3)}$$

Where RC_{water} is the radon concentration in water (BqL^{-1}), 10^{-4} is the ratio of radon in air to water; 0.4 is the equilibrium factor between radon and its progenies, 7000 is the average number of hours that an individual can spend indoor per year, and $9nSvBq^{-1}h^{-1}m^3$ is the dose conversion factor for inhalation of radon.

Equation 4 was used to estimate the annual effective dose for lungs (AED_{lungs}) and stomach (AED_{sto}),

$$AED_{lung,sto}(\mu Sv) = W_T \times AED_{inh,ing} \quad \text{…….. (4)}$$

Where W_T is the tissue weighting factor equal to 0.12 for the stomach and lungs (ICRP 1991)

RESULTS AND DISCUSSION

Radon concentrations in water: Radon concentrations measured in water samples from Namtumbo District are presented in Table 1. From this table, it is evident that radon concentrations varied from 0.20 BqL^{-1} to 86 $BqL^{-1}$with a mean value of 10.3 BqL^{-1}. The highest value was measured in water sample number 14 collected from the Likuyuseka River flowing through Likuyu Mandela village, Likuyuseka ward. The lowest concentration was measured in well water (sample number 18) located in the same village. The highest concentrations in other water sources were 28.1Bq/L and 18.1Bq/L for springs and wells, respectively. The mean values for rivers, springs and wells were 28.1Bq/L, 11.3Bq/L and 6.8Bq/L, respectively.

These results show that the maximum and mean values from all water sources are well below the limit of 100 BqL^{-1}as set by the World Health Organization – WHO (WHO 2017). However, 45% of the values exceeded the USEPAs maximum contaminant level (MCL) of 11.1 BqL^{-1}set out in US EPA 1999. Although, radon levels in all samples are within the maximum level of 100Bq/L, spatial variations in radon concentration are evident in Table 1. Such variations could be attributed to geological structure of the area, soil type, depth of the water source, and also geo-hydrological processes that occurs in the area (Alharbi *et al.* 2015).

Table 2. Radon levels in groundwater from different parts of the world

Country	^{222}Rn concentrations (Bq/L)	Reference
Brazil	0.95 – 36.00	Marques, *et al*. 2004
Turkey	0.70–31.70	Yalim *et al*. 2007
Mexico	1.78–39.75	Villalba *et al*. 2005
Italy	1.80–52.70	D'Alessandro and Vita, 2003
India	11.7–381.2	Ravikumar and Somashekar 2014
Brazil	1.6–215	Corrêa *et al*.,2014
India	0.50–85.7	Rani, *et al*. 2013
Saudi Arabia	10-100	Shabana, *et al*.2013
Saudi Arabia	0.76- 4.69	El-Taher, 20212
India	17 – 68	Kumar *et al*. 2022
Kenya	0–371	Otwoma and Mustapha 1998
India	7.5–389.6	Sethy *et al*. 2015
Yemen	1–896	Abdurabu *et al*. 2016
Germany	9–1220	Trautmannsheimer, *et al*. 2002
Tunisia	0 – 2860	Faten *et al*. 2018.
Tanzania	0.2 – 86	This study

Levels of radon in water reported in different parts of the world are presented in Table 2 for comparison. From Table2, it is evident that the maximum radon concentrations in water were measured in Tunisia, German, Yemen and India exceeding the WHO limit of 100BqL^{-1} by a factor of 28.6; 12.2; 8.96 and 3.89 for Tunisia, German, Yemen and India, respectively. In the East African region, the reported maximum value for Kenya is 3.71 folds above the WHO limit.

CONCLUSIONS

Radon concentrations were determined in water samples collected from springs, rivers and wells in Namtumbo District Southern Tanzania, where commercially viable uranium deposits are found. Results revealed that radon concentrations in all samples lies well within the limit of 100 BqL^{-1}as set by the World Health Organization (WHO) but the USEPA limit of 11.1Bqm^{-3} was exceeded by some samples. The annual effective dose due to ingestion and inhalation of radon were far below the dose limit of 1mSvy^{-1} for the members of the public. Also, the mean values of effective doses per annum to the lungs and stomach as well as the estimated total annual effective doses were below the limit of 1mSvy^{-1}. These results suggest that radon levels in the sampled water sources are not of concern. Results can be used as baseline values prior to commencement of uranium mining and milling operations in Namtumbo District.

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