Natural radioactivity levels and evaluation of radiological hazards in Usakos marble, Erongo region, Namibia

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ABSTRACT

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Background: Most parts of the Erongo region of Namibia have shown high background radiation. The aim and objective of this study is to determine the natural radioactivity levels and evaluate the radiological hazards in Usakos marble dome in the Erongo region. Materials and Methods: A high purity germanium (HPGe) detector was used in this study to measure the activity concentrations ²²⁶Ra, ²³²Th, and ⁴⁰K in marble samples from 20 different points in Usakos marble dome in the Erongo region, Namibia. Result: The mean activity concentrations were found in the range of 0.37 to 339.60 Bq kg^{-1} for 226 Ra, 0.20 to 210.30 Bq kg^{-1} for 232 Th, and 2.96 to 928.70 Bq kg^{-1} for ⁴⁰K, respectively. The activity concentrations of ²²⁶Ra, ²³²Th, and ⁴⁰K were used to estimate values of radium equivalent activity (Ra_{eq}), Absorbed dose rate (Dr), internal and external hazard indices (Hex & Hin) and the activity utilization index (Iy). The calculated mean values were Ra_{eq} (226.75 Bg kg⁻¹), (Dr) (102.99 nGyh⁻¹), H_{ex}(0.61), H_{in} (0.95) and Iy (1.59). *Conclusion*: From a radiological point, the results of (Dr) and (Iy) were found above the world recommended average and the performed correlation analysis showed strong positive correlation amongst ²³²Th and ²²⁶Ra, which confirms these radionuclides as the main contributors to gamma radiation in the Usakos marble dome. The high activity concentrations at some sampling points higher than the world average value pose possible radiological hazards for the marble samples usage for building materials.

INTRODUCTION

Humans are continuously being exposed to ionizing radiations that originate from both natural man-made sources. Natural background radiation makes up about eighty-seven percent of the total doses received by human populations in a year (1). The main contributor to naturally occurring radioactive materials (NORM) are ²³⁸U, ²³²Th and ⁴⁰K (2,3). Although, the parent element, ²³⁸U does not pose adverse effects on the environment, the inhalation of its daughter nuclide 226Ra is known to carry a high degree of risk to human organs particularly the lungs resulting to lung cancer (3). Naturally occurring radioactivity in building materials is responsible for the internal and external radiation exposure of human populations living in dwellings made from such building materials (4-8). The distribution of natural radionuclides is not uniform hence, an understanding of their concentrations in human

dwellings is important to quantify human population exposure to NORM, since most humans spend 80-90% of their lifetime indoors (9-11).

Earlier studies have shown that the worldwide average indoor effective dose due to gamma rays originating from building materials is estimated to be about 0.4mSvy⁻¹ (12-13). The presence of natural radionuclides in building materials in an amount exceeding the internationally acceptable limit has received global attention (13, 14-17). The activity of radionuclides depends on geological and geographical setting as well as the geochemical characteristics of the materials (9, 18).

Many studies have investigated the radiological elements in different soil, sediment and rock samples. From a radiological point, the Erongo region in Namibia has attracted the attention of several authors (19-24). However, there exists little or no information on the radiological hazards associated with the marble deposits in the region. The

understanding of the activity concentrations of the marble deposits is needed to properly quantify the radiological hazards from their usage as building materials. The main aim of this study is to ascertain natural radioactivity levels and evaluation of radiological hazards in Usakos marble and to establish a reference line for further and future study on NORM in and around the Usakos, Erongo marble dome.

MATERIALS AND METHODS

Study area

Usakos is a mining town in the Erongo region of Namibia (figure 1). It is located at latitude 22° 00′ 0.00″S and longitude 15°35′59. Usakos has a population of 3,000 inhabitants and has a catchment area of 58 km². The town of Usakos, as with most towns in the Erongo region, experiences seasonal variation in rainfall. The wet season is from December to April with about 262 mm of rainfall in a year. The rest of the months are generally dry and hot with an average annual temperature of 26°.

Sampling and sample preparation

Twenty marble samples were collected, once from each of 20 locations 500m apart, at the study area during the year 2018. Initial labeling was done for easy identification. The samples were dried for 120 °C for 24 h, and then crushed using laboratory jaw crusher to 2mm particles, after which 1kg each of the sieved samples was weighed out. All the crushed samples were then carefully homogenized, after which the samples were transferred into a clean empty radon-tight marinielli beaker, marked, and sealed for 31 days to allow the daughter products to attain radioactive secular equilibrium with their parent radionuclides ²²⁶Ra and ²³²Th, after which they were counted for 53200 seconds.

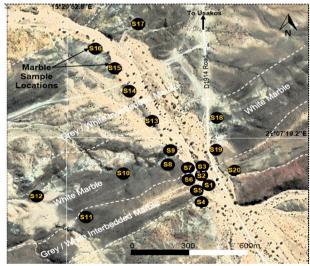


Figure 1. Marble sample collection sites from Usakos marble dome.

Experimental radiometric analysis

The counting of radionuclides present in marble samples was analyzed with a high-resolution gammaray spectrometer using a coaxial (62.80 X 64.80 mm) Canberra high purity germanium (HPGe) detector Model No. GC4520 SN 10882 with 45 % relative efficiency and a resolution of 2.00 keV full width at half maximum (FWHM) at 1.33 MeV peak of 60Co and 1.200 keV (FWHM) at 122 keV. The detector is shielded with 15 cm lead encasement to reduce the background radiation and cooled using liquid nitrogen. A computer-based Multichannel Analyser (MCA), Genie 2000 software from Canberra, was used for data acquisition and analysis of gamma spectra. Each marble sample was counted for 53200 s in a reproducible sample detector geometry, and the same configuration and geometry was used throughout the analysis. The gamma spectrometry system was energy and efficiency calibrated using a range of gamma-ray energies ranging from 0.060 MeV to 2 MeV mixed radionuclides standard in a 500 ml Marinelli beaker. This energy range was analysed for the absolute photo-peak efficiency and energy calibration of the HPGe detector using a multi-nuclide calibration standard with an initial activity of 40 kBq homogeneously distributed in silicone matrix, which was supplied by Eckert & Ziegler Nuclitec GmbH, Germany, SN. AM5599. The 295.22 keV, 351.93 keV for ²¹⁴Pb and 609.32 keV, 1120.29 keV and 1764.49 keV for 214Bi gamma lines were used in the assessment of activity concentration of 226Ra, while 911.21 keV for ²²⁸Ac and 968.97 keV and 238.63 keV for 212Pb were used for 232Th. The40K activity was obtained from the measurement of the single gamma line at 1460.8 keV. The background activity counting due to naturally occurring radionuclides in the room housing the detector was subtracted from obtained peak of each marble samples. Equation 1 was used to calculate the activity concentration A(C) in Bq kg-1 of the levels of the radioactivity of ²²⁶Ra, ²³²Th and ⁴⁰K found in each marble samples and the results are presented in table 1.

$$A(C) = \frac{C_{net}}{\epsilon(E) \times I_{\gamma} \times t \times m}$$
 (1)

Where C_{net} is the counting rate for a specific gamma line given in count per second corrected for background. $\epsilon(E)$ represent absolute photopeak efficiency, Iy is the intensity of gamma-ray line, t is the time for data collection in seconds and m is the mass of each sample in kg.

Radium equivalent activity (Ra_{eq})

The concentrations of radionuclides from the activity of 226 Ra, 232 Th and 40 K in marble samples are not uniformly distributed. The uniform distributions in respect of exposure to ionizing radiation can be written in terms of Radium equivalent dose (Raeq). The Raeq activity of the measured radionuclides is

used to make comparison of each 226 Ra, 232 Th and 40 K in the marble samples. The Ra_{eq} activity is measured in the unit of Bq kg⁻¹. It was calculated using equation 2.

$$Ra_{eq} = AC_{Ra} + 1.43AC_{Th} + 0.077AC_{k}$$
 (2)

Where AC_{Ra} , AC_{Th} and AC_k are the specific activity concentration of 226 Ra, 232 Th and 40 K respectively. The Ra_{eq} defines the weighted sum of each activity of 226 Ra, 232 Th and 40 K bearing in mind that the radionuclide 226 Ra and the index Ra_{eq} is 10 Bq kg⁻¹. For 232Th, Ra_{eq} is attributed with 7 Bq kg⁻¹ while for 40 K, Ra_{eq} it is 10 Bq kg⁻¹.

Absorbed dose rate D_r(ngy/h)

In this present study, the absorbed dose rates in air at 1m above ground was estimated using the formula in equation 3.

$$D_r(ngy/h) = 0.427A_{Ra} + 0.662A_{Th} + 0.043A_k$$
 (3)

Where

 A_{Ra} = specific activity concentration of ^{226}Ra A_{Th} = specific activity concentration of ^{232}Th A_{k} = specific activity concentration of ^{40}K

Radiological hazards

The contribution to gamma dose is from different radionuclides. It is therefore important to present radiological hazards as a single quantity. This quantity is known as hazard index, and is calculated according to the model proposed by Krieger ⁽²⁵⁾. The external and internal hazard indices are computed using equation 4 and 5 ^(12, 25-27).

$$H_{\text{ext}} = \frac{A_{\text{Ra}}}{370} + \frac{A_{\text{Th}}}{259} + \frac{A_{\text{K}}}{4810} \tag{4}$$

$$H_{\text{int}} = \frac{A_{\text{Ra}}}{185} + \frac{A_{\text{Th}}}{259} + \frac{A_{\text{K}}}{4810} \tag{5}$$

To keep gamma radiation dose minimal from building material, $H_{ext} \le 1$ (28-30).

Activity utilization index Iy

To measure the level of γ -radiation hazard from marble samples, another radiation level index called the activity utilization index is evaluated. The I γ , was suggested by OECD (16) and was evaluated using equation 6:

$$I\gamma = \frac{A_{R2}}{150} + \frac{A_{Th}}{100} + \frac{A_{K}}{1500}$$
(6)

For the safe use of the marble stone, evaluated values should be less than unity.

Statistical analysis

MS – Excel 2013 software was used for statistical analysis and the statistical test of analysis

of variance (ANOVA) and correlation analysis that were employed to test for statistical significance and correlation that exist amongst the identified radionuclides.

MATERIALS AND METHODS

The results in table 1 summarize the activity concentration that ranges from 0.37 to 339.60 Bg kg-1 for ²²⁶Ra, 0.20 to 210.30 Bq kg-1 for ²³²Th and 2.96 to 928.70 Bq kg⁻¹ for ⁴⁰K respectively. The activity concentrations were calculated from equation 1. The mean and standard deviation of the activity concentrations of ²²⁶Ra, ²³²Th and ⁴⁰K of all 20 marble samples collected from the Usakos marble dome were found to be 117.39±35.30 Bq kg⁻¹, 60.05±21.05 Bq kg-1 and 304.95±88.67 Bq kg-1 respectively. Equation 2 was used to determine Ra_{eq} which is based on the estimation that 370 Bq kg⁻¹ of ²²⁶Ra, 259 Bq kg⁻¹ of ²³²Th and 4810 Bq kg⁻¹ of ⁴⁰K each produce an identical gamma ray dose rate and their specific activities are presented in table 1 of column 6. The obtained values shows the minimum radium activity for Us-06 (grey) (0.88 Bq kg-1) and maximum associated with Us-15 (Black) (688.54) with a mean value of 226.75 Bq kg⁻¹. The absorbed dose rate (D_r) in air due to gamma radiation was calculated using equation 3 and the results were presented in table 2. The calculated D_r in unit of (nGy/h) due to the activity concentrations of ²²⁶Ra, ²³²Th and ⁴⁰K were found in the range of 0.42 to 311.15 nGy/h with a mean value of 102.99. The average value of Dr is found to be higher than the world recommended value of 55 nGy/h (12), implying that the measured absorbed dose rate due to the activity concentrations of ²²⁶Ra, ²³²Th and ⁴⁰K in air from the studied sites is 1.9 times the world allowed value. For building materials used for dwelling purposes to be acceptable, it is recommended that the external and internal hazard indices (Hext), (Hin) should have values less than unity (12, 31-33). The Hext and Hin were calculated using equation 4 and 5 and the obtained values presented in table 2 shows the external and internal hazards indices ranging between 0.00 to 1.86 for H_{ext} and 0.00 to 2.78 for H_{in} . The values of samples with codes Us-08, Us-09, Us-10, Us-15 and Us-19 were higher than the recommended value of unity for both H_{ext} and H_{in} respectively. However, sample Us-03 was higher than the value of unity for H_{in} but lower for Hext while Us-07 was lower for Hext and higher for H_{in} . The activity utilization index (I γ) was calculated using equation 6 and the results presented in column 6 of table 2. The Iy results have a minimum value of 0.00, maximum value of 4.78 and, mean value of 1.59. Clearly, the mean value of Iy was found to be higher than the recommended world average value of $<1^{(12)}$. The tested values of the correlation between the specific activities of 226Ra, 232Th, and 40K as shown in Figure 3, 4 and 5, showed weak positive correlations with coefficient ($R^2 = 0.1882$) for 40 K and 226 Ra, ($R^2 = 0.1123$) for 40 K and 232 Th, and a strong positive

Table 1. Activity concentrations of Radium-226 (²²⁶Ra), Thorium-232 (²³²Th), Potassium-40 (⁴⁰K) and radium equivalent activity (Raeq) in the marble samples from Usakos.

		Activity			
Codes	Colour	²²⁶ Ra	(Bq/kg) ²³² Th	⁴⁰ K	Ra _{eq} activity (Bq/kg)
Us-01	Grey	19.30	3.91	63.94	29.81
Us-02	Grey	45.04	37.72	11.53	99.87
Us-03	Grey	180.35	13.99	409.60	231.89
Us-04	Grey	17.06	8.02	128.4	38.42
Us-05	White	6.28	1.15	10.93	8.77
Us-06	grey	0.37	0.20	2.96	88.0
Us-07	grey	149.70	120.76	293.90	345.02
Us-08	grey	253.50	92.72	162.20	398.58
Us-09	White + grey	232.90	149.21	276.30	467.55
Us-10	Beige	179.35	201.53	430.50	500.69
Us-11	White + grey	75.11	26.73	366.70	141.57
Us-12	Beige	57.69	19.27	928.70	156.76
Us-13	Beige	80.14	23.86	200.00	129.66
Us-14	Beige	57.29	17.30	67.58	87.23
Us-15	Black	339.60	210.30	626.10	688.54
Us-16	Grey	71.68	28.27	540.10	153.69
Us-17	White +grey	63.55	20.78	307.80	116.97
Us-18	Grey	134.30	23.93	578.60	213.07
Us-19	Grey	322.90	186.23	485.90	626.62
Us-20	Grey	61.73	15.16	207.20	99.36
Minimum		0.37	0.20	2.96	88.0
Maximum		339.60	210.30	928.70	688.54
Mean±SE		117.39±3 5.30	60.05± 21.05	304.95± 88.67	226.75± 15.94
World's average (12) 32 30 420 370					

Where, Standard Error (SE) = is the standard deviation, N is the number of observation, Us = Usakos.

correlation with coefficient (R^2 = 0.7312) for 232 Th and 226 Ra respectively.

Table 2. Radiological parameters of Absorbed dose rate (Dr), External hazard index (Hex), Internal hazard index (Hin and Activity utilization index (AUI) in the marble samples from

	Colour	Absorbed	External	Internal	Activity	
Codes			hazard	hazard	utilization	
04		Dr (nGy/H)		index (H _{in}) 0.13		
Us-01	Grey	13.58	80.0		0.21	
Us-02	Grey	44.70	0.27	0.39	0.69	
Us-03	Grey	103.88	0.63	1.11	1.62	
Us-04	Grey	18.12	0.10	0.15	0.28	
Us-05	White	3.91	0.02	0.40	0.06	
Us-06	grey	0.42	0.00	0.00	0.00	
Us-07	grey	156.50	0.93	1.34	2.40	
Us-08	grey	176.60	1.08	1.76	2.73	
Us-09	White + grey	210.11	1.26	1.89	3.23	
Us-10	Beige	228.51	1.35	1.84	3.50	
Us-11	White + grey	65.54	0.38	0.59	1.01	
Us-12	Beige	77.32	0.42	0.58	1.20	
Us-13	Beige	58.62	0.35	0.57	0.91	
Us-14	Beige	38.82	0.24	0.39	0.60	
Us-15	Black	311.15	1.86	2.78	4.78	
Us-16	Grey	72.55	0.42	0.61	1.12	
Us-17	White +grey	54.13	0.32	0.49	0.84	
Us-18	Grey	98.07	0.58	0.94	1.52	
Us-19	Grey	282.06	1.69	2.57	4.33	
Us-20	Grey	45.30	0.27	0.44	0.70	
Minimum		0.42	0.00	0.00	0.00	
Maxi	mum	311.15	1.86	2.78	4.78	
	n±SE	102.99 ± 31	.0.61 ± 0.1	0.95 ± 0.1	l1.59±0.35	
World's average (12)		55	≤1	≤1	<1	

Where, Standard Error (SE) = $\,$ is the standard deviation, N is the number of observation and Us = Usakos.

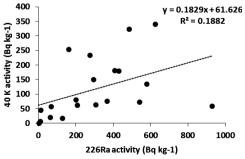


Figure 3. The correlation between ⁴⁰K and ²²⁶Ra activity.

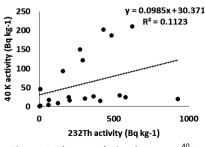


Figure 4. The correlation between ⁴⁰K and ²³²Th activity.

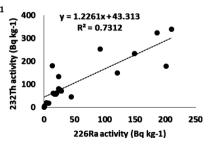


Figure 5. The correlation between ²³²Th and ²²⁶Ra activity.

DISCUSSION

The United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR-2000) proposed the acceptable values of activity concentrations of $^{226}\rm{Ra}$ (32 Bq kg-¹), $^{232}\rm{Th}$ (30 Bq kg-¹) and $^{40}\rm{K}$ (420 Bq kg-¹) (¹²²). A comparison of the mean activity concentrations of $^{226}\rm{Ra}$, $^{232}\rm{Th}$ and $^{40}\rm{K}$ were made with the world average recommended values as well as some studies carried out in other parts of the globe and the results are given in table 3.

It is clearly seen from Table 3 that the average values of 226Ra and 232Th in the collected marble samples were higher than the world's average values by a factor of 3.7 for 226Ra, and 2.0 for 232Th. The findings also showed that the activity concentrations of ²²⁶Ra and ²³²Th in this present study were higher than other published data in Algeria, Cameroon and Saudi Arabia (34-36) as well as some other studies conducted in other countries of the world such as Egypt, Kuwait, Nigeria, China, India and Brazil (37-42). The high concentrations of the radionuclides of ²²⁶Ra and ²³²Th measured in this study may be attributed to the geography and geological contents of the Erongo region of Namibia that is characterized by high background radiation (19-24). This is worrisome because high levels of activity concentrations in the marble samples may render the mineral rock radiologically unfit and hazardous for use in building and construction purposes.

However, the mean activity concentration for ⁴⁰K is lower than the world allowed value of 420 Bq kg⁻¹ by a factor of 0.73 ⁽¹²⁾ but higher than the reported values for the listed countries of Egypt, Kuwait, Nigeria, China, India and Brazil presented in table 3.

Table 3. Comparative average values for ²²⁶Ra, ²³²Th and ⁴⁰K of present work with world average and other countries.

of present work with world average and other countries.						
Country	²²⁶ Ra	²³² Th	⁴⁰ K	Ref		
Namibia, Usakos	117.39±	60.05±	304.95±	Present		
INdillibid, USAKUS	35.30	21.05	88.67	study		
UNSCEAR-2000	32	30	420	(12)		
Egypt	56.78±2.5	5.95±1.2	1.42±1.7	(37)		
Kuwait	3.9±0.5	0.22±0.08	3.7±0.5	(38)		
Nigeria	2	1	7	(39)		
China	8-157	6-166	44-1353	(40)		
India, Gujarat	12±3	3±2	10±3	(41)		
Brazil, Espirito Santo	3.0±0.3	2.2±0.6	18.0±0.1	(42)		

Ra = radium, Th = thorium and K = potassium

The Radium equivalent activity (Ra_{eq}) is a radiological parameter that is used to relate the external and internal doses due to radon and its progenies ⁽⁴³⁻⁴⁵⁾. The Ra_{eq} values from the marble dome site at Usakos, Erongo region were found to be lower than world recommended value 370 Bq kg⁻¹ ⁽¹²⁾, while exceeded the allowed values for sample sites with codes Us-08, Us-09 and Us-10 as well as some other sampling sites with codes Us-15 and Us-19. This indicates that the marble samples of

these five locations may have radiological implications if used for building purposes.

The mean absorbed dose rate was calculated and found to be higher than the world average value 55 nGy h⁻¹ by a factor of 1.87 (12). The average value in this study was also found to be higher than some countries of the world (34, 36) as well as other findings reported in literatures for countries like Nigeria and Cameroon (3, 35).

The radiological indices of internal hazards (H_{in}) and external hazards (H_{ex}) are important indices for the control of exposure originating from radon and its progenies that are carcinogenic in nature since the present health risk to the respiratory organs $H_{in}>1$ was observed in marble samples Us-03, Us-07, Us-08, Us-09, Us-10, Us-15 and Us-19. Also, for $H_{ex}>1$ was estimated for samples with codes Us-08, Us-09, Us-10, Us-15 and Us-19. Although the obtained mean value for H_{in} and H_{ex} were less than unity which is the recommended value $^{(25)}$, the high values recoded at these sites carries a high radiological risk from the use of the samples for construction and building purposes.

The activity utilization index (AUI)>1 was observed in samples Us-03, Us-07, Us-08, Us-9, Us-10, Us-11, Us-12, Us-15, Us-16, Us-18 and Us-19, with an a mean value greater than 1 being recommended worldwide (12,46).

The analysis of variance of the mean activity concentrations of ²²⁶Ra, ²³²Th, and ⁴⁰K (n=20) at p<0.05 are statistically significant (p=0.0000221) (Table 4). This suggests different lithogenic sources of inputs of the radionuclides recorded in the marble samples. According to M. Ngachina *et al.*, natural radionuclides are present in all rocks including marble in varying amounts depending on their concentration levels in the parent rock materials ⁽³⁵⁾.

The strong positive correlation amongst the radioactive nuclides and the associated variables at p<0.05 suggest that the marble samples from the Usakos marble dome are endowed with thorium and radium, and these elements significantly contribute to the gamma dose from the marble sampling sites. Also, the correlation analysis indicates that both thorium and radium originate from the same decay chain (44,47).

Table 4. Statistical significant test

Table 4. Statistical significant test.							
Groups	Count	Sum	Average	Variance			
Column 226Ra		2347.84	117.392	10632.2			
Column 232Th	20	1201.04	60.052	5185.693			
Column 40K	20	6098.94	304.947	59836.37			
ANOVA							
Source of Variation	SS	df	MS	F	P-value	F crit	
Between Groups	656255.4	2	328127.7	13.0116	2.21E-05	3.158843	
Within Groups	1437431	57	25218.09				
Total	2093687	59					

Between groups, ANOVA: single factor of paired means (n = 20) are statistically significant (p < 0.05).

CONCLUSION

From this study, the mean and range of the activity concentrations of $^{226}\rm{Ra},~^{232}\rm{Th},~$ and $^{40}\rm{K}$ were 117.39 (0.37-339.60), 304.95 (2.96-928.70) and 226.75 (0.88-688.54) Bq kg $^{-1}$ respectively. The mean values of Ra $_{\rm eq}$ activity was found to be less than the acceptable value of 370 Bq kg $^{-1}$. The mean value of absorbed dose rate is 102.99 nGy/h, which is higher than the world recommended value of 55 nGy/h. The mean values of external and internal hazards indices were below unity however, the mean value of activity utilization index was found to be higher than unity.

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