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

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ABSTRACT

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 ^{226}Ra, ^{232}Th, and ^{40}K 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 ^{40}K, respectively. The activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K were used to estimate values of radium equivalent activity (Ra_{eq}), Absorbed dose rate (Dr), internal and external hazard indices (H_{ex} & H_{in}) and the activity utilization index (Iγ). The calculated mean values were Ra_{eq} (226.75 Bq kg^{-1}), (Dr) (102.99 nGyh^{-1}), H_{ex}(0.61), H_{in} (0.95) and Iγ (1.59). ***Conclusion***: From a radiological point, the results of (Dr) and (Iγ) were found above the world recommended average and the performed correlation analysis showed strong positive correlation amongst ^{232}Th and ^{226}Ra, 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 and 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 ^{238}U, ^{232}Th and ^{40}K [(2, 3)]. Although, the parent element, ^{238}U does not pose adverse effects on the environment, the inhalation of its daughter nuclide ^{226}Ra 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 22^0 00’ 0.00’’S and longitude 15^{0}35’59. Usakos has a population of 3,000 inhabitants and has a catchment area of 58 km^2. 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 26^0.

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 ^{0}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 ^{226}Ra and ^{232}Th, after which they were counted for 53200 seconds.

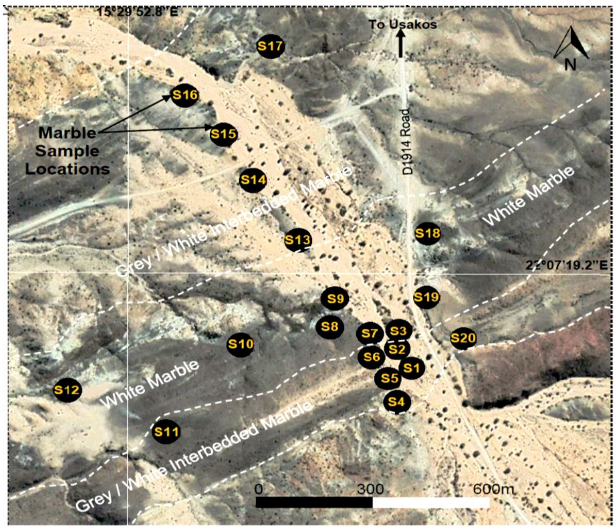

Figure 1. Marble sample collection sites from Usakos marble dome.

Experimental radiometric analysis

The counting of radionuclides present in marble samples was analyzed with a high-resolution gamma-ray 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 ^{60}Co 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 ^{214}Pb and 609.32 keV, 1120.29 keV and 1764.49 keV for ^{214}Bi gamma lines were used in the assessment of activity concentration of ^{226}Ra, while 911.21 keV for ^{228}Ac and 968.97 keV and 238.63 keV for ^{212}Pb were used for ^{232}Th. The^{40}K 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 ^{226}Ra, ^{232}Th and ^{40}K found in each marble samples and the results are presented in table 1.

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

Where C_{net} is the counting rate for a specific gamma line given in count per second corrected for background. ε(E) represent absolute photopeak efficiency, $I\gamma$ 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 (Ra_{eq}). The Ra_{eq} 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.

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

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.

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

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].

$$H_{ext}=\frac{A_{Ra}}{370}+\frac{A_{Th}}{259}+\frac{A_{K}}{4810} \quad (4)$$

$$H_{int}=\frac{A_{Ra}}{185}+\frac{A_{Th}}{259}+\frac{A_{K}}{4810} \quad (5)$$

To keep gamma radiation dose minimal from building material, $H_{ext} \leq 1$ [28-30].

Activity utilization index Iγ

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

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

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 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 ^{40}K respectively. The activity concentrations were calculated from equation 1. The mean and standard deviation of the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K 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 ^{226}Ra, 259 Bq kg^{-1} of ^{232}Th and 4810 Bq kg^{-1} of ^{40}K 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 ^{226}Ra, ^{232}Th and ^{40}K were found in the range of 0.42 to 311.15 nGy/h with a mean value of 102.99. The average value of D_r 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 ^{226}Ra, ^{232}Th and ^{40}K 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 (H_{ext}), (H_{in}) should have values less than unity [12, 31-33]. The H_{ext} and H_{in} 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 H_{ext} while Us-07 was lower for H_{ext} and higher for H_{in}. The activity utilization index (Iγ) was calculated using equation 6 and the results presented in column 6 of table 2. The Iγ results have a minimum value of 0.00, maximum value of 4.78 and, mean value of 1.59. Clearly, the mean value of Iγ 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 ^{226}Ra, ^{232}Th, and ^{40}K 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 correlation with coefficient (R^2 = 0.7312) for ^{232}Th and ^{226}Ra respectively.

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

		Activity Concentration (Bq/kg)			
Codes	Colour	^{226}Ra	^{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	0.88
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	0.88
Maximum		339.60	210.30	928.70	688.54
Mean±SE		117.39±35.30	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.

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 Usakos.

Codes	Colour	Absorbed dose rate Dr (nGy/H)	External hazard index (H_{ex})	Internal hazard index (H_{in})	Activity utilization index (AUI)
Us-01	Grey	13.58	0.08	0.13	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
Maximum		311.15	1.86	2.78	4.78
Mean±SE		102.99± 31	0.61± 0.1	0.95± 0.1	1.59± 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.

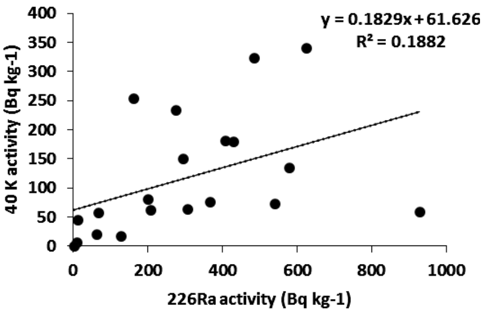

Figure 3. The correlation between ^{40}K and ^{226}Ra activity.

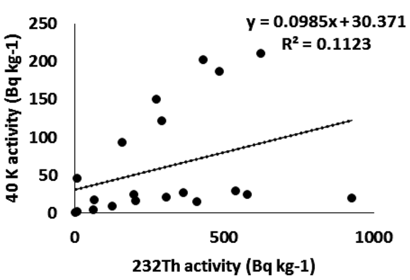

Figure 4. The correlation between ^{40}K and ^{232}Th activity.

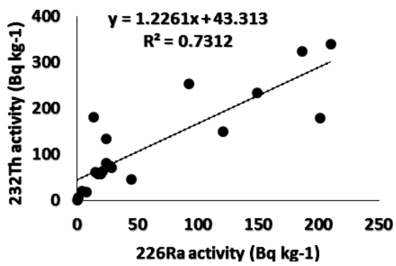

Figure 5. The correlation between ^{232}Th and ^{226}Ra activity.

DISCUSSION

The United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR-2000) proposed the acceptable values of activity concentrations of ^{226}Ra (32 Bq kg^{-1}), ^{232}Th (30 Bq kg^{-1}) and ^{40}K (420 Bq kg^{-1}) [12]. A comparison of the mean activity concentrations of ^{226}Ra, ^{232}Th and ^{40}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 ^{226}Ra and ^{232}Th in the collected marble samples were higher than the world's average values by a factor of 3.7 for ^{226}Ra, and 2.0 for ^{232}Th. The findings also showed that the activity concentrations of ^{226}Ra and ^{232}Th 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 ^{226}Ra and ^{232}Th 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 ^{40}K 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 ^{226}Ra, ^{232}Th and ^{40}K of present work with world average and other countries.

Country	^{226}Ra	^{232}Th	^{40}K	Ref
Namibia, Usakos	117.39± 35.30	60.05± 21.05	304.95± 88.67	Present 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 ^{226}Ra, ^{232}Th, and ^{40}K (n=20) at $p<0.05$ are statistically significant (p=0.0000221) (Table 4). This suggests diiferent 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.

Groups	Count	Sum	Average	Variance		
Column ^{226}Ra	20	2347.84	117.392	10632.2		
Column ^{232}Th	20	1201.04	60.052	5185.693		
Column ^{40}K	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}Ra, ^{232}Th, and ^{40}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_{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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