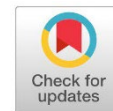


Research Article

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Assessment of Radiation Hazards Parameters Associated with Natural Radionuclides in Granite Used in Al-Bayda, Libya



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Abstract

The levels of natural radionuclides in granite samples used as building materials, collected from local markets in Al-Bayda, Libya, were investigated using a gamma ray spectrometer equipped with a NaI (Tl) scintillation detector. For ^{226}Ra , ^{238}U , ^{232}Th , and ^{40}K , the respective activity concentrations ranged from 40.96 ± 1.47 to $205.48 \pm 0.13 \text{ Bqkg}^{-1}$, 52.25 ± 1.53 to $182.65 \pm 2.36 \text{ Bqkg}^{-1}$, 49.72 ± 1.17 to $144.32 \pm 1.51 \text{ Bqkg}^{-1}$, and 49.41 ± 1.18 to $271.75 \pm 2.74 \text{ Bqkg}^{-1}$. According to the data, all granite samples exhibited radioactive levels for ^{226}Ra , ^{238}U , and ^{232}Th that exceeded the worldwide recommended limits set by UNSCEAR. Conversely, the activity concentration levels of ^{40}K in all granite samples were found to be below the UNSCEAR worldwide recommended values. The radiological hazard parameters associated with these natural radionuclides were subsequently evaluated. Comparison of the results with other global studies and world-recommended values revealed that while some parameters were lower than or within the recommended limits, others showed values higher than internationally accepted thresholds. Nevertheless, the findings indicate that for most of the studied granite samples, the radiation hazards from terrestrial radionuclides are within acceptable limits for their use as building materials.

Keywords: Natural Radioactivity; Granite Absorbed Dose Rate; Building Material; Annual Effective Dose.

INTRODUCTION

Natural radioactivity originates from terrestrial radioactivity and cosmic radiation. Humans primarily experience two types of exposure: internal exposure from inhaled radon-222 (^{222}Rn) gas and its decay products, and external exposure from gamma rays emitted by terrestrial radionuclides like potassium-40 (^{40}K) and the uranium (^{238}U) and thorium (^{232}Th) series (Sivakumar et al., 2014). Recent investigations in regions with high natural background radiation have raised awareness of risk assessment due to inhabitants' exposures to long-term low-level radiation (Akpanowo et al., 2020). These high radiation levels often stem from concentrated radio nuclides in granite rocks, soils, sediments, and other geological materials frequently used in construction and infrastructure (Abbasi, 2013). To accurately assess human exposure to natural radiation sources, it's crucial to understand public dosage limits and measure ambient background radiation levels from the ground, air, water, food, and within buildings (Kovacs et al., 2017). Consequently, information about the



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concentrations of these radio nuclides in the environment is fundamental for estimating the level of public exposure to ionizing radiations (Lee et al., 2019). Currently, there is limited information available concerning the radioactive levels of building materials in Libya. Therefore, determining the activity concentrations of these materials is necessary to evaluate potential radiological risks to building inhabitants (Hanfi et al., 2022). This study aims to determine the natural radioactivity in granite samples, commonly used as building materials in Al-Bayda, Libya, by utilizing a gamma spectrometer to measure the activity concentrations of ^{226}Ra , ^{238}U , ^{232}Th , and ^{40}K .

MATERIALS AND METHODS

Sampling Collected and Preparation

In this study, gamma-ray spectroscopy with a sodium iodide thallium-doped NaI (TI) detector with a "1.5×1.5" crystal, model No. PM-9266B, serial No. WA00012638. The detector was encased within a lead shield of sufficient thickness to minimize background radiation contributions from cosmic rays and ambient laboratory sources. Gamma spectra acquisition and subsequent analysis were performed utilizing the Cassy Lab software system. The activity concentrations of the ^{238}U and ^{232}Th decay series and ^{40}K were determined in 13 granite samples imported from India, which are commonly used as building materials and decorative materials in the city of Al-Beida, Libya. The solid samples were pulverized into a fine powder and passed through a standard 2 mm sieve, and then the samples were dried at a temperature of 110°C for two hours in an oven used to remove any moisture and achieve homogeneity. Before being analyzed using a gamma spectrometer, the samples were weighed and placed in 250 cm³ polyethylene containers, weighed, and stored for more than 30 days to reach equilibrium in the radioactivity between ^{238}U and ^{232}Th and their corresponding daughters. In order to ensure that the daughter remains in the sample and that the radon gas is contained inside the volume, this step is necessary. For gamma analysis, these samples were placed directly above the detector. The counting time for each sample was 7200 seconds. Figures 1) show gamma ray spectroscopy.



Figure: (1). The gamma spectroscopy system.

Calculations of Radioactivity Concentration Level

Concentration of activity (A) The rate at which an isotope decays is known as the radioactivity of a radioactive source. The quantity of radiation produced over time can be thought of as "radioactivity. Gamma spectroscopy measurements of each peak were used to determine the radioactivity levels of the different radionuclides that had been identified. The formula below was used to determine the associated activity (A) (Orosun et al., 2020).

$$A = \frac{(CPS)}{I \cdot \epsilon \cdot M} \quad (1)$$

Where: CPS: the energy-related net counts per second.

I: is the probability of gamma ray emission at the energy peak, ϵ : the absolute efficiency at photopeak energy, M: the sample's mass in kg, T: is the sample spectrum collection time (sec).

Calculations of Radiological Parameters

Radium Equivalent

The equivalent radioactivity, the radiation index evaluates a material's suitability for construction using the assumption that the gamma dose rates produced by 370 Bq kg⁻¹ of ²²⁶Ra, 259 Bq kg⁻¹ of ²³²Th, and 4810 Bq kg⁻¹ of ⁴⁰K are equal. To determine the radium equivalent activity, use the formula:

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

Where: A_{Ra} , A_{Th} , and A_K , represent the specific activity concentrations of ²²⁶Ra, ²³²Th, and ⁴⁰K, for safe use building materials should not exceed 370 Bq kg⁻¹ (Agora and Hashim, 2015; Ahmed Etal., 2022).

Gamma Radiation Level Index

Gamma radiation index I_γ is used to estimate the level of gamma radiation hazard associated with the natural radionuclides in building materials. It is identifying materials that may be hazardous to health when used for the construction of buildings. I_γ calculated using an equation based on (Mahmoud et al., 2020):

$$I_\gamma = \frac{A_{Ra}}{150} + \frac{A_{Th}}{100} + \frac{A_K}{1500} \quad (3)$$

Alpha Index

There is an association between alpha particle indices and radon inhalation from construction materials. The index of alpha is calculated using the relation (El-Feky et al., 2022):

$$I_\alpha = \frac{A_{Ra}}{200} \quad (4)$$

Internal Hazard Index

The internal radiation hazard index (H_{in}) provides an estimate of radon exposure and its daughter product, which is defined as (Alaboodi et al., 2020).

$$H_{in} = \frac{C_{Ra}}{185} + \frac{C_{Th}}{259} + \frac{C_K}{4810} \leq 1 \quad (5)$$

The External Hazard Index Hex

The external hazard index H_{ex} is the assessment of the hazard of γ -radiation. The H_{ex} values are detected via the following formula (Najam et al., 2015):

$$H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \leq 1 \quad (6)$$

Gamma Absorbed Dose Rate

The absorbed dose rate D_R (nGyh⁻¹) due to terrestrial gamma rays at 1 m above the ground according to the activity concentrations of ²²⁶Ra, ²³²Th, and ⁴⁰K in the granite samples, was

determined using the equation (Mansor et al., 2020):

$$D_R = 0.462 A_{Ra} + 0.604 A_{Th} + 0.042 A_K \quad (7)$$

The Annual Effective Dose Rate

In order to establish the annual effective dose rate in the air, the conversion coefficient between the absorbed dose in the air and the effective dose received by an adult must be considered. This value for environmental exposure to gamma rays with moderate energy is 0.7 SvGy^{-1} , according to UNSCEAR(2000). The occupancy factor for indoor measurements is about 0.8, as is the case for building materials, and the indoor annual effective dose equation becomes:

$$E_{in} (\text{mSvy}^{-1}) = D_R (\text{nGyh}^{-1}) \times 8760 (\text{hy}^{-1}) \times 0.8 \times 0.7 (\text{SvGy}^{-1}) \times 10^{-6} \quad (8)$$

There is about a 0.2 outdoor occupancy factor. Equation (9) gives the outdoor annual effective dose equivalent (Darwish et al., 2015)

$$E_{out} (\text{mSvy}^{-1}) = D_R (\text{nGyh}^{-1}) \times 8760 (\text{hy}^{-1}) \times 0.2 \times 0.7 (\text{SvGy}^{-1}) \times 10^{-6} \quad (9)$$

Excess Lifetime Cancer Risk (ELCR)

A person's risk of acquiring cancer increases with radiation exposure during their lifetime. ELCR was determined from the formula below :

$$ELCR = E_{out} \times DL \times RF \quad (10)$$

Where, according to the ICRP (2012), RF is a constant risk factor that is distributed to the community at a rate of 0.05 Sv (Taskin et al., 2009), DL is the life expectancy (70 years), and E_{out} is the outdoor annual effective dose equivalent (Yalcin et al., 2020).

RESULTS

The specific activity concentration values are recorded in Table (1). The values for radionuclides varied from 40.96 ± 1.47 to 205.48 ± 0.13 , 52.25 ± 1.53 to 182.65 ± 2.36 , 49.72 ± 1.17 to 144.32 ± 1.51 , and 49.41 ± 1.18 to $271.75 \pm 2.74 \text{ Bq kg}^{-1}$ for ^{226}Ra , ^{238}U , ^{232}Th , and ^{40}K , respectively, with an average of 119.88 ± 1.61 , 121.94 ± 2.49 , 97.17 ± 1.85 , and $169.97 \pm 2.46 \text{ Bq kg}^{-1}$, respectively, as shown in Figure (2).

Table (1): The specific activity concentrations (Bqkg^{-1}) of the radio nuclides in the investigated samples.

Sample No.	^{226}Ra	^{238}U	^{232}Th	^{40}K
G1	100.98 ± 1.74	86.00 ± 1.78	116.46 ± 1.36	126.27 ± 1.87
G2	40.96 ± 1.47	52.25 ± 1.53	55.39 ± 1.22	111.17 ± 1.76
G3	117.93 ± 1.80	116.95 ± 1.78	69.34 ± 1.13	131.76 ± 1.91
G4	134.17 ± 1.86	182.65 ± 2.36	63.93 ± 1.05	57.64 ± 1.27
G5	144.05 ± 1.90	94.22 ± 1.88	127.29 ± 1.56	52.15 ± 1.21
G6	136.99 ± 1.88	127.98 ± 2.06	112.78 ± 1.47	82.35 ± 1.52
G7	165.94 ± 1.98	156.7 ± 2.27	65.98 ± 1.14	49.41 ± 1.18
G8	127.81 ± 1.84	119.69 ± 2.04	90.61 ± 1.32	116.66 ± 1.80
G9	60.02 ± 1.56	72.87 ± 1.71	72.51 ± 1.83	111.17 ± 1.76
G10	135.58 ± 1.44	160.31 ± 1.38	144.32 ± 1.51	68.62 ± 0.71
G11	205.48 ± 0.13	182.24 ± 9.94	100.44 ± 7.39	223.71 ± 12.39
G12	122.87 ± 1.82	99.36 ± 1.99	105.30 ± 1.92	271.75 ± 2.74
G13	65.67 ± 1.59	94.32 ± 1.68	49.72 ± 1.17	126.27 ± 1.87
Average	119.88 ± 1.61	121.94 ± 2.49	97.17 ± 1.85	169.97 ± 2.46
P. L.	50	50	50	500

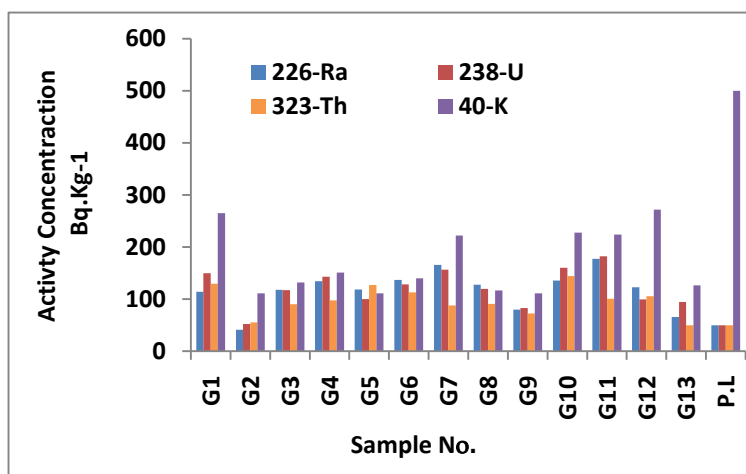


Figure (2). The specific activity concentrations of granite samples.

The average values of activity concentrations of radionuclides in the samples under study exceeded the globally recommended limit values of 50, 50, and 50 Bqkg⁻¹ for radionuclides ²²⁶Ra, ²³⁸U, and ²³²Th, respectively (UNSCEAR, 2010). However, the calculated mean value for ⁴⁰K was lower than the 500 Bqkg⁻¹ worldwide mean (UNSCEAR, 2010). The radiological hazards caused by exposure due to radionuclides were presented in Tables (2).

Tables (2): The radiological hazard parameters in investigation samples.

Sample No.	Hazard indices					D_R (nGy.h ⁻¹)	E_{in} (msv.y ⁻¹)	E_{out} (msv.y ⁻¹)	$ELCR$ ×10 ⁻³
	R_{aeq}	I_γ	I_α	H_{in}	H_{ex}				
G1	320.08	1.12	0.57	1.17	0.86	142.24	0.70	0.17	0.57
G2	128.72	0.45	0.20	0.46	0.35	57.04	0.27	0.06	0.23
G3	256.87	0.89	0.59	1.01	0.69	114.42	0.56	0.14	0.46
G4	284.98	0.98	0.67	1.13	0.77	127.12	0.62	0.16	0.51
G5	309.21	1.07	0.59	1.16	0.83	136.36	0.67	0.17	0.55
G6	309.04	1.06	0.68	1.20	0.83	137.29	0.67	0.17	0.55
G7	308.86	1.07	0.83	1.28	0.83	139.14	0.68	0.17	0.56
G8	266.37	0.92	0.64	1.06	0.72	118.68	0.58	0.14	0.48
G9	192.05	0.67	0.40	0.73	0.51	85.33	0.42	0.10	0.34
G10	359.50	1.24	0.68	1.34	0.97	159.38	0.78	0.19	0.64
G11	338.09	1.17	0.89	1.39	0.91	151.94	0.74	0.19	0.61
G12	294.37	1.03	0.61	1.13	0.79	131.78	0.65	0.16	0.53
G13	146.49	0.51	0.33	0.57	0.40	117.74	0.58	0.14	0.48
Average	269.43	0.93	0.59	1.04	0.72	124.49	0.60	0.13	0.50
P. L	370	1	1	1	1	84	1	0.07	0.29

The results of the R_{aeq} values calculated for the granite samples ranged from 128.72 to 359.50 Bq kg⁻¹, with an average value of 269.43 Bqkg⁻¹. The obtained values in this study are lower than the world-recommended value of 370 Bqkg⁻¹ (UNSCEAR, 2010). The values obtained of I_γ for samples ranged from 0.45 to 1.24, with an average value of 0.93. These results indicate that the gamma

radiation index exceeded the recommended value of 1 (UNSCEAR, 2010) for most samples. For I_α , the result ranged from 0.20 to 0.89 with an average value of 0.59. Consequently, the value of the internal hazard index ranged from 0.46 to 1.39, with an average value of 1.04. The values of most samples exceed the recommended limit of 1 (UNSCEAR, 2010), and some individual samples also showed values above this limit. Also, the values of the external hazard index ranged from 0.35 to 0.97, with an average value of 0.72. The results showed that H_{ex} values for all studied samples are lower than unity (UNSCEAR, 2010). On the other hand, the absorbed dose rate ranges from 57.04 to 159.38 nGyh⁻¹, but the average absorbed dose rate value for granite samples was 124.49 nGyh⁻¹. This average is higher than the world-recommended value of 84 nGyh⁻¹ (UNSCEAR, 2010), with the exception of sample G2, which showed a lower value. The values of the indoor annual effective dose varied from 0.27 to 0.78 mSvy⁻¹, with an average value of 0.60 mSv y⁻¹. The average values for all measured samples were less than the 1 mSvy⁻¹ limit (UNSCEAR, 2017, and Muiyiwa et al., 2020). The outdoor annual effective dose ranged from 0.06 to 0.19 mSvy⁻¹ with an average value of 0.13 mSvy⁻¹. These values of E_{out} are higher than the recommended value of 0.07 mSvy⁻¹ (UNSCEAR, 2010). The recorded values for ELCR range from 0.23 to 0.64 with an average value of 0.50. For all samples, these values were higher than the recommended world value of 0.29×10^{-3} (UNSCEAR, 2010). The current study will be compared with some previous studies, as shown in Tables (3)

Table (3): Comparison of radiological hazard indices in present work with those in other countries of the world.

Hazard index					D_R (nGyh ⁻¹)	Annual effective dose (mSvy ⁻¹)		$ELCR$ $\times 10^{-3}$	Ref.
Ra_{eq} (Bqkg ⁻¹)	I_γ	I_α	H_{in}	H_{ex}		E_{in}	E_{out}		
251.87	0.87	0.48	0.94	0.67	111.62	0.54	0.13	0.45	Present work
1016.92	8.29	-	3.03	2.75	534.42	-	-	-	(Sroor, 2013)
487	1.87	-	-	1.2	261	-	-	-	(Obaid et al., 2015)
176	0.67	0.19	0.58	0.48	160	-	-	-	(Imani et al., 2021)

CONCLUSIONS

This study systematically assessed the natural radioactivity levels in granite samples utilized as building materials in Al-Beida, Libya. The investigation revealed that while ⁴⁰K activity concentrations were below globally recommended values, the concentrations of ²²⁶Ra, ²³⁸U, and ²³²Th exceeded established acceptable limits. Regarding radiological hazard parameters, most samples exhibited higher than world-average recommended values for the gamma radiation index (I_γ), internal hazard index (H_{in}), absorbed dose rate, and lifetime cancer risk. Consequently, the outdoor annual effective doses for all studied granite samples were also higher than the world-recommended value. These results are crucial for enhancing the database on natural radioactivity in Libyan construction materials, which will facilitate further risk assessment and contribute to awareness efforts for mitigating hazardous material exposure.

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Author contributions: A.B and C developed theoretical formalism, performed the analytic calculations, and performed the numerical simulations. Both A.B and C. authors contributed to the final version of the manuscript. A.C. supervised the project.

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