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Quantification of Radiological Hazards Associated with Natural Radionuclides in Rock Samples from Al-Meshal Region, Libya

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ABSTRACT

The specific activity concentrations of primordial radionuclides in eight1samples of rock collected from specific locations in Al-Meshal, Al-Gabal Al-Akhdar region, Libya, were measured using a1gamma-ray1spectrometer system with a sodium iodide detector. The results revealed that the specific activity1concentrations of 226Ra, 238U, and 232Th in the analyzed-1samples were higher than their respective established permissible levels. Conversely, the specific activity1concentrations of 40K in all rock samples were found1to be below the permissible limit. The assessment of the radiological1hazard parameters, including radium equivalent activity, internal and external hazard1indices, and representative level index, indicated values were mostly within recommended world limits. However, for two specific samples, RE1 and RW2, the representative level index and internal hazard index values were higher than the recommended world1value. Although the radiological investigation indicates no immediate acute health hazard to the area's population, there is a need to consider the long-term health effects related to low-level exposure to radiation.

KEYWORDS: Natural1radionucles, radiological hazards, representative1level index, Pearson's correlation coefficients.

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1. INTRODUCTION

Human are continuously exposed to natural adionucides. The primary sources of exposure to naturally occurring radionuclides include terrestrial sources, such as the uranium and thorium decay series and potassium-40, as well as cosmic rays. Terrestrial background radiation, in particular, is influenced by the global distribution of radionuclides, local geological formations, geographical location, altitude, and regional geochemistry [1]. The advancement of the nuclear industry and other technologies involving radioactive isotopes has underscored the necessity of quantifying natural background radionuclide levels. This is crucial for discerning anthropogenic contamination, thereby enabling effective measures for population and environmental protection [2]. Human exposure to radionuclides occurs from external and internal exposure through both direct and indirect pathways. External exposure is caused by direct gamma radiation, whereas internal exposure results from inhalation of radioactive radon gas (radon-222) and its short-lived decay products [3]. Radon, a daughter element resulting from the decay chain of uranium, has the strongest effect on humans, as it poses a significant threat of lung cancer to individuals with prolonged exposure to it [4]. Recognizing the potential health implications, and understanding radiation levels within living and occupational environments is paramount [5]. Therefore, many researchers worldwide are quite interested in measurements of natural radioactivity in rock. To evaluate the terrestrial gamma dose rate for outdoor, it is very important to estimate the natural radioactivity level in rock. From the review of the literature, it is observed that though the data on the content of radioactive elements is available for some regions in Libya, there is no data available for the Al-Meshal region in Libya. Detailed investigations of natural radioactivity have been carried out for the first time [6]. This study presents the activity concentration levels and associated radiological hazard parameters in rock samples collected from Al-Meshal, Libya. In the context of Libya, several studies have investigated natural background radiation levels. Rock samples in the Gondola region reported similar dominant contributions from the uranium and thorium decay series [7]. Furthermore, investigations in the eastern region of Libya on rock samples around El-Beida City high-

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lighted the influence of local geology on the distribution of natural radionuclides, a factor also considered pertinent to the Al-Meshal area [8]. While specific data on rock samples from Al-Meshal might be limited, the general understanding of radionuclide distribution in Libya suggests that the levels are influenced by the country's diverse geological formations, similar to global trends. This research on Al-Meshal contributes to the growing body of knowledge on natural radioactivity in Libya.

2. MATERIALS AND METHODS

2.1. Study Area

Al-Meshal is a rural agricultural region situated in northeastern Libya, geographically located at coordinates 32° 28' 46.7" N, 21° 40' 42.2" E. The area is characterized by fertile soil, conducive to both agriculture and livestock husbandry.

The primary economic activity in this region is livestock rearing, which significantly contributes to local agricultural output. Consequently, understanding the environmental characteristics of this area, including its soil, rocks, and vegetation (grasses), is of considerable importance. Figure (1) illustrates the geographical location of the study area, as visualized using Google Earth and GPS data.

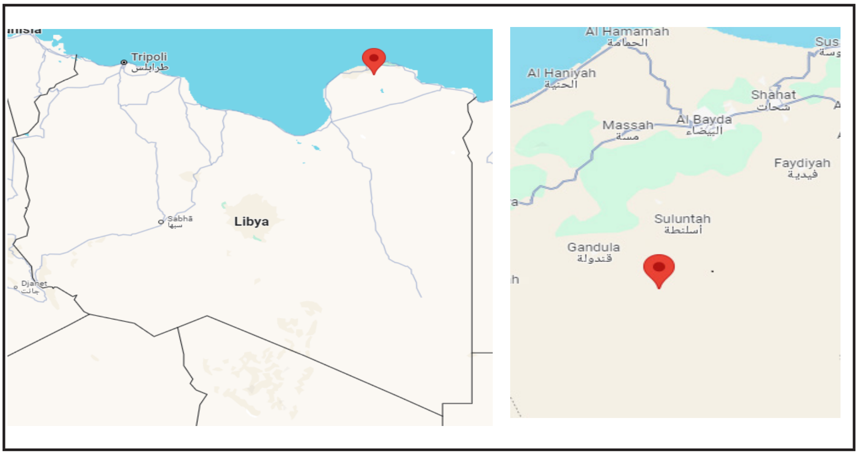

Figure.(1). Study region and locations of rock sample sites.

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2.2. Sample Collection and Preparation

The Al-Meshal area contains fourteen former weapons storage facilities. In this study, one of these facilities was selected as the sampling site. Eight rock samples were collected from this site, specifically from the four cardinal directions (east, west, north, and south) at distances of one kilometer and two kilometers. This sampling strategy was consistently applied in each direction. The unique identifiers (codes) assigned to each rock sample are detailed in Table (1).

Table (1). The code of the investigation samples.

.Sample No	Description
RE1	(Rocks from the first kilometer (East
RE2	(Rocks from the second kilometer (East
RN1	(Rocks from the first kilometer (North
RN2	(Rocks from the second kilometer (North
RS1	(Rocks from the first kilometer (South
RS2	(Rocks from the second kilometer (South
RW1	(Rocks from the first kilometer (West
RW2	(Rocks from the second kilometer (West

Sample preparation was conducted at the Advanced Nuclear Physics Laboratory, Omar Al-Mukhtar University, Libya. The collected samples were pulverized into fine powder. The rock samples were sieved using a 250 µm sieve aperture to ensure maximum homogeneity and to mitigate potential underestimation of radionuclide concentrations in bulk samples. Subsequently, the powdered samples were dried in a laboratory oven at approximately 120 °C for 2 hours to eliminate any residual moisture. Following the drying process, the samples were precisely weighed and transferred into polyethylene containers with a standardized volume of 250 cm^3. All prepared samples were then hermetically sealed and stored for a minimum period of 30 days to allow for the establishment of secular radioactive equilibrium between the ^{238}U and ^{232}Th decay series and their respective short-lived daughter radionuclides prior to gamma-ray spectroscopic analysis.

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2.3. Gamma Spectroscopy1Analysis

The concentrations of naturally occurring radionuclides in the collected samples were determined using gamma-ray-spectroscopy with a sodium iodide thallium-doped NaI(Tl) 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 were determined indirectly by quantifying the gamma emissions of specific short-lived daughter radionuclides, assuming secular equilibrium within the respective decay chains. Specifically, ^{238}U was quantified through the photopeaks of ^{214}Pb (at 295 and 352 keV) and ^{214}Bi (at 609, 768.2, 1120, 1378, and 1768 keV). Similarly, ^{232}Th was quantified using the photopeaks of ^{228}Ac (at 92, 209, 338, and 911 keV), ^{212}Bi (at 727 keV), and ^{208}Tl (at 583 keV). The activity levels of ^{40}K and ^{226}Ra were directly measured at their energies of 1460 keV and 186.2 keV [9]. For each sample, the gamma-ray spectrum was acquired over a counting time of 7200 seconds to ensure adequate statistical precision.

2.4. Measurement of Activity Concentration

The specific activity of a radionuclide is defined as the rate of its nuclear disintegration. It can be specified with the aid of equation [10]:

$$C = \frac{(CPS)}{I_\gamma \xi . M} \quad ---- \quad (1)$$

Where:

C: represents the activity concentration of the radionuclide corresponding to the gamma-ray photopeak energy, expressed in (Bq/kg).

Cps: denotes the net count rate detected at the specific gamma-ray photopeak energy, measured in counts per second.

ξ: signifies the counting system efficiency at the specific gamma-ray photopeak energy, which is dimensionless.

M: is the mass of the measured sample, expressed in (kg).

I_γ: is the probability of the absolute gamma-ray emission for the radionuclide of in-

$$Ra_{eq} = C_{Ra} + 1.34\, C_{Th} + 0.077\, C_K \quad -------(2)$$

terest, which is dimensionless.

3. RADIOLOGICAL HAZARDS

3.1. Radium Equivalent

A common radiological index used to represent the gamma radiation dose rate arising from various combinations of ^{226}Ra, ^{232}Th, and ^{40}K present in a sample indicates the radium equivalent activity, Raeq. It is calculated using the following relationship[11]:

$$H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \le 1 \quad --------(3)$$

Where C_{Ra}, C_{Th} and C_K are the activity concentrations (Bq/kg) for ^{226}Ra, ^{232}Th and ^{40}K, respectively. The maximum value of Ra_{eq} must be less than 370 Bq/kg [12].

3.2. External Hazard Index

The radiological parameter used to assess the external gamma radiation exposure risk arising from the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in a material is the external hazard index (H_{ex}). It is computed using the following relationship [13]:

3.3. Internal Hazard Index

Exposure to ^{222}Rn and its short-lived decay progeny presents several health risks. The alpha-emitting radionuclides, specifically radon (^{222}Rn), thoron (^{220}Rn), and their respective decay products present in ambient air are the primary contributors to the radiation-induced health hazards experienced by populations exposed to environmental radioactivity. The internal hazard index (H_{in}) is calculated using the following equation [14]:

$$H_{in} = \frac{C_{Ra}}{185} + \frac{C_{Th}}{259} + \frac{C_K}{4810} \le 1 \quad -------(4)$$

3.4. Representative Level Index

The representative level index (RLI) is a dimensionless radiological parameter that quantifies the gamma radiation dose rate arising from the specific activity concentrations of primordial radionuclides 226Ra,

$$RLI = \frac{C_{Ra}}{300} + \frac{C_{Th}}{200} + \frac{C_k}{3000} \leq 1 \quad ---- \; (5)$$

232Th, and 40K present within a material. This index is very important for the quality control of regulating annual effective doses to the population and monitoring overall radiation exposure to the human body. Its primary purpose is to ensure that internationally established acceptable dose limits are not exceeded. The RLI is calculated using the following equation [15]:

4.RESULTS AND DISCUSSION

The activity concentration values for rock samples varied from (72.03±0.13) to (117.93 ± 0.13) Bq/kg, (78.91 ± 5.84) to (120.52 ± 5.18)] Bq/kg, (71.10 ± 4.85) to (112.81 ± 6.21) Bq/kg and (301.95 ± 14.39) to (465.27 ± 17.87) Bq/kg for- ^{226}Ra, ^{238}U, ^{232}Th and ^{40}K respectively, with an average (84.92, 91.17, 84.31 and 388.30) Bq/kg respectively, for radionuclides ^{226}Ra, ^{238}U,^{232}Th and ^{40}K, as shown in Table (2).

Table (2). Activity concentrations of natural radionuclides for rock.

Sample No.	^{226}Ra	^{238}U	^{232}Th	^{40}K
RE1	117.93 ±0.13	120.52 ±5.18	102.60 ±6.11	465.27 ±17.87
RE2	73.44 ±0.13	87.98 ±6.63	71.10 ±4.85	428.22 ±17.14
RN1	73.44 ±0.13	82.93 ±6.59	78.78 ±4.77	458.41 ±17.74
RN2	72.03 ±0.13	79.18 ±5.86	76.76 ±4.40	303.32 ±14.43
RS1	77.68 ±0.13	79.03 ±5.53	74.79 ±5.31	409.00 ±16.75
RS2	76.97 ±0.13	78.91 ±5.84	72.73 ±4.74	301.95 ±14.39
RW1	106.63 ±0.13	114.8 ±85.35	112.81 ±6.21	430.09 ±13.15
RW2	81.21 ±0.13	86.00 ±6.03	84.92 ±5.35	310.18 ±14.59
Average	84.92	91.17	84.31	388.30
P. L	50	50	50	500

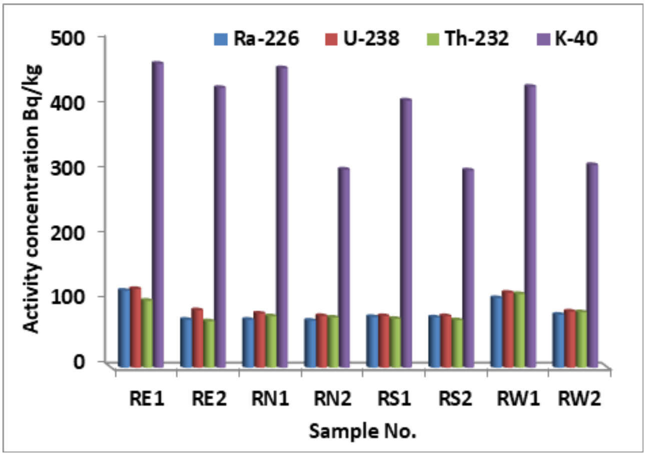

Figure.(2): Specific activity (Bq/kg) concentration of radionuclides for rock Samples.

The specific activity concentrations of all rock samples were higher than the permissible level for ^{226}Ra, ^{238}U, and ^{232}Th (50, 50 and 50 Bq/kg) [16]. However, the activity concentration of ^{40}K was lower- than the permissible level (500 Bq/kg) [16].

Radium equivalent activity (Ra_{eq}), internal hazard index (H_{in}), external hazard index (H_{ex}), and representative level index (RLI) were determined based on the specific activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K; the results are presented in Table (3).

Table (3). The radium equivalent, hazards indices and representative level index of rock samples.

Sample No.	$Ra_{eq\ (Bq/kg)}$	H_{in}	H_{ex}	RLI
RE1	300.47	1.13	0.81	1.06
RE2	208.09	0.76	0.56	0.74
RN1	221.09	0.79	0.59	0.79
RN2	205.14	0.75	0.55	0.72
RS1	216.13	0.79	0.58	0.77
RS2	204.21	0.76	0.55	0.72
RW1	301.12	1.10	0.81	1.06
RW2	226.53	0.83	0.61	0.80
Average	235.35	0.85	0.64	0.83
P. L	370	1	1	1

Statistical analysis revealed that Ra_{eq} values ranged from 204.21 to 300.47 Bq/ kg, with an average value of 235.35 Bq/ kg. For all inspected rock samples, Ra_{eq} values are lower than the assigned international allowed limit of 370 Bq/ kg [16]. The H_{in} varied between a minimum of 0.75 and a maximum of 1.13, with an average value of 0.85, which is below the established recommended value [16]. The values of H_{in} ranged between 0.51 to 0.81, with a mean of 0.64. The obtained values of H_{in} for most rock samples are less than unity [15]. The representative level index (RLI) values were found to be lower than the permissible limit of 1 for most samples [16], spanning from 0.72 to 1.06, with a mean of 0.83. Although samples RE1 and RW1 exhibited elevated radiological hazard indices, overall, all samples fell within the acceptable ranges for radiological hazard limits, consistent with established global values [16].

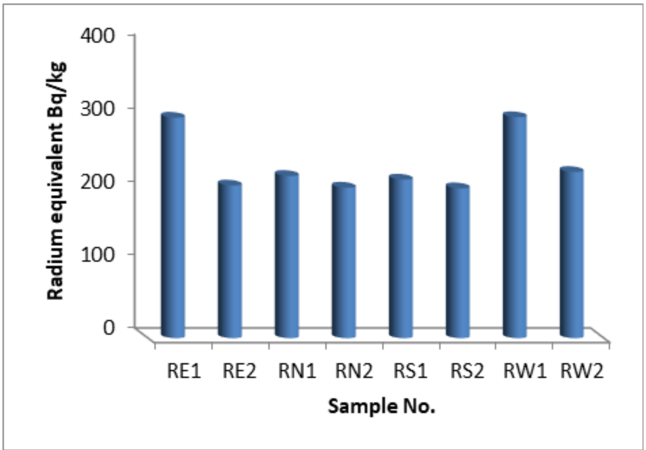

Figure. (3). Radium equivalent activity levels in rock samples.

The results showed that all1values of H_{ex} and RLI were lower than1unity. All values of internal hazard for the1studied1rock1samples are lower than unity, with the exception of RE1 and RW1, which were higher than the recommended value [1], as shown in Figure. (4).

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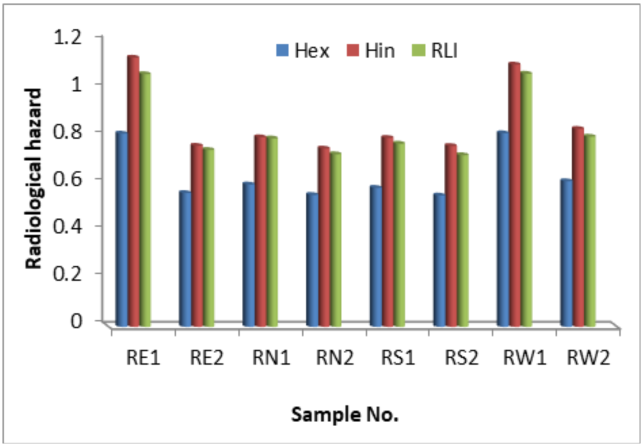

Figure. (4). A comparison between internal, external gamma Indices and the representative level index.

4.1. Pearson's Correlation Coefficients Analysis of Radioactive Variables

Pearson correlation coefficient analysis was employed to determine the pairwise mutual relationships and the strength of linear association between the studied parameters, utilizing bivariate statistics. The results of the Pearson correlation coefficients between all measured radionuclides and radiological parameters for the rock samples are presented in Table 4. A strong positive correlation coefficient was observed between ^{232}Th and ^{238}U, which is attributable to the thorium and uranium decay series in natural geological formations [17]. A moderate positive correlation coefficient was noted between ^{40}K and both ^{238}U and ^{232}Th. This is expected, as ^{40}K originates from a distinct decay scheme. Furthermore, radium equivalent activity (Ra_{eq}), the external hazard index (H_{ex}), the internal hazard index (H_{in}), and the representative level index (RLI) exhibited strong positive correlation coefficients with ^{238}U and ^{232}Th, and moderate positive correlation with ^{40}K.

Table (4). Pearson correlation matrix among the radionuclides and radiological parameters.

	^{226}Ra	^{238}U	^{232}Th	^{40}K	Ra_{eq}	H_{ex}	H_{in}	*RLI*
^{226}Ra	1							
^{238}U	0.97	1						
^{232}Th	0.91	0.92	1					
^{40}K	0.47	0.57	0.40	1				
Ra_{eq}	0.97	0.97	0.97	0.55	1			
H_{ex}	0.97	0.97	0.97	0.55	0.99	1		
H_{in}	0.98	0.98	0.96	0.53	0.99	0.991	1	
RLI	0.97	0.97	0.97	0.57	0.99	0.99	0.99	1

4.2. The Clark Values

The Clark values for the rock samples and the elemental concentrations (in ppm) of the studied radionuclides at each sampling location are presented in Table (5). The majority of the analyzed locations exhibit a Th/U ratio follows aligns along a linear trend with an average value of 2.83, which is lower than the reported recommended Th/U ratio of 3.5 [18]. The findings of the present study suggest a relatively low thorium content in the rock samples investigated from the Al Meshal region.

Table (5). Clark values and radionuclides ppm contents.

Sample No.	RE1	RE2	RN1	RN2	RS1	RS2	RW1	RW2
^{238}U ppm	9.72	7.09	6.69	6.38	6.37	6.37	9.26	9.93
^{232}Th ppm	25.27	17.51	19.40	18.91	18.42	17.91	27.79	20.92
^{40}K %	1.49	1.37	1.46	0.97	1.31	0.96	1.37	0.99
Clark Values	2.60	2.47	2.90	2.96	2.89	2.81	3.00	3.02

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5.CONCLUSION

The natural radioactivity levels in rock samples collected from various locations in Al-Meshal, Al-Gabal Al-Akhder, Libya, were evaluated using gamma-ray spectrometry. The average activity concentrations of ^{226}Ra, ^{232}Th, and ^{238}U were higher than the world-recommended values identified by UNSCEAR [16]. The average values of radium equivalent activity (Ra_{eq}) of the analyzed rock samples were lower than the recommended value of 370 Bq/kg, according to UNSCEAR [16]. The external hazard index (H_{ex}), internal hazard index (H_{in}), and representative level index (RLI) values were generally lower than the recommended maximum values set by UNSCEAR [16], except for the H_{in} and RLI values for two specific samples RE1 and RW1, which were lower than unity. Based on these findings, it is recommended that a periodic assessment of the activity concentrations of natural radionuclides and associated radiological hazards in the Al-Meshal study area be implemented. This proactive monitoring is essential to ensure that radiation exposure levels are maintained, thereby safeguarding public health and the environment in this region of Libya.

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