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Natural Radioactivity in Carbonate Rocks Outcropping at Al-Lajjun Region, South Jordan by Gamma-Ray Spectroscopy

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Abstract

Soil samples were collected from the Al-Lajjun area near the dam and analyzed for activity concentrations of primordial radionuclides using gamma-ray spectrometry. A low background configuration pure germanium (HPGe) detector was used for this purpose. The mean activity concentrations for ^{40}K, ^{226}Ra (^{238}U), ^{232}Th, ^{238}U, and ^{235}U at the location of Al-Lajjun Dam at Al-Karak Governorate in central Jordan were found to be in the range of 12-560, 234-6037, 4.3-28.4, 459-54147, and 43102-3656 Bq/Kg, respectively. Some of the obtained values were much higher than the world average values, except for ^{40}K and ^{232}Th.

To assess the radiological risk, radium equivalent activity, absorbed dose rate, and annual effective dose equivalent were calculated. The external hazard value (Hex) ranged from 0.638 to 16.316, which indicates significant activity in certain locations. In this study, the concentration of ^{238}U was found to be much higher than the acceptable value reported by UNSCEAR-2000. Therefore, cautionary measures with intense and continuous monitoring should be implemented for the dam site and water. Therefore, cautionary measures with intense and continuous monitoring should be implemented to the dam site and water.

Keywords: Gamma-ray spectrometry; Annual effective dose; External hazard index; Natural radioactivity; ^{238}U; ^{226}Ra; ^{232}Th; ^{40}K

Introduction

Radioactivity is a natural occurrence found in the environment. The most common terrestrial radio elements that produce gamma rays are ^{238}U, ^{232}Th, and ^{40}K. The level of natural environmental radioactivity varies depending on geological lithology and structure and appears at different levels in the soils of overlain various geological regions. Primordial radionuclides, such as ^{40}K, and radioisotopes from ^{238}U, and ^{232}Th series, along with their products such as ^{226}Ra are the primary sources of natural radioactivity pollution. It is also the largest source of radiation that humans are exposed to.

Researchers from various parts of the world have used a high-purity germanium detector (HPGe) to measure the activity concentration of gamma-emitting radionuclides in various soil and rock samples collected from different locations. Because Gamma-ray spectrometry is a fast, simple, and non-destructive method that can be used to collect data for many radionuclides at once. Furthermore, Gamma spectrometry is commonly used to determine the levels of ^{238}U, ^{232}Th, ^{226}Ra, and ^{40}K in environmental samples due to its simple sample preparation process. These samples were taken from rivers, coastal beaches, phosphate mines, types of cement, and medical plants. Samples of soil studies conducted in different countries using gamma spectroscopy can

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be found in the references and their sources [1-10].

Several studies have been conducted using gamma spectroscopy to measure natural and artificial radioactivity in the environment. Researchers from Jordanian universities and research centers have studied various regions of Jordan, from the north to the south and east to west. They have even analyzed different sources such as irrigation water, drinking water, milk products, dust, cement, and phosphate.

Abusini et al. conducted a study to measure the specific activity of naturally occurring radionuclides such as ^{238}U, ^{232}Th, and ^{40}K in soil cores collected from six different locations in Araba Valley using gamma-ray spectrometry measurements. They also measured the concentrations of various chemical oxides in the collected samples. The study's main objective was to establish a baseline map for the area. This map would serve as a reference to assess any changes in the radioactivity background level due to the alteration in the area's topography, development, and settlements around it, or any artificial environmental influences [11].

The aim of the study conducted by Zaid et al. was to determine the concentrations of gamma-emitting radionuclides present in sediment cores collected from different water depths along the Jordanian coast of the Gulf of Aqaba. Core sediments were collected from five representative locations at three different water column depths (5, 15, and 35 m). The results showed that the activity concentrations of ^{238}U, ^{235}U, and ^{226}Ra for both seafloor and beach sediments from the phosphate loading berth location were higher than those from other investigated locations and more than twice as high as the worldwide average [12].

A study was conducted by Malkawi et al. to determine the level of background radiation present in soil samples collected from the campus of Jordan University of Science and Technology (North of Jordan). The study involved selecting sampling locations around the proposed site of the Jordan Research and Training Reactor using a random systematic approach. The collected samples were analyzed using a high-purity Germanium detector coupled with GENIE 2000 spectroscopy software [13].

Farag studied the concentration levels of naturally occurring radioactive materials (NORMs) of ^{238}U, ^{232}Th, and ^{40}K in surface soil samples from selected areas in Zarqa, Jordan. Using gamma-ray spectrometry, the soil samples were analyzed [14].

In roadside soil cores collected from eight sites along the Amman-Aqaba National Highway, Al-Jundi et al. ascertained the specific activity and the gamma-absorbed dose rates of terrestrial naturally occurring radionuclides (^{238}U, ^{232}Th, and ^{40}K) [15]. Ababneh et al. investigated the radioactivity levels in soil and vegetables in the northern Jordan Rift Valley and estimated the corresponding doses [16].

Al-Bedri et al. evaluated the concentration levels of naturally occurring radioactive materials of ^{238}U and ^{232}Th series and ^{40}K in surface soil samples of an old phosphate mine at Russaifa, Jordan. The activity concentrations of naturally occurring radioactive materials in soil samples were determined through a gamma-ray spectrometry system using a hyper-pure germanium (HPGe) detector in a low background configuration [17].

Saleh and Abu-Shayeb investigated the natural radioactivity in Maan soil from five main distribution sites. They estimated and evaluated the radiological health hazard by collecting soil samples [18]. Manal et al. examined the cement properties and potential radiological risks of natural radioactivity in types of cement used in Jordan [19].

Hamarneh et al. studied surface and core soil samples collected from different regions of Jordan. Also, determination of the natural radioactivity was carried out, employing a gamma-ray spectrometry system, in surface soil samples collected from various geological formations in urban areas of the northern highlands of Jordan. Furthermore, ^{226}Ra, ^{238}U, ^{232}Th, and ^{40}K radionuclides have been determined using gamma-ray spectrometry in soil samples collected from urban areas in the southern governorates of Jordan [20-22].

The researchers Abu-Saleem et al. have conducted a study where they measured the activity concentrations of artificial radionuclides (^{134}Cs, ^{137}Cs, and ^{60}Co) and natural radionuclides (^{40}K) in eighteen surface soil samples from different locations around the Jordan Research and Training Reactor. To do this, they used a gamma-ray spectrometer that was based on a high-purity germanium detector [23].

An extensive study was conducted by Al-Omari et al. in Jordan to measure the activity concentrations of natural and artificial radionuclides including ^{226}Ra, ^{232}Th, ^{40}K, and ^{137}Cs in soil samples from each governate. A high-purity germanium detector was used to measure a total of 370 samples [24].

Please refer to Table 3 for additional information and results of the studies and consult the references listed therein for further details [11-24].

Recently, a review study of the dams in Jordan threats was done by [25], they considered the natural threats to affect many of these dams and the Al-Lajjun dam was among them.

The main aim of this is study is to analyze the levels of radioactive elements, such as radium, thorium, and potassium, in the Al-Lajjun region of Jordan. The results will be used for health risk assessment and to prepare the industry to produce electricity using nuclear reactors. Additionally, we will investigate the Al-Lajjun region to complete the survey of environmental radioactivity in Jordanian areas, particularly in a region full of different types of minerals to learn about its various radiation elements. The samples were collected from a region near Al-Lajjun Dam, which is used as a source of

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water for irrigation and animal husbandry. Also, the study aims to help the local community by identifying any potential risks and taking necessary precautions. In future work, the team should collect vegetable and water samples from different locations in the region for further investigation.

Study area settings:

The sampling site is exactly at the left abutment of the Al-Lajjun Dam, which is in Eastern part of Al-Karak governorate, on Wadi Al-Ghobyan, approximately located 1 km from the Amman-Al-Karak main street, and about 4 km to the north of Karak Industrial Estate, with coordination: 31o13'55''N 35o51'00''E, as shown in (Figure 1). The study area is dominated by a semi-arid climate and classified as a marginal area because it has less than 250 mm/year annual rainfall [26]. Therefore, it has low vegetation cover that only pushes for grazing; it is not populated only a few temporary Bedouins and Shepherds are seasonally live there. The main firm in the area is Al-Lajjun dam, which is an aggregate dam, it is 140 m long and 26m high, with a storage capacity of 1MCM, with a 0.13 km^2 lake area, as it has a limited catchment area of about 46 km^2.

The geology of Al-Lajjun Dam site begins with Amman Silicified Limestone (ASL) of Campanian age with 30-55 m thickness, consisting of bedded brown-black chert that is interbedded with limestone, marl, granule phosphatic chert, and 8m of oyster-coquina limestone. Overlain by the Al-Hisa Phosphorite (AHP) formation of Campanian-Maastrichtian with a 40–6 m thickness of interbedded phosphatic chert, granular phosphatic limestone, micritic limestone, and marl, the upper part of the thickness range is 2–15 m composed of phosphatic chert, phosphatic limestone, marly limestone, and marl. At the surface, outcrops the alluvial deposits of Pleistocene–Holocene age [27,28]. The surficial yellowish uranium mineralization was found mainly within the AHP formations as veins along joints and as encrustation in marly

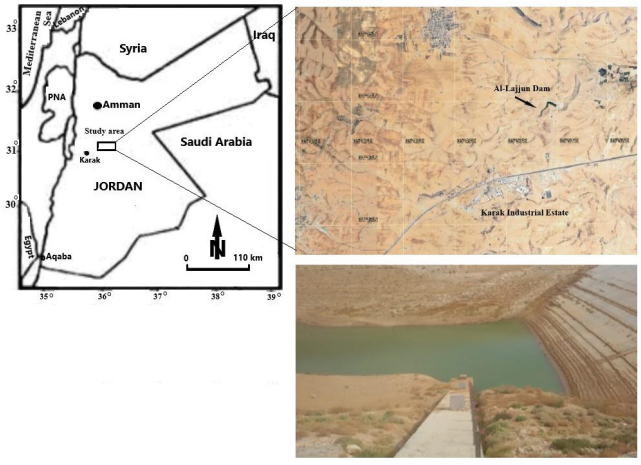

Figure 1: Study area location map; showing the Al-Lajjun Dam, Eastern region of Al-Karak governorate, with a photo of Al-Lajjun Dam upsream view.

limestone, phosphatic chert, marl, and phosphatic limestone layers of the upper part of AHP as illustrated in Figure 2. These deposits were outcrops due to the dam excavation works.

Experimental Procedure

Sampling

The studied samples are rock samples were collected from outcropped Upper Cretaceous carbonate rocks from the left abutment of the Al-Lajjun dam, from the eastern region of Al-Karak province (Figure 1). The samples were selectively collected based on U-mineralization signs or encrustation (Figure 2). The collected samples were properly labeled and sent to the laboratory for processing. Once collected, the samples were crushed into fine powder using a mortar and pestle, then sieved sieve with a small mesh size to obtain high-quality samples. The mass of each soil sample was accurately measured using a 0.001-gram balance and then weighed, packed, and sealed in an airtight PVC container. Before experimentation, the collected samples were saturated for up to 40 days to ensure that the parent nuclides were in secular equilibrium with their daughter's nuclides.

Measurement of Natural Radioactivity

The activity was measured using a high-resolution gamma spectrometry system and a high-purity germanium (HPGe) detector. GAMMA-X (GMX) N-type coaxial HPGe detector is designed for high-performance gamma spectroscopy in the energy range of 3 keV to 10 MeV. The GMX detector is a type of coaxial Germanium (Ge) detector that has an extremely thin entrance window. The detector's entrance window is a 0.3-µm-thick ion-implanted contact that extends the useful energy range to around 3 keV. The ion implantation process results in a stable contact that does not deteriorate with repeated cycling. Additionally, the N-type HPGe detector is resistant to fast neutron damage and is shielded by lead on all sides to minimize system background. (The detector is manufactured by EG&G, ORTEC, Oak Ridge, USA).

Figure 2: U-mineralization yellow encrustations along the joints of the marly limestone layers of AHP (Upper Cretaceous).

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Cryogenic cooling is required for germanium semiconductor radiation detectors. Therefore, a liquid nitrogen system is used. The system is calibrated to produce a gamma spectrum of intensity (number of counts/s) versus energy in MeV before experiments. Four sources were used to calibrate the Multi-Channel Analyzer (MCA): 60Co (1173.24 keV and 1332.50 keV), 137Cs (661.67 keV), 133Ba (356.01 keV), and 241Am (59.54 keV). The detector has a resolution of 2.0 keV and a relative efficiency of 20% for the 1.332 MeV gamma energy of 60Co. The output of the detector was analyzed using a 4K multichannel analyzer system connected to a PC. The gamma spectra were obtained using Genie 2000 spectroscopy software (supplied by Canberra, 2004) after collecting data for 86400 seconds to minimize statistical errors.

Theory and Evaluation

To determine the concentration of radionuclides such as ^{40}K, ^{235}U, ^{226}Ra, ^{232}Th, and ^{238}U, we analyzed the gamma peaks in the obtained spectra. For the ^{40}K activity concentration, we used its direct gamma line (1461.8 keV, yield = 10.67%). To calculate the radium (^{226}Ra) activity concentration, we considered the photo peaks of its daughter isotopes, ^{214}Bi (energy line 609.3 keV, yield = 46.3%) and ^{214}Pb (energy line 351.9 keV, yield = 37.2%), as provided by:

$$A(^{226}_{88}Ra) = \left(\frac{A_{Bi}}{\sigma^2_{Bi}} + \frac{A_{Pb}}{\sigma^2_{Pb}}\right) \Big/ \left(\frac{1}{\sigma^2_{Bi}} + \frac{1}{\sigma^2_{Pb}}\right) \quad (1)$$

where A is the activity concentration and σ is the uncertainty in A. The activity concentration of ^{238}U equals the obtained value of ^{226}Ra [10,29]. The concentration of activity (A) of ^{232}Th can be determined by analyzing the photo peaks of its daughter isotopes, namely ^{228}Ac (with line energy of 911.60 keV and yield of 27.7%), 208Tl (with line energy of 583.0 keV and yield of 30.9%), and ^{212}Pb (with line energy of 238.62 keV and yield of 44.6%).

$$A(^{232}_{90}Th) = \left(\frac{A_{Ac}}{\sigma^2_{Ac}} + \frac{A_{Tl}}{\sigma^2_{Tl}} + \frac{A_{Pb}}{\sigma^2_{Pb}}\right) \Big/ \left(\frac{1}{\sigma^2_{Ac}} + \frac{1}{\sigma^2_{Tl}} + \frac{1}{\sigma^2_{Pb}}\right) \quad (2)$$

when they are in secular equilibrium, the activity concentration of ^{232}Th can likewise be estimated from the activity of ^{228}Ra. Finally, the activity concentration of ^{235}U is calculated using the photo peak line (186.2 keV, yield = 57.2%) that coincides with the ^{226}Ra photo peak line (185.7 keV, yield = 3.28%).

$$A(^{235}_{92}U) = A(^{226}_{88}Ra\ at\ 186keV) - A(^{226}_{88}Ra) \quad (3)$$

Figures 3 and 4 display the gamma spectrum obtained for samples 4 and 6, respectively. In Figure 2, the highest number of counts per second is 20000 counts/s, while in Figure 4, it is 2000 counts/s, with the activity being one-tenth of the first. This difference could be attributed to varying uranium concentrations in some of the collected samples. Table 1 summarizes the findings obtained from the different samples. One sample collected from the Aqaba region in the south of Jordan falls within the acceptable range of activity concentration, indicating a normal case.

The acquired data show that we have high activity concentration levels in numerous samples compared to the global average concentrations. The Aqaba sample demonstrates the typical activity. To verify the accuracy of the results. Also, we sent the samples to the Jordan Atomic Energy Commission (JAEC) for further examination using the RID-SOP-006/Gamma ISO 18589-3 method. Even though different values were obtained by the two methods, our calculations were supported by the results presented in Table 2 from JAEC. Only the sample of Al-Lajjun-6 shows ^{234}Th with A=36312±500 and ^{137}Cs with A=6.65±3.35 Bq/Kg. The results are compared with previous results obtained from other scholars from different regions in Jordan in Table 3. There is a large activity concentration in some of the studied samples concerning previous studies.

The external hazard index (*Hex*) is given by a model proposed by Krieger (1981):

$$H_{ex} = \left(A^{226}Ra/370\right) + \left(A^{232}Th/259\right) + \left(A^{40}K/4180\right) \quad (4)$$

H_{ex} must not exceed the limit of unity for the radiation hazard to be negligible. On the other hand, the internal hazard index (Hin) gives the internal exposure to carcinogenic radon and its short-lived progeny. Is is given by the following formula (Beretka and Mathew, 1985):

$$H_{ex} = \left(A^{226}Ra/370\right) + \left(A^{232}Th/259\right) + \left(A^{40}K/4180\right) \quad (5)$$

The values of H_{in} must also be less than unity to have negligible hazardous effects of radon and its short-lived progeny on the respiratory organs [32]. The gamma absorbed dose rate in the air out the doors, D at 1 m above the ground surface due to specific activity concentrations of ^{238}U, ^{232}Th, and ^{40}K is defined as given in equation (6), to the effective dose received by adults. However, D values were calculated by the following equation [32]:

$$D(nGy\ h^{-1}) = 0.462A^{238}Ra + 0.604A^{232}Th + 0.0417A^{40}K \quad (6)$$

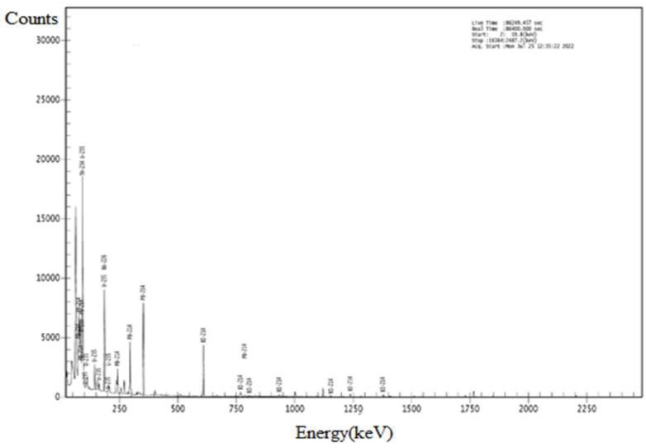

Figure 3: Gamma spectrum of sample #4.

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Table 1: The activity concentration A (Bq/kg) and the activity uncertainty of the collected samples of the radionuclides (^{40}K, ^{226}Ra, ^{238}U, ^{235}U) calculated by Genie 2000.

Sample	A(^{40}K)	A(^{214}Bi)	A(^{214}Pb)	A(^{226}R)=A(^{238}U)	A(^{235}U)
Al-Lajjun2	38.923±0.001	1073±10	1467±11	1250±7	472±11
Al-Lajjun3	23.581±0.010	205±3	270±3	234±2	102±3
Al-Lajjun4	-	-	16040±115	-	2782±54
Al-Lajjun5	-	5526±13	7502±21	6037±11	3656±69
Al-Lajjun6	559.770±0.002	3160±23	4057±21	3653±15	463±11
Al-Lajjun7	110.39±0.003	309±4	610±8	377±4	243±13
Al-Lajjun8	60.731±0.001	1985±12	2770±12	2378±8	1161±23
Al-Lajjun9	11.828±0.002	1418±7	1818±8	1583±5	357±8
Aqaba	54.3±2.5	229.3±4	280.8±3.80	249.9±2.6	43.1±2.2
Range	12-560	205-3160	270-16040	234-6037	102-3656
World average concentrations	400	-	-	35	-

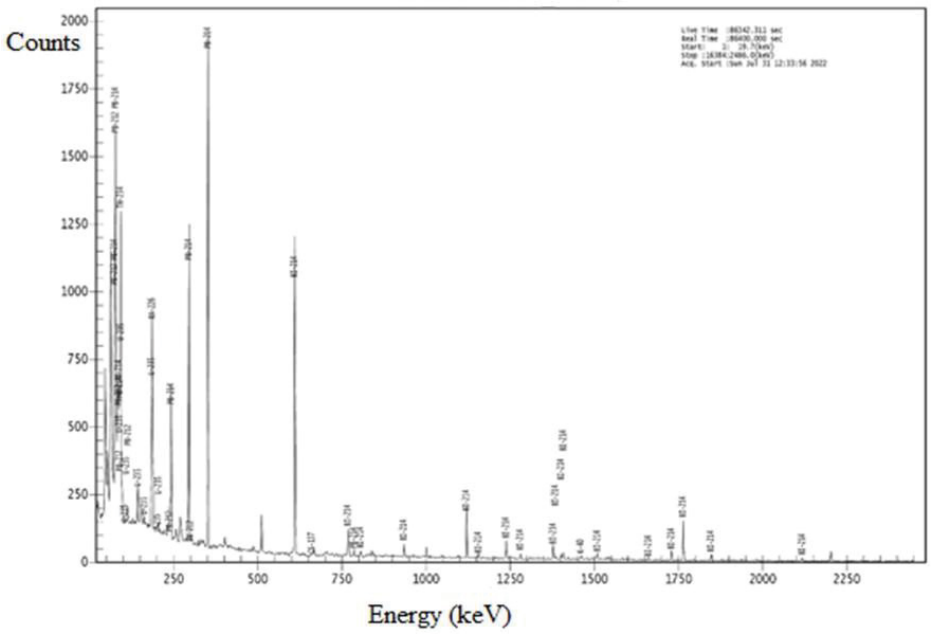

Figure 4: Gamma spectrum of sample #6.

Table 2: Results of activity concentration and the activity uncertainty obtained by Jordan Atomic Energy Commission A^{238}U.

No	Sample ID	Client ID	Radionuclide	Activity Concentrations Bq/kg
1	D2G2ST011	Al-Lajjun 2	^{238}U	4472±220
2	D4G2ST012	Al-Lajjun 3	^{238}U	1518±86
3	D2G2ST013	Al-Lajjun 4	^{238}U	34270±1526
4	D4G2ST014	Al-Lajjun 5	^{238}U	32164±1426
5	D2G2ST015	Al-Lajjun 6	^{238}U	817±52
6	D4G2ST018	Al-Lajjun 7	^{238}U	2020±134
7	D4G2ST016	Al-Lajjun 8	^{238}U	11638±542
8	D2G2ST017	Al-Lajjun 9	^{238}U	2531±131

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where $A^{238}U$, $A^{232}Th$ and $A^{40}K$ are the activity concentration of ^{238}U, ^{232}Th and ^{40}K in Bq/kg, respectively. Exposure Radiation is defined in terms of many parameters. Radium equivalent activity (Raeq) is a widely used hazard index. It is calculated, as given by equation (7), assuming that 370 Bq/kg of ^{226}Ra, 259 Bq/kg of 23kg, and 4810 Bq/kg of ^{40}K produce a gamma-ray dose rate (Beretka and Mathew, 1985):

$$Ra_{eq}\left(\frac{Bq}{kg}\right) = A^{226}Ra + 1.43A^{232}Th + 0.077A^{40}K \quad (7)$$

where $A^{226}Ra$, $A^{232}Th$ and $A^{40}K$ are the activity concentration of ^{226}Ra, ^{232}Th, and ^{40}K in Bq/kg, respectively. Note the total value must be equal to or less than 370.

Annual Effective Dose Equivalent (AEDE) due to the activity of the samples was calculated using equation (8) [14]

$$AEDE(mSvy^{-1}) = D(nGyh^{-1}) \times 8760(hy^{-1}) \times 0.2 \times 0.7(SvGy^{-1}) \times 10^{-6} \quad (8)$$

where the value 0.7 SvGy-1 is the conversion coefficient from the absorbed dose in the air to the effective dose received

Table 3: Activity concentrations in Bq/Kg in different studied regions in Jordan.

	Activity Concentration Bq/Kg (The average value of the studied Nuclei)					
Element / Region	**^{40}K**	**^{226}Ra(238U)**	**^{235}U**	**^{232}Th**	**^{238}U**	**^{137}Cs**
Zarqa 2016	212.87	211.44		11.10		
Russaifa 2014	207.10	265.95		0.895		
The Northern Jordan Rift Valley 2009	156.0	33	2.2	11.2		3.5
Maan 2014	138.1	57.7,44.9		18.1		
The types of cement in 2022	354.70	79.52		30.99		
Irbid 2013 Jordan University of Science and Technology	312.39	20.84		24.45	83.88	2.43
Amman Aqaba Highway 2003	560			82	84	
Urban areas in the southern governorates of Jordan 2018	233	39		23	45	
Urban areas of the Northern Highlands of Jordan 2009	291	42.5		26.7	49.9	
Jordan Research and Training Reactor 2018	340.3					2.94-25
Each governate of Jordan 2019	309	42		23		3.7
Araba Valley, Jordan 2008	94-762			14.3-35	19-38.7	
The Aqaba Gulf 2010		626			57 - 677	
Worldwide rang**	140-850	17-60		11-64		
Worldwide Median Value*	400	35		30		
Present Research	12-560	234-6037	43-3656	4.3-28.4	459-54147	6.65

Table 4: *Hex*, *Hin*, and the annual effective dose equivalent.

Sample	**Raeq (Bq/Kg)**	**D (nGy/h)**	**AEDE (mSv/y)**	***Hex***	***Hin***
Al-Lajjun2	1253.21	579.13	0.71	3.388	6.767
Al-Lajjun3	235.67	109.11	0.134	0.638	1.27
Al-Lajjun 4	-------	------	--------	--------	------
Al-Lajjun5	6036.86	2789.09	3.421	16.32	32.63
Al-Lajjun6	1643.56	1728.19	2.119	10.12	19.99
Al-Lajjun7	385.36	178.64	0.219	1.045	2.063
Al-Lajjun 8	2382.24	1101.18	1.35	6.434	12.87
Al-Lajjun9	1582.91	731.84	0.898	4.281	11.21

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by adults, 8760 is the time in hours in one year, 0.2 represents the outdoor occupancy factor [32] and is the observed dose rate. The calculated values are given in Table 4:

Conclusions

In the current study, the concentration levels of naturally occurring radioactive materials (NORMs) of ^{40}K, ^{226}Ra (^{238}U), ^{232}Th, and ^{235}U in the surface samples of selected areas in Al-Lajjun in south Jordan were investigated. The collected samples were analyzed employing gamma-ray spectrometry. The radioactivity concentration of the naturally occurring radionuclides ^{40}K, ^{226}Ra (^{238}U), ^{232}Th, and ^{235}U in the samples varied from 12-560, 234-6037, 4.3-28.4, 43-3656 Bq/Kg, respectively.

Based on the sample analysis results showing high radioactivity, we recommend analyzing the water stored in the dam at different depths. There is no harm in studying the plants grown in the area and irrigated by dam water. Also, more soil samples from different locations at different depths must be collected and investigated from the same region.

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Conflict of interest

We have no conflicts of interest to disclose. All authors declare that they have no conflicts of interest.

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