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Determination and Comparative Analysis of Natural Radioactivity Levels Using Gamma Spectrometry in Shore Sediment Samples from the East Coast, Libya

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ABSTRACT

The occurrence of natural radioactivity in the environment is attributable to the primordial and cosmogenic nuclides in the earth's crust where the exposure associated with this natural radioactivity relies principally on the geological and geographical environments. In this paper, Natural-radionuclide levels ^{238}U , ^{226}Ra , ^{232}Th and ^{40}K were determined and investigated in seashore sediment samples collected from an area on the Eastern Coast of Libya (Al Jabal Al Akhdar region). The goal of this research is to evaluate radionuclide's concentrations and their distribution in this region. The measurement of natural radioactivity concentrations in the sediment samples was performed employing gamma-ray spectrometry techniques using a pure germanium detector (HPGe). The extracted values of the activities for ^{238}U , ^{226}Ra , ^{232}Th , and ^{40}K were in the range of values obtained in similar studies in other countries and are within the average worldwide values. The calculated average values of radioactivity concentrations for ^{238}U , ^{226}Ra , ^{232}Th and ^{40}K were 7.75, 8.53, 3.16 and 13.26 Bq/kg, respectively. The descriptive statistical features of radioactive levels in sediment samples are also presented. Due to the limited similar studies in the study area, the obtained results could assist in setting up a reference line of radiometric records for radioprotection purposes and environmental monitoring schemes in the study area and in Libya in general.

تقدير وتحليل مقارن لمستويات النشاط الإشعاعي باستخدام مطيافية جاما في عينات رسوبيات الشاطئ من الساحل الشرقي، ليبيا

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الكلمات المفتاحية:

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الملخص

حدوث النشاط الإشعاعي في البيئة يعزى للنويدات البدائية والكونية في قشرة الأرض حيث التعرض للإشعاع المرتبط بهذا النشاط الإشعاعي يعتمد أساساً على البيئات الجيولوجية والجغرافية. في هذه الورقة، مستويات النشاط للنويات الطبيعية ^{40}K , ^{232}Th , ^{226}Ra , ^{238}U تم قياسها وفحصها في عينات من رسوبيات الشاطئ جمعت من الساحل الشرقي في ليبيا (منطقة الجبل الأخضر). الهدف من الدراسة كان فحص تركيزات النويات وتوزيعها في منطقة الدراسة. تركيزات هذه النويات تم باستخدام مطياف أشعة جاما بالاعتماد على مكشاف الجرمانيوم ثنائي المحور عالي النقاوة. القيم المتحصل عليها للنويات ^{40}K , ^{232}Th , ^{226}Ra , ^{238}U كانت في مدى القيم المتحصل عليها في دراسات مماثلة في دول أخرى وتقع ضمن المدى المتوسط العالمي. متوسط القيم المحسوبة لتركيزات

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النشاط الإشعاعي لأجل ^{40}K , ^{232}Th , ^{226}Ra , ^{238}U كانت على التوالي 13.26, 7.75, 3.16, 8.53 بوحدة Bq/kg الخصائص الوصفية الإحصائية للمستويات الإشعاعية لعينات الرسوبيات تم أيضا عرضها. ونظرا لمحدودية دراسات مشابهة في منطقة الدراسة فإن النتائج المتحصل عليها يمكن أن تساعد لتكون مرجع للقياسات الإشعاعية لأغراض الحماية الإشعاعية ومبادرات المراقبة البيئية في منطقة الدراسة وليبيا عموما.

1. Introduction

One of the main sources of radioactivity for human beings is continuous exposure to natural radionuclides. This natural radioactivity exists in the environment as a result of the primordial and cosmogonist nuclides in the Earth’s crust. The exposure associated with natural radioactivity, which is mainly due to gamma rays, relies principally on the geological and geographical environments [1]. As a result, radionuclides are emitted into the surrounding ecosystem through various sources and activities [2]. Hence, naturally occurring radionuclides find their way to marine environments and, in particular, to sea sediments, the subject of the present study [3]. Assessment of radionuclides in the marine environment, either water, biota, sand, or sediments is the goal of many studies worldwide [4-8]. These studies are important to evaluate the hazards that might be linked to the absorption of radionuclides into the food chain by humans [9]. As for sediments, which are considered a host for this environmental radioactivity, and their role in accumulating and transporting various pollutants in sea biota, it is important to investigate their contents as far as radiological activities are contemplated [10-11]. Keeping in mind that 80% of the radiation exposure to humans is from natural radionuclides, there are sizable portions of radiation from manmade radionuclides which may end up in the human food chain. The coastal states, like Libya, are required to protect the wellbeing of their coastal sites and beaches. Libya has about 2200 km of coastline along the Mediterranean Shore. This coastal line is of significant economic, ecological, and leisure significance. Therefore, collecting a set of data for the radionuclides’ distribution of in sediments for the Libyan Coast is very important. Only few data are available for the study area. Sediments act as the eventual sink for the radionuclides. This study has been undertaken on parts of the Eastern Libyan Coast due to their proximity to coastal farming lands. This anticipated accumulation of these radionuclides may be due to fallout leading to sediments in waters having a higher biological productivity to be a principal sink for these elements. The present study is initiated to assess the levels of some natural radionuclides in sediment samples along the Al Jabal Al Akhdar region Coastline as well as to initiate a reference point for the radioactivity levels in the study area [12].

2. Study Area

The investigated area extended along the Coast of Al Jabal Al Akhdar region (about 144 km along), from 32°53'38" N to 20°55'54" E in the west (Toukra region) to 32°89'58" N to 21°91'02" E in the east (Sousa region). Fig. 1 and Table 1 show the eight sites where the sediment samples were collected in the Eastern Coast of Libya. To be noted that there are various drainage routes flowing towards the Mediterranean Sea from Al Jabal Al Akhdar which may be another source of coastal sediments.



Fig. 1: Sites of collected samples along the northern east coast of Al Jabal Al Akhdar region.

Table 1: Latitude and longitude lines of the sites of the collected samples.

s. no	Location	Latitude	Longitude
1	Tawkraa	32°53'38"N	20°55'54"E
2	Potraba	32°65'07"N	20°82'30"E
3	Telmitha	32°71'48"N	20°94'78"E
4	Bataa	32°76'94"N	21°17'14"E
5	Gargarima	32°78'63"N	20°40'34"E
6	AL-Haniya	32°83'49"N	21°50'60"E
7	AL-Hmama	32°91'76"N	21°62'23"E
8	Sosaa	32°89'58"N	21°91'02"E

3. Materials and Methods

The sediment samples were gathered employing a special grabber and at depths of ranging from 10 to 20 cm. The samples were investigated for their radiological natural activity and were then compared with results available in the earlier studies[13]. The distance between these sites was an average of about 20 km. The wet weight of each sample was about 1 kg. To get rid of the moisture content and to obtain a constant weight for each sample. An oven was used to dry the samples at 110°C without causing any heat damage to the samples. The samples were from the solid type and not carbonate or clay ones that might dissolve due to heating or undergo compositional and/or structural changes. The samples were grounded to achieve a powder form using mortar and pestle. The obtained powder was sieved with a 2 mm mesh to attain homogenized samples. Each sample was set to have a net weight of 350 g and packed in containers with a particular geometry analogous to that of the calibration source used in the radioactivity analysis. To preclude any loss of radium-isotope near the container walls and to avert any micro-organisms growth, the samples were placed in a concentrated HNO₃ acid. A gamma-ray spectrometry using a pure germanium detector (HPGe) was utilised to measure the activity levels of ^{238}U , ^{226}Ra , ^{232}Th and ^{40}K . The relative efficiency of the detector was about 30% with a resolution energy of 1.9 keV FWHM for a gamma transition of ^{60}Co with 1332 keV. The background activity was subtracted from the measured values using a spectrum analysis program. An empty Marinelli beaker, under the same measurement circumstances was used to estimate the background radiation. The activity levels were estimated from the gamma lines radiated from the decay products of the radio nuclides. The weighted mean of the photo peak activities of the daughter product ^{234}Th (63.3 keV) of ^{238}U was used to measure the activity concentration of ^{238}U . To measure the activity of ^{226}Ra , the gamma lines 295.2 and 351.9 keV for ^{214}Pb and 609.3, 1120.3 and 1760.5 keV for ^{214}Bi were employed. The activity concentration of ^{232}Th was determined using the gamma lines 338.3 and 911.6 keV for ^{228}Ac , and 583.0 keV for ^{208}Tl . While the activity of ^{40}K was determined by its gamma-line 1460.8 keV. To lessen the statistical counting error, a counting rate of at least 36 000 s was employed[14].

4. Results and Discussion

The activity levels of ^{238}U in the samples ranged from 6.3 to 9.5Bq/kg, with an average of 7.75Bq/kg; ranged from 6.7 to 10.6 Bq/kg for ^{226}Ra , with an average of 8.53Bq/kg; ranged from 2.5 to 3.7 Bq/kg for ^{232}Th , with an average 3.16 Bq/kg, where as the levels of ^{40}K ranged from 22.0 to 29.2 Bq/kg, with an average of 13.26 Bq/kg. These values are for dry weight samples and assuming secular equilibrium in each of the ^{238}U and ^{232}Th series. These measured values indicate that the radioactivity levels in the samples have low risk values[15-16]. The samples radioactivity activity levels (in Bq/kg) of ^{238}U , ^{226}Ra , ^{232}Th , and ^{40}K are presented in Fig 2. and Table 2. The recorded average activities of ^{238}U , ^{226}Ra and ^{232}Th are lower than the world average limits[1]. Along with the error treatment calculation (shown in Table 2), descriptive statistical calculations, including ranges skewness, kurtosis, standard deviations and means, of the activity concentrations for sediments samples are shown in Table 3. In particular, skewness

and kurtosis are used to estimate if the normality assumption is satisfactory or not. Kurtosis and skewness coefficients are expected to be low, which is the case here, to have a normal distribution (see Table 3).

Table 2: Natural activity concentrations of ²³⁸U, ²³²Th, ²²⁶Ra and ⁴⁰K in sediment samples (in Bq/kg).

S. No	²³⁸ U	²²⁶ Ra	²³² Th	⁴⁰ K
1	8.1±0.48	8.6±0.7	2.5±1.0	28.1±3.8
2	9.5±1.1	10.6±1.3	3.6±0.2	22.0±4.0
3	6.3±0.72	7.0±0.6	2.6±0.64	< D.L.
4	8.4±0.55	9.1±0.8	3.0±0.86	< D.L.
5	7.8±0.76	8.6±0.9	3.6±0.81	< D.L.
6	7.6±1.1	8.7±1.2	3.7±1.2	29.2±3.5
7	6.3±0.7	6.7±0.5	2.9±0.7	< D.L.
8	8.0±0.86	8.9±0.7	3.4±0.7	26.8±3.0
Ave.	7.75	8.53	3.16	13.26

Table 4 presents a comparison of radioactivity levels of the current study with some regions of the world. The ²³⁸U and ²³²Th activities are in fair agreement with the values reported in some of the studies. On the other hand, ⁴⁰K activity measured value in the present study is lower than values obtained by other studies in different locations of Egypt (an eastern neighbour to Libya) and the other parts of world (see Table 4). In particular, noting that Elmzainy et al.(2022), who performed a study in the same study area (even though the sites may differ), found in their study that the average values for radionuclides ²²⁶Ra, ²³²Th and ⁴⁰K (in Bq/kg) as 8.26, 5.95 and 66.1, respectively [13]. These values are in fair agreement with the ones obtained in the present study except for ⁴⁰K. Elmzainy et al. (2022) used a NaI detector which is less efficient than the HGe detector [13]. Even though, NaI detectors are the very common scintillation detectors using NaI crystals doped with thallium (NaI(Tl)), they have poor energy resolution. On the other hand, HPGe detectors have high resolution

Table 4: Comparison of radioactivity levels (in Bq/kg) of the current study with some regions of the world.

Location	²³⁸ U	²²⁶ Ra	²³² Th	⁴⁰ K	Place	Ref.
Libya	7.75(6.3-9.5)	8.53(6.7-10.6)	3.16(2.5-3.7)	13.26(22.0-29.2)	North East of Libya	Present Work
Libya	-	8.26	5.95	66.1	North East of Libya	[13]
Egypt	17.3(12.6-19.9)		10.03(8.5-10.6)	299.7(258.8-316.8)	Burullus lake, Egypt	[17]
Egypt	24.6(5.2-105.6)		31.4(2.3-222)	428(98-1011)	Red Sea Coast	[18]
Egypt	20.99(7.06-30.15)		14.8(5.7-20.35)	244.7(68-352)	Qarun sediment, Egypt	[5]
USA	37.8(11.1-74.2)	21.4(11.4-41.2)	45.3(13-185.8)	609.3(385.9-1046.9)	Reedy River, USA	[19]
Jordan	11.2-677	5-31	3.6-32.8	71.5-901.1	Gulf of Aqaba	[20]
Greece	28(9-43)	27(10-37)	30(12-46)	483(218-686)	Patras-Rion sub-basins, Greece	[21]
India	3.67(2.2-20.9)		37.23(2.1-233.9)	387.2(313.3-482.5)	Coastal sediments, India	[22]
Worldwide	33	32	45	420		[1]

Each series of ²³⁸U, ²²⁶Ra and ²³²Th shows a significant positive correlation with each other. This indicates that there is a high similarity in the local circumstances and accumulation sources. For example, the average ²²⁶Ra/²³⁸U activity ratio is 1.09 which is considered to be low. The adsorption or de-adsorption of radium from the sediments depends on chemical and physical conditions. Generally, sediments at the drain sites have radium adsorbed to the particulate matter at the surface. After that, radium is desorbed whenever saline surroundings is available [18].

5. Conclusion

The radioactivity activity levels of natural radionuclides ²³⁸U, ²²⁶Ra, ²³²Th, and ⁴⁰K in sediment samples were measured using gamma-ray spectrometry in the Eastern Coast of Al Jabal Al Akhdar region (Libya). The activity concentrations of these radionuclides are estimated to be in normal range and, in general, are in fair agreement with corresponding ones in some regions of the world. The obtained values are less than the average worldwide values. Consequently, the study area presents no damaging radiation effects to the public going to the seashores for leisure or to the fishermen fishing in this area. The results in this study may be as a reference for future monitoring of radioactivity in the region and as a baseline for future surveys.

References

abilities. While, NaI(Tl) detectors are cheaper and simpler, their ability to resolve gamma peaks and lines are limited.

Table 3: Statistical variations of radioactivity values of the sediment samples.

Element	²³⁸ U	²²⁶ Ra	²³² Th	⁴⁰ K
Mean	7.75	8.53	3.16	13.26
St. deviation	1.06	1.21	0.473	9.134
Skewness	-0.04	0.021	-0.174	1.717
Kurtosis	1.79	2.027	1.034	2.738
Minimum	6.3	6.7	2.5	22.0
Maximum	9.5	10.6	3.7	29.2
Variance	1.12	1.465	0.224	83.432
Frequency	8	8	8	8

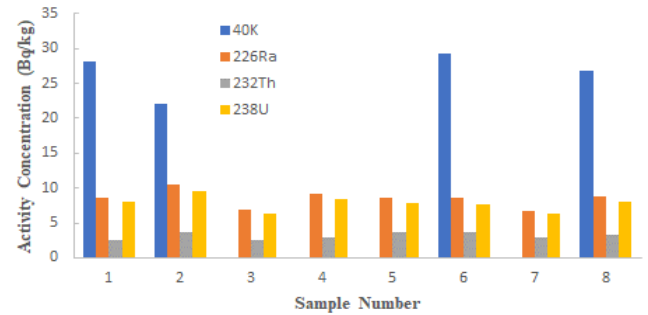


Fig. 2: Activity-level concentrations of ²³⁸U, ²²⁶Ra, ²³²Th and ⁴⁰K in the sediment samples collected along the northern east coast of Al Jabal Al Akhdar region.

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