NATURAL RADIOACTIVITY LEVELS ALONG THE MEDITERRANEAN SAND BEACH BETWEEN TAJOURA AND MISRATA, LIBYA

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المخلص

تحظى قياسات مستويات النشاط الإشعاعي في الشواطئ باهتمام كبير خلال العقود الماضية لعديد الدراسات في جميع أنحاء العالم. وتوفر هذه الأبحاث امكانية رسم خرائط لمستويات الإشعاع توثيق البيانات المرجعية للتأكد من التغيرات المحتملة في النشاط الإشعاعي البيئي بسبب الأنشطة النووية والصناعية وغير ها من الأنشطة البشرية. والجدير بالذكر أن ليبيا لديها إمكانات كبيرة للسياحة مع توفر 1700 كم من السواحل.

أجريت هذه الدراسة لتحديد مستويات تركيز النشاط الإشعاعي الطبيعي في سلاسل اليورانيوم-238 والثوريوم-232 والبوتاسيوم-40 في ثلاثين عينة من رمال الشاطئ التي تم جمعها من مواقع مختلفة من الساحل بين تاجوراء ومدينة مصراته. تم تحليل عينات الرمال بواسطة نظام التحليل الطيفي لأشعة جاما باستخدام كاشف الجرمانيوم عالي النقاء (HPGe) الموجود في مختبر الإشعاع النووي في قسم الهندسة النووية بجامعة طراباس ليبيا.

كانت تركيزات النشاط الإشعاعي لنويدات U 238 ، Th (238 و 40 K بناءً على القيم المتوسطة الموزونة عبر العينات ($0.38\pm 6.728\pm 0.183\pm 29.673$) و (0.576 ± 3.010) و ($0.38\pm 29.673\pm 0.183\pm 29.673$) و التوالي.

تراوح تركيز النشاط U^{238} و Th في عينات رمال الشاطئ من 3.668 ± 0.381 إلى تراوح تركيز النشاط U^{238} و H^{0} في عينات رمال الشاطئ من 3.668 ± 0.381 إلى 2.806 ± 0.365 يكريل/كجم و 25.656 ± 2.405 يكريل/كجم و 2.405 ± 1.201 بيكريل/كجم و 1.460 ± 1.201 إلى 88.881 ± 0.755 بيكريل/كجم لكل من U^{238} من U^{238} و H^{0} على التوالي.

ABSTRACT

Measurements of the levels of radioactivity in beaches are of great interest during the last decades for many studies worldwide. Such investigations can be useful for mapping of radiation levels and recording reference data to ascertain potential environmental radioactivity changes due to nuclear, industrial, and other human activities.

This study was conducted to determine the natural radioactivity concentration levels of ²³⁸U, and ²³²Th natural decay chains and ⁴⁰K in 30 beach sand samples collected from different sites of the coast between Tajoura and Misrata, Libya.

Representative sand samples have been analysed based on the gamma spectroscopy system using High Purity Germanium (HPGe) detector housed in the Nuclear Radiation Laboratory at the Department of Nuclear Engineering, University of Tripoli, Libya. The activity concentrations of 238 U, 232 Th and 40 K based on the weighted mean values across samples were (6.728±0.385), (3.010±0.576) and (29.673±0.183) Bq/kg, respectively.

The activity concentration of ²³⁸U, ²³²Th and ⁴⁰K in the beach sand samples ranged from 3.668 \pm 0.381 to 25.656 \pm 2.405 Bq/kg, 0.755 \pm 0.271 to 13.081 \pm 1.231 Bq/kg and 15.935 \pm 1.460 to 88.881 \pm 6.20 Bq/kg for ²³⁸U, ²³²Th and ⁴⁰K respectively. The annual effective dose equivalent in µSv/y ranged between 4.092 \pm 0.494 and 21.694 \pm 0.214 µSv/y while the world average annual effective dose equivalent (AEDE) from outdoor or indoor terrestrial gamma radiation is 290 µSv/y. The activity concentrations of ²²⁶Ra, ²³²Th, and ⁴⁰k were below the global limit. Thus, there are no harmful radioactive effects resulting from sea sand.

The achieved results will be used as reference information to track any change in the radioactivity background levels in this area and may provide good achievement for mapping the radioactivity background.

KEYWORDS: Activity Concentration; Gamma Spectroscopy; Dose, HPGe; Activity in Beach Sand.

INTRODUCTION

The coastline of Libya extends over 1970 km and is characterized by its relative homogeneity since there is no marked differentiation of its topography and geomorphology. Marine pollution generated by oil activities and sewage is affecting the marine environment in Libya in some areas [1]. Sand beaches produce from the rocks on land, which over time, rain, ice, wind, heat, cold, and even plants and animals break rock into smaller particles. These soil particles are transported by the flowing river to the low lying areas and deposited there as sand. The environment quality and beach areas and surroundings attract the tourists and other holidaymakers.

Environmentalists have started to put more attention to continuous monitoring of the radioactive concentration levels in the environment. Due to the serious damage that may occur to the aquatic ecosystem. There are two known main causes of the pollution; natural and anthropogenic [2, 3].

The surface contamination of water systems can be owed to increase in industrial activities and rapid urbanisation. The main source of radiation exposure for humans is the environmental natural radioactivity associated with the terrestrial and cosmic radiation. The natural radiation contributes with about 80% of a year's effective exposure for humans [4].

Individuals are exposing to radiation in two pathways; the external exposure to gamma radiation emitted from the terrestrial radionuclides; ²³⁸U ($T_{1/2} = [4.468 \pm 0.003] \times 10^9$ years), ²³²Th ($T_{1/2} = [1.405 \pm 0.006] \times 10^{10}$ years) and ⁴⁰K ($T_{1/2} = [1.277 \pm 0.008] \times 10^9$ years) and internal exposure from the inhalation of radioactive radon and its daughters [3-7].

The terrestrial γ -ray radiation with about 20% of the total annual dose (2.4 mSv) or 0.48 mSv due to natural sources and inhalation of radon as internal exposure accounting for 48% (1.15 mSv) of this dose [8-10].

Individuals long term exposure to radiation causes significant health effects. Many diseases such as lung cancers, the pancreas hepatic, due to the radon gas inhalation and ingestion of radioactive food [4,11]. Disasters, such as the Chernobyl and Fukushima, are other sources of radionuclides in the environment. Consequently, periodic radiological surveys, monitoring and screening the environment, are all necessary.

The external exposure contributes to the environmental hazardous from the natural radionuclides present in beach sands [2,5,6,12]. Measurements of radioactivity levels in beaches are of a great interest during the last decades for many studies worldwide. Such investigations can be useful for mapping of radiation levels and recording reference data to ascertain potential environmental radioactivity changes due to nuclear, industrial and other human activities [5,13,14].

Measurements of natural radioactive nuclides in beach sands have been reported in many studies worldwide. Various techniques have been used to assess natural radionuclides in different environmental media. The most common used methods include direct gamma spectroscopy, alpha spectrometry, and liquid scintillation counting [1,2,5,7,12,15-17].

The primary aim of this study is to determine the activity concentration of the natural radionuclides ²³⁸U, ²³²Th, and ⁴⁰K using gamma spectrometry based on a high purity germanium (HPGe) detector. Beach sand samples collected from the coast of Libya, the region from Tajoura to the City of Mistrata, were analysed to measure and quantify natural gamma-emitting radionuclides.

MATERIALS AND METHODS Sample collection and preparation

In total, 30 beach sand samples were collected from different locations on the west of Libya along the Mediterranean coast from Tajoura to the City of Misrata. Figure (1) shows these locations on the map, the distance between these sites was 4-7km. Samples of 1.5 to 2kg were collected from the closest distance from the seashore with a mold of area $20x20 \text{ cm}^2$ and a depth between 15-20 cm from the surface. Three samples were collected from each location, where the distance between them was 10 m. Samples then well-mixed and one sample was kept for each location.



Figure 1: The regions of collected sample

Samples were weighed, dried in oven at 100°C and then sieved through a 0.1-mm mesh sieve for a uniform particle size. The homogenized sample was contained in cylindrical plastic container (71 mm in diameter with 47 mm height and thickness of 1.1 mm), well-sealed with adhesive tape, weighted and labelled (according to its name, weight, and storage date). This container was chosen to match the standard source geometry.

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All samples then stored for one month at the Department of Nuclear Engineering, the University of Tripoli to reach the state of the equilibrium [18,19]. Figure (2) shows the samples preparation tools and method. Table (1) lists the collected samples.



Figure 2: Samples Collection protocol

Sample Label	Sample Location	Sample Label	Sample Location	Sample Label	Sample Location
Sa-1	Quara Polly	Sa-11	Ahmed Palace Resort	Sa-21	The Palm Resort (Al Khums)
Sa-2	Steam Station	Sa-12	Al Khums Port	Sa-22	Ganaima
Sa-3	Mirbat Beach	Sa-13	Zliten Port	Sa-23	Sur Sauod Beach
Sa-4	Bsees Beach	Sa-14	Zliten Dump	Sa-24	Al-Guaia HT
Sa-5	Tajoura Energy	Sa-15	Al Aman Resort	Sa-25	Toba Beach
Sa-6	Um ElZain Beach	Sa-16	Ain Kaam	Sa-26	Leptis Magna
Sa-7	Falfool Beach	Sa-17	Daphnia	Sa-27	Celine Beach
Sa-8	Trailer Factory Beach	Sa-18	Popular Market	Sa-28	Ispan
Sa-9	Iron and Steel Resort	Sa-19	Marsa Al-Quara Polly	Sa-29	Mara Tajoura
Sa-10	The Industries Resort	Sa-20	Zliten Beach	Sa-30	Zureik Beach

Table 1: Collected samples and their locations

Gamma-Ray Spectrometry

A high-resolution hyper-pure germanium detector-based, gamma-ray spectroscopy, low-background counting system by ORTEC was used in this study. The detector is housed in one of the nuclear radiation laboratories of the University of Tripoli at the Nuclear Engineering Department. A lead castle of approximately 10 cm thick with a copper lining on the inside to absorb any lead K X-rays, shields the detector. The relative efficiency of the detector used in these experiments is examined and compared with the manufacture nominal relative efficiency to the scintillation counter (86.4%) and found to be 76.2%. The analysis was performed using the software Ortec <u>GammaVision[®]</u> (Ortec GammaVision-32 v. 6 2010).

Energy and Efficiency Calibration

HPGe detector energy calibration was measured over a wide range of energies. The calibration was conducted by using full-energy peaks from a standard source (SYRNORM 2005) in the shape of a cylindrical plastic container with Ra-226 activity of 9775 Bq/kg.

Minimum Detectable Activity (MDA)

The minimum detectable activity (MDA) for each radionuclide was obtained from the background radiation spectrum for the same counting time (24hr) as for the soil samples. The values were calculated using equation (1) [20].

$$MDA = \frac{N_d}{\epsilon P_{\gamma} T m}$$
(1)

Where; $N_d = L_c + 2.706$, $L_c = 4.653 \sigma_{NB}$

Where, N_d , σ_{N_B} , ϵ , P_{γ} , L_c are the counts, the standard deviation of the background, detector efficiency, the emission probability of the gamma line corresponding to the peak energy, critical level, respectively, and T is the sample counting time. *m* is sample mass.

RESULTS AND DISCUSSION

Detection Limit and Minimum Detectable Activity (MDA)

The minimum detectable activity obtained was in the range of 0.110 ± 0.001 to 1.01 ± 0.002 Bq/kg as shown in Figure (3). This range of values was used to check if the activity concentrations found within the samples are of measurable values throughout this study.



Figure 3: Calculated MDA for listed radionuclides

Activity Concentrations

The activity concentrations of each identified radionuclide from ²³⁸U, ²³²Th series and ⁴⁰K of seashore sand samples were calculated using the following equation:-

$$A(Bq/kg) = \frac{N-B}{\varepsilon \times t \times I_{\gamma} \times m}$$
(2)

Where ε is the absolute efficiency at a given photo peak energy, I_{γ} is the emission probability of the gamma line corresponding to the peak energy, *m* is the mass of the measured sample (kg), *N* and *B* are the areas under the photopeaks of the sample and the background, respectively [5,12].

The gamma-energy lines of 609.32 (45.4%) keV, 1120.29 (14.8%) keV, emitted by 214 Bi were used to represent the 238 U (226 Ra) series, while 911.20 (25.8%) keV and 968.97 (15.8%) keV from 228 Ac were used to represent the 232 Th series and 1460.82 (10.66%) keV for 40 K gamma-line.

Then obtained values were compared with the reported natural radionuclides mean activities in Libyan soil by UNSCEAR (2008) global survey on exposures to natural radiation sources of ²³⁸U, ²³²Th series and ⁴⁰K, which are 10.5, 8.8, 9.5 and 270 Bq/kg, respectively [20].

In addition, these activities were compared with world limit (33 Bq/kg for ²³⁸U, 45 Bq/kg for ²³²Th, and 420 Bq/kg for ⁴⁰K) reported by UNSCEAR 2008. The activity concentration of ²³⁸U, ²³²Th and ⁴⁰K based on the weighted mean values across samples as shown in Figures (4-6) were (6.728 ± 0.385),(3.010 ± 0.576) and (29.673 ± 0.183) Bq/kg, respectively. The lowest radioactivity of ²³⁸U was about (3.668 ± 0.381) Bq/kg in sand sample Sa-01 collected from Quara Polly. Sample Sa-9 from Iron and Steel Resort contained the lowest activity of ²³²Th, (0.755 ± 0.271) Bq/kg.

For ⁴⁰K the lowest activity concentration obtained in sample Sa-22 collected from Ganaima (15.935 \pm 1.460) Bq/kg, while the largest activity recorded in sample S-14 from Zliten Dump with (88.881 \pm 6.201Bq/kg). Sample S-20 collected from Zliten Beach (32°30'21.1"N 14°30'01.6"E) revealed the highest values of ²³⁸U and ²³²Th series (25.656 \pm 2.405) and (13.081 \pm 1.231) Bq/kg respectively. These values were higher than limits of activity of natural radionuclides activity in Libyan soil reported in UNSCEAR (2008).



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Figure 5: Th-232 radioactivity concentrations in Bq/kg



Sample No. Figure 6: K-40 radioactivity concentrations in Bg/kg

Radiation Hazard Assessment

Gamma Absorbed Dose Rate Assessment

The Radium equivalent activity in each sample was calculated by:

$$Ra_{eq} = A_{Ra} + 1.43A_{Th} + 0.077A_{K}$$

(3)

Where A_{Ra} , A_K and A_{Th} are the specific activities of ²³⁸U, ⁴⁰K and ²³²Th in (Bg/kg). The values of Ra_{eq} found to be in the range of 7.743±0.359 to 44.681±0.149 and Bq/kg, which were below the world's mean values 370 Bq/kg [14,16,21].

The gamma dose rate (D) in nGy/hr caused by naturally occurring radioactive materials in air at 1 m above the ground surface can be estimated using the following formula [5]:-

$$D = 0.461A_{Ra} + 0.623A_{Th} + 0.0414A_{K}$$

(4)

Where A_{Ra} , A_{Th} and A_K are the specific activities of ²²⁶Ra, ²³²Th and ⁴⁰K in (Bq/Kg).

The range of calculated gamma dose rate in this study was found to be between 4.092 ± 0.494 and 21.69 ± 0.214 nGy/h. Figure (7) illustrates the Dose rate, Ra_{eq} distribution for all samples.

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Figure 7: Dose rate, Ra_{eq} distribution for all samples.

External Hazard and Annual effective dose equivalent (AEDE)

External hazard index for each sample was found to be less than unity, and Table (2) lists these results, which calculated by:-

$$H_{ex} = \frac{C_K}{4810} + \frac{C_{Th}}{259} + \frac{C_{Ra}}{370}$$
(5)

Where C_K , C_{Th} and C_{Ra} are the activities concentrations of 40 K, 232 Th and 226 Ra (in Bq/kg) respectively. This expression indicates that the value of this index must be less than unity for the radiation hazard to be insignificant. Thus, the maximum values of H_{ex} equal to unity correspond to the upper limit of Ra_{eq} being 370 Bq/kg.

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Sample #	Hex	Sample #	H _{ex}	Sample #	Hex	
Sa-1	0.032	Sa-11	0.028	Sa-21	0.029	
Sa-2	0.032	Sa-12	0.032	Sa-22	0.023	
Sa-3	0.035	Sa-13	0.030	Sa-23	0.030	
Sa-4	0.048	Sa-14	0.041	Sa-24	0.034	
Sa-5	0.035	Sa-15	0.037	Sa-25	0.030	
Sa-6	0.028	Sa-16	0.028	Sa-26	0.031	
Sa-7	0.028	Sa-17	0.036	Sa-27	0.027	
Sa-8	0.031	Sa-18	0.046	Sa-28	0.038	
Sa-9	0.036	Sa-19	0.039	Sa-29	0.051	
Sa-10	0.030	Sa-20	0.128	Sa-30	0.034	

Table 2: External Hazard Index from 30 Samples.

The Annual Effective Dose Rate

In order to estimate the annual effective dose rate in air, the conversion coefficient from absorbed dose in air to effective dose received by an adult must be considered. This value is published in UNSCEAR 2000 and UNSCEAR 1993, to be 0.7 SvGy⁻¹ for environmental exposure to gamma rays of moderate energy. The outdoor occupancy factor is about 0.2 .The annual effective dose equivalent is given by the following equation [14].

AEDE (
$$\mu$$
Sv/y) = D(nGy/h) ×8760(h/y) ×0.2×0.7(Sv/Gy) ×10⁻³ (6)

The annual effective dose equivalent in μ Sv/y shown in Figure (8) ranged between 5.019 and 26.606 μ Sv/y, which was within the allowed limits that equal (1 mSv/y) for all samples and all regions that were selected in the present study.



Figure 8: annual effective dose equivalent for each sample

CONCLUSION

Gamma spectrometer of HPGe was used to measure the activity concentration of natural terrestrial radionuclides, ²³⁸U, ²³²Th, and ⁴⁰K in beach sands collected from different sites of the coast between Tajoura and Misrata, Libya. Reported values are comparable to the average worldwide range.

The achieved results were compared with previously published study, which was conducted to measure radioactivity concentrations of beach sand samples from different locations along the coast of Tripoli, northwest Libya, using high resolution γ -ray spectroscopy. Their findings showed that the radioactivity concentrations for ²²⁶Ra, ²³²Th, ⁴⁰K in samples taken at depth 5-10 cm have an average 7.5 ± 2.5 Bq/kg, 4.5 ±1.3 Bq/kg, 28.5 ± 6.7 Bq/kg respectively. This study showed that the activity of ²³⁸U, ²³²Th and ⁴⁰K based on the weighted mean values across samples were (6.728±0.385), (3.010±0.576) and (29.673±0.183) Bq/kg, respectively, which is consistent with the previously published study [21].

The range of the absorbed dose rate obtained for the soil samples was between 2.7n Gy/h and 6.1 nGy/h, while the mean value of the absorbed dose rates in air was 4.4 ± 1.3 nGy/h. In this work the absorbed dose rate values were higher and found to be between 4.092 ± 0.494 and 21.69 ± 0.214 nG/h. [21].

All the results obtained in this study showed that the beaches in the region of study are safe in terms of radiological hazard, with no estimated values exceeding the permissible levels. Obtained results will be employed as reference information to track any change in the radioactivity background levels within the area covered in this study and may provide good achievement for mapping the radioactivity background.

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