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Natural Radioactivity of Beach Sand Samples in the Tripoli Region, Northwest Libya

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Abstract

A comprehensive study was conducted to determine the radioactivity concentrations of beach sand samples from different sites along the coast of Tripoli, northwest Libya, using high resolution γ-ray spectroscopy. Collection of samples was carried out during low tide, where it was possible to collect sediments from the wet region that was covered by sea water during high tide. From the measured γ-ray spectra, elemental concentrations were determined for ^{226}Ra, ^{232}Th, ^{40}K, and ^{210}Pb at depths 5-10 and 50-70 cm from the surface. The activity concentration ratios of ^{210}Pb/^{226}Ra for some samples were calculated to show the disequilibrium between the ^{210}Pb and its parent nuclei ^{226}Ra in the coastal marine sediments. The mean value of the absorbed dose rates in air obtained in the studied area was 4.4 $\pm$ 1.3 nGy h^{-1}, which is lower than the world average (55 nGy h^{-1}).

Key words: Natural radioactivity, Radionuclide, Beach sand, Gamma ray spectroscopy, Libya.

Introduction

Radionuclides have been an essential constituent of the Earth since its creation. The Earth contains many radioactive elements; the origin for part of them goes back to the formation of our world, while the others are continuously produced through nuclear reactions in the universe.

Measurements of natural radioactivity in soil have been performed in many parts of the world, mostly for assessment of the dose and risk resulting from them(Beck, 1972a, 1972b; Sheppard and Evenden, 1988; Nellis, 1990; Rudge et al., 1993; Clouvas et al., 2001; Copplestone et al., 2001; Dowdall et al., 2004; El-Bahi et al., 2005). It was observed that most of natural radioactive elements present in soil are primordial radionuclides from the uranium series, thorium series, and ^{40}K. The minimum radioactivity was found in sand soil by Lauria and Godoy (2002) and Saleh (2002). Several radionuclides in the radioactive decay chains starting from ^{238}U and ^{232}Th are highly radiotoxic, in particular ^{226}Ra and ^{228}Ra. The radioactive gas (radon) that arises from the disintegration of ^{238}U and ^{232}Th in the Earth's crust is considered the main source of exposure to ionizing radiation for humans, representing 40% of the annual accumulated dose (UNSCEAR, 1993). Measurements of ^{210}Pb in air and in surface soils will afford quantitative information about the flux of radon gas (^{222}Rn) and its daughters in the atmosphere (Harley et al., 2000; Winkler and Rosne, 2000; Mèliéres et al., 2003; Paatero et al., 2003; Gaffney et al., 2004; Ioannidou et al., 2005). It can help in uranium exploration and monitoring

transfers of radionuclides of uranium series in soils and aquatic systems. In the context of luminescence dating, the ^{210}Pb/^{226}Ra activity ratio can give the proportion of ^{222}Rn that can escape from given sediment, such data being important in the calculation of annual radiation dose rate. Furthermore, ^{210}Pb and its grand-daughter radionuclide ^{210}Po are included in the group of most highly toxic radioisotopes and provide the major internal natural radiation dose to humans. It is approximately 18% of the average dose to the population from internal irradiation due to ingested radionuclides. For some members of the public the dose due to ingestion of ^{210}Pb and ^{210}Po may be far higher due to high intakes of specific foodstuffs such as shellfish (Clayton and Bradley, 1995; Pilleyre et al., 2006). The high quantities of the radioactive nuclides require the use of appropriate methods of elimination to minimize their health hazards(Sorg, 1991; Herranz, 1999).

There is no previous work to give a complete database about the distribution of radioactive nuclides and their concentrations in the coastal region of Tripoli, northwest Libya. Therefore, the present study aimed to measure the concentrations of natural and artificial radionuclides in the surface soil samples from the coastal part along the Tripoli region to assess the level of background radiation arising from them. In addition, these data will be useful for subsequent evaluations of possible future environmental contamination due to non-nuclear industries or any future activities.

Experimental Procedure

Soil samples were collected along the shore of the Mediterranean Sea a few meters (10-15 m) from the water. Forty soil samples were collected randomly from different locations alongside the coast of Tripoli, northwest Libya, during low tide over a distance 3 km for each location, where 4 samples were collected using a template 25 × 25 cm^2 area and thoroughly mixed together (USDOE, 1992). The samples were taken at depths 5-10 and 50-70 cm from the surface. The collected samples were prepared for radiation counting by sieving through 2 mm mesh size. After that, samples were homogenized and dried in a vacuum drier at 110 °C for 48 h and then weighed and transferred to 1-l Marinelli beakers and stored for more than 1 month to ensure that secular equilibrium between the nuclear chain members had been attained.

Before determining of the radioactivity concentration in samples, an empty 1-l Marinelli beaker was counted for 24 h under identical geometry to measure the background spectrum in the laboratory of measurement. This spectrum is necessary to establish a high confident background level to be used for determination of the specific activities of the analyzed samples.

The experimental arrangement consisted of an HPGe detector with relative efficiency ≈ 10% relative to a 3" × 3" NaL(Tl) detector, active volume 62.3 cm^3 and resolution 1.8 keV at 1.33 MeV γ-line. The detector was coupled through an amplifier to the computer with an MCA plug-in card. Because of the sensitivity of the HPGe detector, it is usual to shield them from the environment. Therefore, a lead shield of thickness 5 cm was used in this work.

The energy calibration of the spectrometer was performed using the 1-l Marinelli calibration sources, which contained well-known standard sources (^{22}Na, ^{60}Co, ^{57}Co, and ^{241}Am). The absolute efficiency of the detector was determined accurately to evaluate the radionuclide concentrations precisely. This was undertaken using multinuclide standard sources distributed in a sand matrix to be homoconditioned with the investigated soil samples.

To determine the radioactivity concentration in the soil samples, each sample was placed on the HPGe detector and counted for the same counting time (24 h). It was found that the detected gamma lines belong to the naturally occurring series radionuclides and a non-series natural radionuclide ^{40}K.

The specific activity concentration of ^{226}Ra was determined indirectly by using the γ-lines 351.90 keV from ^{214}Pb, and 609.32 keV, 1120.28 keV, and 1764.49 keV from ^{214}Bi. For ^{232}Th, the specific activity concentration was determined by using the gamma lines 338.40 keV from ^{228}Ac, 583.14 keV from ^{208}Tl, and 911.07 keV, 964.60 keV, and 968.90 keV from ^{228}Ac. In the case of ^{40}K, the specific activity concentration was estimated directly by its gamma line of 1460.75 keV. The gamma line with energy 46.50 keV was used to determine the activity concentration of ^{210}Pb.

The activities of the radionuclides were calculated from the net area of the peak, accumulation time, absolute peak efficiency at close geometry, absolute γ-ray emission probability, and the sample volume. Triplicate analyses were conducted on all the samples to check the reproducibility of the results and the stability of the counting technique.

The minimum detection limit (MDL) for each activity was calculated using the following equation (Palmer and McInerney 1994):

$$MDL = \frac{A}{NC} \times 3.3\sqrt{B} \quad (1)$$

where NC is the net count for a sample of activity A in Bq/kg, and B is the background count below the peak of interest. The MDL for ^{226}Ra, ^{228}Ra, and ^{40}K was 0.35, 0.20, and 1.2 Bq/kg, respectively.

Results and Discussion

The activity concentrations of radionuclides ^{226}Ra, ^{232}Th, and ^{40}K in the 40 soil samples considered are presented in Table 1. The corresponding results for ^{210}Pb in some soil samples are presented in Table 2. The values are given in Bq/kg on a dry weight basis.

The samples were ordered so that they follow the location along the coast from west to east. It is clear that, in general, the ^{226}Ra and ^{232}Th concentrations at depth 5-10 cm are nearly equal or greater than those at depth 50-70 cm with the exception of some locations (2, 16-20 for ^{226}Ra; and 8, 17-20 for ^{232}Th). The activity concentrations of ^{226}Ra in samples taken at depth 5-10 cm ranged from 4.1 Bq/kg to 13.5 Bq/kg with an average 7.5 ± 2.5 Bq/kg, ^{232}Th ranged from 2.8 Bq/kg to 6.7 Bq/kg with an average 4.5 ± 1.3 Bq/kg, ^{40}K ranged from 19.3 Bq/kg to 39.6 Bq/kg with an average 28.5 ± 6.7 Bq/kg, and ^{210}Pb ranged from 6.1 Bq/kg to 15.7 Bq/kg with an average 10.3 ± 2.7 Bq/kg.

The corresponding results at depths 50-70 cm are within the range from 4 Bq/kg to 11.6 Bq/kg with an average 6.7 ± 1.9 Bq/kg for ^{226}Ra, from 2.9 Bq/kg to 6.5 Bq/kg with an average 4.2 ± 1.1 Bq/kg for ^{232}Th, from 19 Bq/kg to 37.5 Bq/kg with an average 26.6 ± 5.9 Bq/kg for ^{40}K, and from 5.2 Bq/kg to 18.6 Bq/kg with an average 9.2 ± 3.9 Bq/kg for ^{210}Pb.

Most of the northwest Libyan coast at Tripoli is composed of continuous beach sand flat of limestone bedrock with large sand particles size. Therefore, the region under investigation is pervious to the flow of water, since the soil samples were collected from sites mostly in the reach of rapidly spreading water due to the breakage of wave fronts at the shore. Consequently, radionuclides deposited from those transported with sea water during high tide are likely to travel downward through this porous sandy soil surface. The present results shown in Tables 1 and 2 confirm this transport mechanism. The relatively high radioactivity concentrations of both ^{226}Ra and ^{232}Th at depths 5-10 cm (locations 1, 3, 5, and 13) are attributed to the existence of dark colored heavy minerals that contain 2 orders of magnitude more of ^{226}Ra and ^{232}Th, compared to those at depths 50-70 cm.

In the investigated area, the Tajura formation in the west makes steep cliffs along the shore of the Mediterranean, and is made of calcareous rock, including shell fragments and minor sandy grains, interbred with occasional salty lenses. The salty interbreeds, which are often lenticular, are not commonly persistent within the succession. Consequently, the radioactivity concentrations of both ^{226}Ra and ^{232}Th at depths 50-70 cm (locations from 17 to 20) are higher than those at depths 5-10 cm.

The average activity ratio of ^{210}Pb/^{226}Ra is 1.5 for depth 5-10 cm and 1.4 for depth 50-70 cm. The variation in these ratios could be due to the presence of varying degrees of disequilibrium between the members of ^{238}U decay series in the coastal marine sediments or some amounts of different pollutants in the sea water. In addition, the ^{210}Pb (^{222}Rn decay product) fallout flux from the atmosphere and/or geochemical behavior of both Pb and Ra in the environment could explain uranium series (U-Ra-Pb) disequilibrium in the environment. Radium forms a complex with chloride and in this form is quite mobile. Conversely, even moderate amounts of sulfate ion inhibit transport because of the co-precipitation of radium sulfate along with barium sulfate. Almost all lead salts are insoluble or sparingly soluble in water.

The radioactivity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K at Tripoli beach are to a great extent identical to the results of Meijer et al. (2001) concerning North Sea studies (light mineral fraction) at the Western Australia Coast and those of Saad and Al-Azmi (2002) concerning the southern areas of Kuwait, as well as the results of El-Mamoney and Khater (2004) concerning the Red Sea coast and those of El-Kameesy (2007) concerning the Gulf coast northeast of KSA.

Table 1. Activity concentrations in Bq/kg of ^{226}Ra series, ^{232}Th series, and ^{40}K for soil samples collected along the northwest coast of Libya.

Loc. No.	^{226}Ra		^{232}Th		^{40}K	
	D=(5-10) cm	D=(50-70) cm	D=(5-10) cm	D=(50-70) cm	D=(5-10) cm	D=(50-70) cm
1	7.4 ± 1.3	4.1 ± 0.7	5.4 ± 1.0	3.1 ± 0.8	39.6 ± 3.4	31.7 ± 5.4
2	10.6 ± 1.1	11.6 ± 1.3	6.4 ± 0.9	4.2 ± 1.2	39.4 ± 5.3	28.5 ± 4.8
3	13.5 ± 1.4	6.6 ± 1.1	6.7 ± 1.1	6.5 ± 1.1	39.6 ± 8.5	37.5 ± 6.1
4	7.5 ± 0.9	6.4 ± 0.9	4.7 ± 1.2	3.8 ± 1.0	21.9 ± 3.5	19 ± 4.3
5	8.8 ± 1.1	4.6 ± 1.2	5.1 ± 1.1	4.1 ± 0.8	23.8 ± 4.2	31.7 ± 4.8
6	8.9 ± 0.7	7.1 ± 1.4	6.1 ± 1.2	3.1 ± 1.1	28.5 ± 5.1	26.9 ± 4.1
7	6.0 ± 0.8	5.7 ± 1.2	3.9 ± 1.3	3.7 ± 1.3	38 ± 3.4	20.9 ± 3..9
8	8.8 ± 1.4	7.5 ± 0.9	3.3 ± 0.9	4.4 ± 1.2	19.3 ± 4.3	22.2 ± 4.2
9	4.1 ± 0.5	4.0 ± 1.0	3.1 ± 1.0	2.9 ± 0.9	27.1 ± 3.2	36 ± 6.1
10	6.2 ± 0.4	5.5 ± 1.1	4.9 ± 1.1	4.9 ± 1.3	22.2 ± 3.9	19 ± 3.8
11	5.5 ± 0.8	4.3 ± 0.8	3.2 ± 0.8	3.1 ± 1.1	22.1 ± 4.6	20.2 ± 5.1
12	8.4 ± 1.0	6.7 ± 1.2	5.2 ± 1.3	3.2 ± 1.1	24.8 ± 5.2	24.3 ± 4.6
13	9.7 ± 1.4	4.9 ± 1.1	6.3 ± 0.9	5.9 ± 1.4	27.5 ± 6.1	29.6 ± 5.2
14	7.3 ± 1.3	6.2 ± 1.3	5.4 ± 1.2	5.2 ± 0.7	25.8 ± 5.6	26.8 ± 4.6
15	6.9 ± 0.7	6.1 ± 0.9	3.1 ± 0.8	3.1 ± 0.9	35.3 ± 8.3	27.5 ± 3.8
16	5.1 ± 0.5	7.9 ± 0.8	4.2 ± 0.6	3.8 ± 1.0	20.9 ± 5.2	21.6 ± 4.1
17	6.5 ± 0.7	8.3 ± 1.3	2.8 ± 0.7	3.9 ± 1.0	23.7 ± 4.9	21.8 ± 5.2
18	5.2 ± 0.4	7.3 ± 1.1	2.9 ± 1.0	3.9 ± 1.1	29.5 ± 6.1	33.4 ± 3.6
19	8.2 ± 0.8	10.3 ± 1.5	3.8 ± 1.2	6.1 ± 1.5	26.8 ± 8.2	34.3 ± 4.2
20	5.4 ± 0.9	8.1 ± 1.4	2.9 ± 1.1	4.8 ± 1.1	33.4 ± 3.6	20.4 ± 2.9
Average ± *SD	7.5 ± 2.5	6.7 ± 1.9	4.5 ± 1.3	4.2 ± 1.1	28.5 ± 6.7	26.6 ± 5.9

*SD: Standard deviation

Table 2. Activity concentrations of ^{210}Pb (Bq/kg) in some soil samples and the activity ratio of ^{210}Pb/^{226}Ra.

Loc. No.	^{210}Pb (D = 5-10 cm)	^{210}Pb (D = 50-70 cm)	^{210}Pb/^{226}Ra (D = 5-10 cm)	^{210}Pb/^{226}Ra (D = 50-70 cm)
1	8.1 ± 1.2	7.8 ± 1.8	1.1 ± 0.1	1.9 ± 0.3
2	15.7 ± 2.3	18.6 ± 2.1	1.5 ± 0.1	1.6 ± 0.1
7	9.8 ± 2.5	8.6 ± 1.9	1.6 ± 0.2	1.5 ± 0.2
8	12.3 ± 2.9	7.5 ± 1.6	1.4 ± 0.2	1.0 ± 0.1
9	6.1 ± 1.6	5.3 ± 1.1	1.5 ± 0.2	1.3 ± 0.2
10	9.9 ± 2.3	7.7 ± 1.9	1.6 ± 0.2	1.4 ± 0.2
13	8.8 ± 1.3	5.2 ± 0.9	0.9 ± 0.1	1.1 ± 0.1
14	12.6 ± 1.8	11.9 ± 1.1	1.7 ± 0.2	1.9 ± 0.2
17	9.7 ± 1.3	9.3 ± 1.3	1.5 ± 0.1	1.1 ± 0.1
20	10.6 ± 1.2	10 ± 1.8	1.9 ± 0.2	1.2 ± 0.1
Average ± *SD	10.3 ± 2.7	9.2 ± 3.9	1.5 ± 0.3	1.4 ± 0.3

*SD: Standard deviation

The activity concentrations of ^{226}Ra, 7.5 ± 2.5 Bq/kg, and ^{40}K, 28.5 ± 6.7 Bq/kg, in our study are comparable to those of Omar (1997) concerning the northeast beaches of Libya (6.6 Bq/kg and 27.8 Bq/kg), respectively. In contrast to these results, the activity concentration of ^{232}Th in the northeast beaches of Libya (2.0 Bq/kg) is less than those in northwest beaches of Libya (4.5 ± 1.3 Bq/kg).

Table 3. The mean and range activity concentrations in Bq/kg of primordial radionuclides for different countries (soil samples).

No.	Mean activity concentration Bq/kg (range)			Country
	^{226}Ra	^{232}Th	^{40}K	
1	–	4.7 (1.1-1.6)	(17-960)	Australia (Meijer et al., 2001).
2	36 (8-72)	6 (2-17)	227(41-492)	Kuwait (Saad and Al-Azmi, 2002).
3	24.6 (5.2-105.6)	31.4(2.3-221.9)	427.5 (97.6-1011.3)	Red Sea, Egypt (El Mamoney and Ashraf, 2004).
4	–	(1.2-68.4)	(35.8-1315)	Alps-Apennines, Italy (Buttaglia and Bramati, 1988).
5	–	18 (2-96)	316(26-653)	The Nile Delta, Egypt (Ibrahiem et al., 1993).
6	–	2 (1.2-2.8)	23.1 (19.7-33.4)	Northeast Libya (Omar, 1997).
7	(5-130)	(5-185)	(75-1400)	Japan (Megumi et al., 1988).
8	38 (9-62)	38 (16-55)	599 (120-1026)	France (Lambrechts et al., 1992).
9	(9-77)	(5-110)	(11-760)	Bulgaria (Strezov et al., 1998).
10	(27-133)	–	(184-632)	Algeria (Noureddine et al., 1998).
11	–	(4.1-40.2)	(255.9-486)	Cyprus (Tzortzis and Tsertos, 2004).
12	–	25 (7-50)	370(100-700)	World (UNSCEAR, 1988).
13	5.0 (3.0-10.8)	2.1 (0.6-7.8)	46 (10.0-86.1)	Mediterranean Sea (Higgy, 2000).
14	212 (24-764)	43 (18-66)	1130 (258-2464)	Greece (Travidon et al., 1996).
15	-	(60-85)	(42-51)	Portugal (Carreira and Sequeira, 1998).
16	45 (13-165)	49 (7-204)	650 (48-1586)	Spain (Baeza et al., 1992).
17	-	24 (5-63)	(220-3202)	Turkey (Kemru, 1997).
Present work	7.5 (4-13.5)	4.2 (2.8-6.7)	27.5 (19-39.6)	Northwest Libya.

The absorbed dose rate in air 1 m above the ground surface for the radionuclides (^{232}Th, ^{226}Ra, and ^{40}K) was computed on the basis of guidelines provided by UNSCEAR (1988, 2000). The conversion factors used to compute absorbed dose rate (D) in air per unit activity concentration in 1 Bq/kg sand correspond to 0.604 nGy h^{-1} for ^{232}Th, 0.462 nGy h^{-1} for ^{226}Ra, and 0.042 nGy h^{-1} for ^{40}K. Therefore, D could be obtained from the relation:

$$D = (0.604C_{Th}+0.462C_{Ra}+0.042C_K).nGyh^{-1} \quad (2)$$

where C_{Th}, C_{Ra} and C_K are the average activity concentrations of ^{232}Th, ^{226}Ra, and ^{40}K in Bq/kg, respectively.

The range of the absorbed dose rate that obtained for the soil samples is from 2.7 nGy h^{-1} to 6.1 n Gy h^{-1} with an average 4.4 ± 1.3 n Gy h^{-1}, while the average effective dose rate is 0.0054 ± 0.0016 m Sv y^{-1} with the range 0.0033-0.0075 m Sv y^{-1}.

The effective dose rate in units of m Sv y^{-1} can be calculated using the following formula:

$$\text{Effective dose rate} = D(nGyh^{-1}) \times 8760h \times 0.2 \times 0.7SvGy^{-1} \times 10^{-6} \quad (3)$$

where the 0.7 Sv Gy^{-1} and 0.2 factors are the conversion factor from absorbed dose in air to effective dose, and the outdoor occupancy factor, respectively (UNSCEAR, 1988).

In Table 3, a summary of results on elemental concentrations is given, derived from similar investigations conducted in other countries worldwide with emphasis on regions.

Conclusion

In this study the radioactivity concentrations for ^{226}Ra, ^{232}Th, ^{40}K, and ^{210}Pb were measured in 40 soil samples from the northwest Libyan coast in the Tripoli region using gamma spectroscopy. It was observed that the activity concentrations of ^{226}Ra, ^{232}Th, ^{40}K, and ^{210}Pb 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, and 10.3± 2.7 Bq/kg, respectively. The corresponding results at depths 50-70 cm are 6.7 ± 1.9 Bq/kg for ^{226}Ra, 4.2 ± 1.1 Bq/kg for ^{232}Th, 26.6 ± 5.9 Bq/kg for ^{40}K, and 9.2 ± 3.9 Bq/kg for ^{210}Pb. In contrast to these results, the activity concentration of ^{232}Th in the northeast beaches of Libya (2.0 Bq/kg) is less than that in the northwest beaches of Libya (4.5 ± 1.3 Bq/kg) (Omar, 1997). Furthermore, the average activity ratio of ^{210}Pb/^{226}Ra, which is greater than 1, implies that there is a possibility that some pollutants come with the sea water, which needs more detailed investigation on the pollution sources and the environment distribution pattern of different pollutants.

The range of the absorbed dose rate obtained for the soil samples is from 2.7 n Gy h^{-1} to 6.1 n Gy h^{-1} with an average 4.4 ± 1.3 n Gy h^{-1}, while the average effective dose rate is 0.0054 ± 0.0016 m Sv y^{-1} with the range 0.0033-0.0075 m Sv y^{-1}.

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