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Investigation of Radioactivity Levels and Evaluation of Radiation Hazards in Soil Samples Collected from the Area Extended from Agdabya to Qaminis, Libya.

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Highlights

- **This monitoring program aims at the determination of natural and man-made radioactivity levels that will help establish a radiological map of Libya.**
- **The results obtained may be considered as a base for future studies.**
- **Measurements were carried out using gamma spectroscopy.**

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ABSTRACT

In order to initiate a radiological assessment program and to establish a baseline map of radioactivity background levels in the area extended from Agdabya to Qaminis in Libya environment, this study has been adopted to identify the radionuclide contents in the soil of this area. Thirty soil samples were collected at a depth of 5 cm in the area extended from Agdabya to Qaminis, measurement of radioactivity and (pH). Measurement of the radioactivity of the Uranium ${}^{238}_{92}U$, Thorium ${}^{232}_{90}Th$, potassium ${}^{40}_{19}K$ and Cesium ${}^{137}_{55}Cs$ were done at the Atomic Energy Center of Tagora in Libya using Hyper Pure Germanium detector. The average radioactivity concentration for ${}^{238}_{92}U$, ${}^{232}_{90}Th$, ${}^{40}_{19}K$ and ${}^{137}_{55}Cs$ were 81.32 Bq/Kg, 32.71Bq/Kg, 184.46Bq/Kg and 2.03Bq/Kg. The variations of the assessed radiological hazard parameters indices Ra_{eq} , H_{ex} , H_{in} , I_γ, I_α and R_D were found to be as follows: 94.89-258.37, 0.26-0.698, 0.37-1.197, 0.33-0.898, 0.175-0.92 and 42.96-120.03, respectively. The pH values ranged from 7.70 to 9.45, with an average of 8.44.

1. Introduction

The human body is exposed to radiation from different sources, including cosmic rays; natural radionuclides; man-made radioactivity from nuclear tests; and applications in medicine. External exposure to gamma radiation comes from natural radionuclides and cosmic radiation, while internal ones come from inhalation and intake through drinks and food (Abdul Adziz and Khoo, 2018). Potassium-40 (${}^{40}_{19}K$) (E=1460 keV), radionuclides belong to ${}^{238}_{92}U(E_{max}$=1764.5 keV) and ${}^{232}_{90}Th(E_{max}$=2614 keV) series, are contaminating soil, rocks, and water by different concentrations due to geological and geographical differences in all over the world (Alshahri and El-Taher, 2019) (Badawy *et al.*, 2013).

The risk from Cs-137 (E=661.6 keV) varies with its diffusion rates in soil. If Cs-137 migrate slowly in soil, the internal irradiation will be higher due to higher absorption by plant roots especially from the top surface of 5 cm depth. However, if Cs-137 diffuse rapidly, the uppermost soil surface acts as a shield against radioactivity found in deeper soil layers and therefore the external radiation will be less (Ahmad *et al.*,2019) (Karataşlı *et al.*, 2016).

In this study, the focus was on natural and manmade radionuclides, Uranium (${}^{238}_{92}U$), Thorium (${}^{232}_{90}Th$), potassium (${}^{40}_{19}K$), and Cesium (${}^{137}_{55}Cs$) by measuring their radioactivity concentrations in soil and evaluation of radiation hazards. Samples of soil were collected from different locations from Ajdabya to Qaminis, Libya. These samples were investigated for their soil chemical characteristics [(pH)]. For Gamma spectroscopy, a gamma-ray spectrometer (HPGe) was the main tool.

2. Materials and Methods:

2.1. Sampling Area

Libya is located in the north of Africa on the Mediterranean coast, it encompasses a geographical area estimated at (1759540 km^2) between (19.30–33°N) and (9.30–25°E) and more than 90% of the country is desert (Bauer *et al.*, 2017). It extends from the Mediterranean in the north to the borders of Niger and Chad in the south, and from the borders of the Egyptian region and Sudan in the east to the borders of Tunisia and Algeria in the west. The elevation ranges from 59 m to 2,314 m. The Libyan climate is characterized by hot, dry summers and mild winters (ELKenawy *et al.*, 2009). The total population amounts to about five million in 1998. The rainfall in the northern part of the country varies between 100–500 mm/year but the southern section receives only as much as 10 mm/year and some parts are completely rainless (Wheida and Verhoeven, 2007). Rainfall is generally concentrated in a short period of the year, usually from October to November on the coast and as late as March or April in the desert (Bauer *et al.*, 2017).

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Table 1

The GPS sites for the soil samples location.

Sample	Latitude	Longitude	Elevation (m)
1	30°45'27.92"N	20°13'13.02"E	3
2	30°46'54.51"N	20°13'41.96"E	5
3	30°48'33.41"N	20°14'4.11"E	6
4	30°50'8.87"N	20°14'23.96"E	16
5	30°51'33.97"N	20°13'23.37"E	15
6	30°52'58.33"N	20°12'22.78"E	11
7	30°54'23.94"N	20°11'21.64"E	11
8	30°55'59.24"N	20°11'1.97"E	6
9	30°57'37.34"N	20°11'3.83"E	7
10	30°59'0.43"N	20°11'4.38"E	6
11	31° 0'36.75"N	20°11'29.42"E	7
12	31° 2'10.47"N	20°12'5.64"E	9
13	31° 4'1.56"N	20°12'49.48"E	8
14	31° 6'5.82"N	20°13'26.92"E	15
15	31° 8'0.04"N	20°13'42.60"E	11
16	31° 9'38.50"N	20°13'49.39"E	7
17	31°11'17.72"N	20°13'48.82"E	9
18	31°13'2.21"N	20°13'39.25"E	7
19	31°15'3.22"N	20°13'15.54"E	8
20	31°17'0.26"N	20°12'51.95"E	15
21	31°18'38.84"N	20°12'22.99"E	16
22	31°19'59.31"N	20°11'58.78"E	19
23	31°22'41.75"N	20°11'13.01"E	27
24	31°24'44.13"N	20°10'26.94"E	28
25	31°26'41.29"N	20° 9'32.82"E	28
26	31°28'11.76"N	20° 8'18.12"E	23
27	31°29'55.27"N	20° 6'50.02"E	18
28	31°31'38.51"N	20° 5'25.59"E	16
29	31°33'15.56"N	20° 4'30.49"E	18
30	31°34'43.02"N	20° 3'41.74"E	20

Note: *Sample 1: was taken from the soil surrounding the Fatimid palace in Ajdabiya*

Fig. 1. The geographical map for all sites of samples using Google Earth

2.2 Samples Collection and Preparation

Thirty soil samples were taken from the area that lay from Agdabya to Qaminis in February in the year of 2018 using the template method (Isaksson, 1997) that is the usual application of this method is to scrape or shovel off layer after layer of soil within a chosen area, which could be defined by some sort of rigid frame, in some cases pressed down into the soil to a certain depth. The area sample was cut out using a template of 25 cm 25 cm for guidance to a depth of 5 cm (Isaksson, 1997).

All soil samples were cleaned from stones and organic matter, they were left to dry in an oven at 80°C for 24 hours. After drying they were crushed and passed through a 2-mm sieve. Their weights were measured and then kept in plastic bags. The meshed soil samples were packed in 500 mL Marinelli beakers and kept sealed for four weeks to attain radioactive equilibrium before being measured.

pH: Thirty soil samples were analyzed by weighing 30 g of air-dried soil into a beaker and then adding 60 mL of double-deionized (d.d.) water. The soil-to-solution ratio used was 1:2. After that the suspension intermittently for 30 min. Then let stand for about 1 h, after this time we immersed the electrode into the clear supernatant and recorded the pH once the reading was constant (Both the glass membrane and the porous salt bridge must be immersed) (Carter and Gregorich, 2008)

2.3. Samples Analysis

The activity concentrations of the radionuclides in the studied samples were measured using a gamma-ray spectrometer with a coaxial p-type HPGe detector having a relative efficiency of 50%. It has an energy resolution of 1.89 keV for the 1332.5 keV ${}^{60}_{27}Co$ gamma-ray line. The detector was shielded using a 10-cm thick low-background lead shield. The amplified signals of the detector were acquired with a 16 K analog-to-digital converter multichannel analyzer (Genie 2000, Canberra, Australia). Each soil sample was placed on the top of the detector. The measuring times ranged from 18,000 to 100,000 s to provide adequate counts under the various gamma-ray photo peaks. Background measurements were taken under the same conditions as sample measurements and subtracted to get net counts for the sample. The energy and efficiency calibrations of the detector were performed using calibration sources *(Elnimr et al.,* 2017).

The energy calibration of the MCA was obtained using standard point sources such as ${}^{22}_{11}Na$, ${}^{57}_{27}Co$, ${}^{60}_{27}Co$, ${}^{133}_{56}Ba$, ${}^{137}_{55}Cs$, etc. The efficiency of the detector for different radionuclides of interest of different energies was determined by mixing standard sources of known activities and different energies such as 122, 245, 344, 411, 444, 779, 963, 1086, 1112 and 1408 keV supplied by Health Physics Division, Atomic Energy Centre, Dhaka and following the standard method. The unknown efficiencies of different radionuclides were then calculated using Eq. (3-1) to draw a standard efficiency curve (Figs. 3-4) (Ferdous *et al.*, 2015). The efficiency calibration curve was drawn up using different energy peaks covering a range of up to 2000 keV to obtain the efficiency of the detector for the particular gamma-ray energy of interest (Ferdous *et al.*, 2015).

Radium equivalent (Ra_{eq}), external hazard index (H_{ex}),internal hazard index (H_{in}), gamma index (I_{γ}), alpha index(I_{α}) and the radiation dose (R_D) were calculated using equations below (Mouandza *et al.*, 2018) (Zubair *et al.*, 2013) (Sowole, 2014) (Fares, 2017) (Ahmed *et al.*, 2019) (Zubair *et al.*, 2013) :

$$Ra_{eq} = A_{Ra} + \frac{10}{7}A_{Th} + \frac{10}{130}A_k \tag{1}$$

$$R_D = 0.462A_u + 0.6044A_{Th} + 0.0417A_k \tag{2}$$

$$H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_k}{4810} \tag{3}$$

$$H_{in} = \frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_k}{4810} \tag{4}$$

$$I_{\gamma} = \frac{A_u}{300} + \frac{A_{Th}}{200} + \frac{A_k}{3000} \quad (5)$$

$$I_{\alpha} = \frac{A_u}{200} \quad (6)$$

2. Results and Discussions

As seen in Table 2, the concentrations $^{238}_{92}U$ and $^{232}_{90}Th$ ranged between (35.09 to 184.69) Bq/Kg and (12.43 to 65.77) Bq/Kg, respectively. The activity concentration values of $^{40}_{19}K$ and $^{137}_{55}Cs$ ranged from (106.82 to 390.50) Bq/Kg and (0.00 to10.10) Bq/Kg, respectively. The average activity concentration of, $^{232}_{90}Th$,$^{40}_{19}K$and $^{137}_{55}Cs$were (81.32, 32.71, 184.46 and 2.03) Bq/Kg, respectively.

Table 2

Activity concentration for the measured radionuclides of $^{238}_{92}U$, $^{232}_{90}Th$, $^{40}_{19}K$and $^{137}_{55}Cs$in the soil samples at depth 5 cm

Samples	$^{238}_{92}U$(Bq/Kg)	$^{232}_{90}Th$(Bq/Kg)	$^{40}_{19}K$(Bq/Kg)	$^{137}_{55}Cs$(Bq/Kg)
1	184.69	30.50	390.50	10.10
2	124.20	47.20	215.31	7.21
3	112.21	20.32	170.70	2.40
4	136.3	12.43	166.32	3.23
5	80.51	23.40	165.32	1.50
6	76.15	19.32	231.78	1.37
7	67.30	19.73	115.13	0.96
8	130.48	22.39	137.05	0.55
9	113.95	41.51	154.35	0.35
10	118.12	22.25	136.25	1.02
11	99.58	20.01	227.47	0.82
12	50.60	44.31	284.14	2.36
13	74.44	28.85	198.91	0.92
14	62.64	27.68	169.11	1.47
15	69.64	31.34	189.10	0.81
16	43.95	31.90	180.20	0.63
17	74.69	65.77	199.57	0.80
18	73.41	51.84	182.08	0.00
19	49.65	39.50	106.82	0.85
20	43.38	32.24	107.72	1.08
21	50.30	49.30	166.50	1.80
22	96.40	27.87	170.04	1.70
23	76.32	31.53	166.90	1.83
24	113.88	36.64	153.51	2.48
25	82.00	32.80	167.45	0.89
26	63.00	33.02	194.41	2.99
27	58.05	31.80	203.85	1.19
28	40.85	30.36	137.93	0.49
29	37.86	40.29	208.73	5.75
30	35.09	35.06	236.71	3.39
Average	81.32	32.71	184.46	2.03

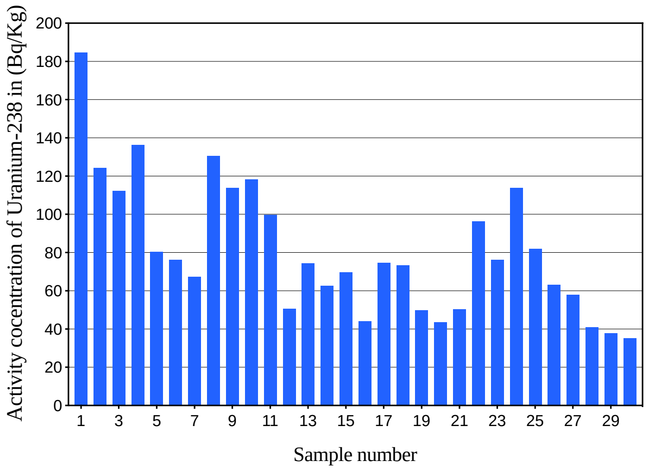

Fig. 2. The Activity concentration of ($^{238}_{92}U$) in (Bq/Kg) for the soil samples.

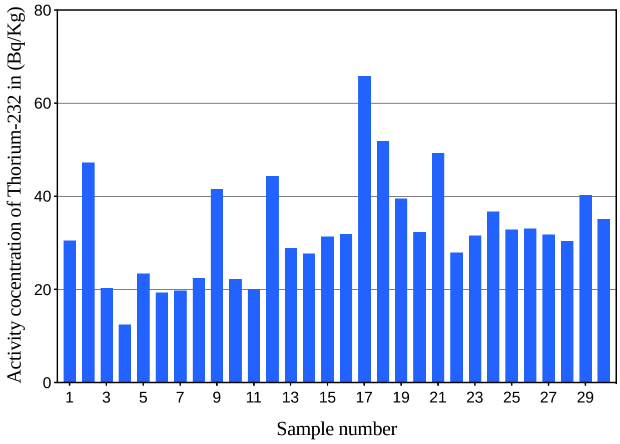

Fig. 3. The Activity concentration of ($^{232}_{90}Th$) in (Bq/Kg) for the soil samples.

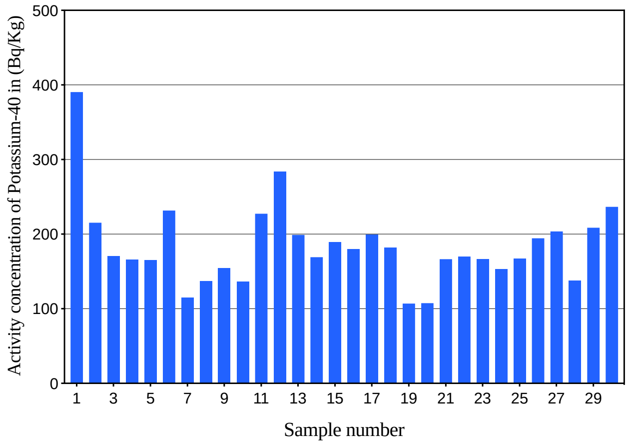

Fig. 4. The Activity concentration of ($^{40}_{19}K$)in (Bq/Kg) for the soil samples.

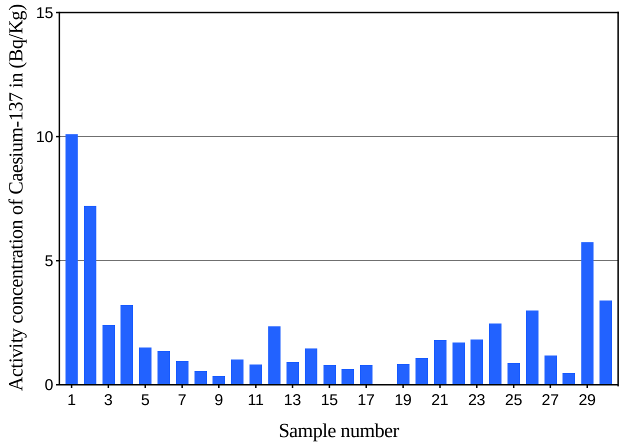

Fig. 5. The Activity concentration of ($^{137}_{55}Cs$) in (Bq/Kg) for the soil samples.

pH measurements are shown in Table 3 It is found that the values of pH ranged from 7.70 to 9.45, with an average of 8.44.

From Table 4 it is found that the radium equivalent (Ra_{eq}) ranges between (94.89 to 258.37) Bq/Kg, with an average value of 142.29 Bq/Kg. External and internal hazard indices ranged from (0.26 to 0.698) and (0.37 to 1.197), respectively, the average values for external and internal hazard indices are (0.38 and 0.60, respectively. For Gamma and Alpha indices values ranged between (0.33 to 0.898) and (0.175 to 0.92), respectively, the average values for Gamma and Alpha indices are (0.496 and 0.406), respectively. The radiation dose ranged from (42.96 to 120.03) nGy/h, with an average of 65.02 nGy/h. The Cs-137 activity was not taken into account because of the limited contribution to the total exposure rate (Gillard *et al.*, 1989).

Table 3

pH Measurements:

Sample №	pH	Sample №	pH	Sample №	pH
1	8.81	11	8.08	21	8.50
2	9.29	12	8.47	22	8.44
3	9.45	13	8.02	23	8.15
4	9.28	14	8.67	24	8.43
5	8.30	15	8.44	25	8.46
6	7.70	16	8.32	26	8.40
7	8.59	17	8.44	27	8.31
8	7.90	18	8.04	28	8.47
9	8.30	19	8.61	29	8.20
10	8.66	20	8.24	30	8.19
Average pH=8.44					

Table 4

The values of radium equivalent, radiation dose, external and internal hazard indices, and gamma and alpha indices in the soil samples at a depth 5 cm.

Sample	Ra_{eq}(Bq/Kg)	H_{ex}	H_{in}	I_γ	I_α	R_D(nGy/h)
1	258.3735	0.6981	1.1973	0.8983	0.92345	120.03263
2	208.27487	0.5627	0.8984	0.72177	0.621	94.867627
3	154.4115	0.4172	0.7205	0.5325	0.56105	71.23249
4	166.88154	0.4509	0.8193	0.5719	0.6815	77.413864
5	126.70164	0.3423	0.5599	0.4405	0.40255	58.223064
6	121.62466	0.3286	0.5344	0.4277	0.38075	56.515806
7	104.37891	0.2820	0.4639	0.3614	0.3365	47.810441
8	173.05055	0.4676	0.8202	0.5926	0.6524	79.520305
9	185.19425	0.5003	0.8083	0.6388	0.56975	84.153335
10	160.42875	0.4335	0.7527	0.5504	0.5906	73.692065
11	145.70949	0.3937	0.6628	0.5078	0.4979	67.577499
12	135.84208	0.3669	0.5037	0.4849	0.253	61.989078
13	131.01157	0.3539	0.5551	0.4587	0.3722	60.111227
14	115.24387	0.3113	0.4806	0.40357	0.3132	52.710287
15	129.0169	0.3485	0.5368	0.4519	0.3482	58.98851
16	103.4424	0.2794	0.3982	0.3661	0.21975	47.08684
17	184.10799	0.4973	0.6992	0.6443	0.37345	82.553929
18	161.56136	0.4364	0.6348	0.5646	0.36705	72.819516
19	114.36014	0.3089	0.4431	0.3986	0.24825	51.250694
20	97.77764	0.2641	0.3814	0.3417	0.2169	44.006444
21	133.6195	0.3609	0.4969	0.4697	0.2515	59.95885
22	149.34718	0.4035	0.6640	0.5174	0.482	68.460948
23	134.2592	0.3627	0.56898	0.4677	0.3816	61.26369
24	178.09547	0.4812	0.7889	0.61397	0.5694	81.144487
25	141.79765	0.3831	0.6047	0.4931	0.41	64.677865
26	125.18817	0.3382	0.5084	0.4399	0.315	57.156977
27	119.22045	0.3221	0.4789	0.42045	0.29025	54.526845
28	94.88541	0.2563	0.3667	0.3339	0.20425	42.961821
29	111.54691	0.3013	0.4036	0.3972	0.1893	50.530521
30	103.45247	0.2794	0.3743	0.3712	0.17545	47.258627
Average	142.294	0.384	0.604	0.496	0.406	65.02

4. Conclusion

The international recommended limits for $(^{238}_{92}U)$, $(^{232}_{90}Th)$ and $(^{40}_{19}K)$ are (35,30 and 400) Bq/Kg, respectively (UNSCEAR,2000). The results showed that in all samples the concentration of $(^{238}_{92}U)$ was higher than international recommended limits and the sample1 showed the highest value at the level of all samples. For $(^{232}_{90}Th)$ the samples (3, 4, 5, 6, 7, 8, 10, 11, 13, 14, and 22) showed lower concentrations, the rest of the samples had higher concentrations than international permitted values and as the sample17 showed the highest value at the level of all samples. For Potassium $(^{40}_{19}K)$, the concentration in all samples were lower than international recommended limits except sample 1 that is considered the highest one, but it is still within international recommended limits. For (Cs-137) sample 1 showed the highest value at the level of all samples, that was 10.10 Bq/Kg.

Radium equivalent in all samples was lower than the international recommended limit which is about 370 Bq/Kg (Najam *et al.*, 2015). The external hazard index for all samples was less than the unity value while the allowed value is unity. On the other hand, the internal hazard index for the sample was higher than unity. The rest of the samples were less than unity (El-Gamal& El-Haddad, 2019). For the Gamma and Alpha index, all samples were less than unity while the allowed limit is a unity(Ahmed Najam et al., 2017). The recommended value for the absorbed Gamma dose rate in air given by (UNSCEAR, 2000) is 55 nGy/h. Absorbed Gamma dose rate in air in samples 1, 2, 3, 4, 5, 6, 8, 9, 10, 11, 12, 13, 15, 17, 18, 21, 22, 23, 24, 25, 26 were higher than the recommended value limit.

It was noted that the concentration of , $(^{40}_{19}K)$,$(^{137}_{55}Cs)$, internal hazard index and the radiation dose in the first sample showed the highest value at the level of all samples, this is due to the rocky soils that formed the monuments. It is known that igneous rocks of granitic compositions (granodiorite, monzogranites, syenogranites and alkali feldspar granite) and their equivalent volcanic rocks (rhyolite, rhyodacite, dacite etc) are strongly enriched in the radionuclides. Radionuclide distribution is not the same in all rock-forming minerals. They sometimes create their minerals like uraninite, thorite, and K-feldspar and sometimes geological processes cause their accumulation in some minerals also. Th and U are accumulated in xenotime, zircon, allanite, apatite, sphene, and epidote among others; K is accumulated in biotite, muscovite, and orthoclase (Heikal & Top, 2018) (Szczepaniak, 2014).

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