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NATURAL RADIOACTIVITY AND ASSOCIATED DOSE RATES OF SOIL SAMPLES IN DIFFERENT LIBYAN SIDES

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Abstract The activity concentrations of ²³⁸U, ²³⁵U, ²²⁶Ra, ²³²Th and ⁴⁰K were measured using HPGe detector for soil samples collected from western and Mid Libya .

The average activity concentration of ²²⁶Ra, ²³²Th and ⁴⁰K for all 20 samples were found to be 39.81±9.28, 26.58±4.82 and 31.05±67.6 Bg/kg while ²³⁵U was detected only for first 6 samples in the average 96.16±19.69Bg/kg. The results obtained for the corresponding nuclides ²²⁶Ra, ²³²Th are slightly agreed with the world wide average values of 30, 45^[1] respectively while ⁴⁰K was smaller than worldwide average (400Bq/kg). The average outdoor absorbed dose and the annual effective dose rates due to 226 Ra, 232 Th and 40 K were observed to be 47.02±7.9nGy/h,57.66±6.69µSv/y,82.5±16 Bg/kg and 0.27±0.06 respectively which acceptable and are less than the world average by 10 %.

Keywords Activity concentration, Absorbed Dose, Effective Dose, world average Activity values.

1. Introduction

The natural radioactivity in the environment is the main source of radiation exposure for human body. Natural radionuclide in soil contributes a significant amount of background radiation exposure to the population through inhalation and ingestion.

The activity concentration of ²²⁶Ra,²¹⁴Bi,²¹⁴Pb are belong to ²³⁸U series ,²²⁸Ac, ²¹²Bi,²¹²Pb are belong to ²³²Th series and ⁴⁰K were measured using HPGe spectrometer .

According to A.L.A.R.A, principle, the radium equivalent ,the external hazard index, the absorbed dose and the annual effective dose were estimated and compared with results of other studies with the world wide average value This work was undertaken to measure the activity concentrations and γ-ray absorbed doses of the naturally occurring radionuclides NORM in soil samples another aim of this work is to create the public awareness about the radiation hazards. This work will also be helpful to establish a research base line in the investigated regions.

2. Materials and Methods

2-1 Sample collection and Preparation

The samples are selected from different geographic and geological regions in western and mid Libya , for each region was taken tow samples A and B with distance about 20km ,the masses of the collected samples varied between 0.300 kg and 0.530 kg .

The samples were collected from western and mid Libya as shown in map **Figure.1** at depth 1-10cm and packed in bag sealed and charged to Cairo by air on October 2014 .The samples were placed in dry place until march 2016 then treated thermally at 60 ^oC for 24 hours after that the samples were sieved to obtain uniform particle size.



Figure.1 Distributed Samples on Map of Libya

Then the soil will filled in beaker which was sealed and then weighted and stored for more than month to reach the secular equilibrium.

2-2 Gamma –ray detector System

In γ -ray spectrometry, the full energy peak efficiency of a (HPGe) detector is the number of γ -rays detected by the detector to the number of photons emitted by the source for a specific energy, which is defined as ^[3,5]

$$\varepsilon(E) = \frac{N_P(E)}{A I_{\gamma}} \qquad (1)$$

Where $N_P(E)$ is the count rate of the photo peak for the corresponding energy E , A is the present activities of the standard reference source and $I\gamma$ is the intensity of gamma energy .The detector has a photo peak relative efficiency of about 30% and an energy resolution of 1.91keV FWHM for 1332keV gamma transition of ⁶⁰Co .The detector was calibrated in absolute efficiency using mixed radionuclides gamma standard inside Enshas laboratory **Figure 2**



Figure.2 The absolute energy peak efficiency measurement using the standard sources ²²⁶Ra and ⁶⁰Co

2-3 Activity Concentrations of Soil Samples

The radioactivity of each sample was measured using HPGe detector of energy resolution 2.0keV at1332keV of 60 Co for a period 8600s .Keeping the samples one by one on the top of the detector and counted for a period of 8600s . The activity concentration (A) of each radionuclide in the sample was determined by using the count rates (N_c) (after subtracting the back ground counts)^[1,3]

$$A = \frac{N_C}{A.I_{\gamma}W} \qquad (2)$$

Where A = Activity concentration of the sample (Bq/kg)

 N_c = Net count rate =Gross counts per second from the samples-Background counts per second, ϵ =Efficiency of the detector for the specific energy

 I_{y} = Intensity of the gamma ray, W= Sample weight (kg). For the analysis of peak areas of gamma spectra, a computer software programming (Gene2000) was used ^[14].

2-4 Radium Equivalent Activity

The important radionuclides in nature ²²⁶Ra , ²³²Th and ⁴⁰K are not uniformly distributed ,this due to disequilibrium between ²²⁶Ra and its decay products .

For uniformity in exposure, estimates the radionuclide concentrations have been defined in terms of radium equivalent activity (Ra_{eq} in Bq/kg). This allows comparison of the specific activity of materials containing different amounts of ²²⁶Ra , ²³²Th and ⁴⁰K according to^[3,12]

$$Ra_{eq} = A_{Ra} + 1.43A_{Th} + 0.077A_{K}$$
 (3)

2-5 External Hazard Index

The external hazard index (H_{ex}) is the indoor radiation dose rate due to the external exposure gamma radiation construction materials which was calculated by ^[8,9]

$$Hex = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_{K}}{4810}$$

(4)

2-6 Absorbed Dose Rates

The external outdoor absorbed gamma dose rates due to terrestrial γ -rays from the nuclides of ²²⁶Ra , ²³²Th and ⁴⁰K at 1m above the ground level was $D = (0.461A_{Ra} + 0.623A_{Th} + 0.0414A_{K})nGyh^{-1}$ calculated as [2]

(5)

About 98% of the external γ dose **rate** from ²³⁸U **series** is delivered by the ²²⁶Ra sub series .So disequilibrium between ²²⁶Ra **and** ²³⁸U will not affect the results of dose calculations from the measurement of ²²⁶Ra.

2-7 Annual Effective Dose Rates

The absorbed dose rate was converted into annual effective dose equivalent by using conversion factor of 0.7SvGy⁻¹ and0.2 for the outdoor occupancy factor by considering that the people on the average spent20% of their time in outdoors. The Effective dose due to natural activity in soil was calculated by^[10]:

 $(E.D = D \times 8760 \times 0.2 \times 0.7 \times 10^3) \mu S v y^{-1}$ (6)

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2-8 Representative level index (Iyr)

This index is used to estimate the level of γ –radiation hazard associated with the natural radionuclides in specific investigated samples , is defined as ^[3]

$$I_{\gamma r} = \frac{A_{Ra}}{150} + \frac{A_{Th}}{100} + \frac{A_{K}}{1500}$$
 (7)

2-9 the Ratio of 238 U/ 232 Th

The ratio of 238 U/ 232 Th concentration activities is less than 1 because the concentration activity of 238 U in soil is less than the concentration of 232 Th .For the investigated samples most their uranium /thorium ratio less than 1

3. Results and Discussion

In this work of soil samples of some sites in Libya the results can be summarized as:

i) Activity concentrations , and ii) Radiological indices

3-1 Activity concentration in soil samples

Activity concentrations for nuclides ²³⁵U, ²³⁸U²²⁶Ra, ²³²Th and ⁴⁰K was determined by equation (2) and the results were tabulated in Table 1. Here the table shows that the highest value is found for sample S6A for ²³⁸U and ²²⁶Ra, S10A for ²³⁸U and ²³²Th and S1A is very high for ⁴⁰K. The highest of the nuclide may vary from place to place in this variation may be due to chemical changes in elements of soil or using agriculture fertilizers or material wastes, weaponsetc. The results for these nuclides also shown independently through Fig3,4,**5**&**6**

S.NO	²³⁵ U	²³⁸ U	²²⁶ Ra	²³² Th	⁴⁰ K	²³⁸ U/ ²³² Th
S1A	160.5±28.8	74.5 <u>+</u> 11.3	76±8.3	63.6±8.5	1910.7±431.2	1.17
S2A	72±10.8	13.7 <u>+</u> 2.6	13±6.2	17.4±2.1	216±42.4	0.78
S3A	14.4±36.6	9.8 <u>+</u> 3.5	6±7.8	4.6±5.0	146.4±26.5	2.13
S4A	31.6±28.9	16 <u>+</u> 2.0	14±2.7	10.2±3.8	242.4±48.5	1.56
S5A	67.5±12.8	19.4 <u>+</u> 1.3	19±4.8	17.7±2.0	255.4±51.5	1.09
S6A	231±60.3	18.2 <u>+</u> 1.6	22±1.6	31.7±1.2	223.6±44.2	0.57
S7A		18.4 <u>+</u> 1.5	13±6.2	12.9±3.1	247±49.5	1.42
S8A		20 <u>+</u> 1.2	20±1.19	28±0.32	364±76.4	0.71
S9A		9 <u>+</u> 3.7	5±8.0	4±5.2	66.5±8.1	2.25
S10A		103.8 <u>+</u> 18.1	124±19.3	118.3±21.0	N.D	0.87
S1B		8.4 <u>+</u> 3.8	11±6.6	10.2±3.8	382±80.5	0.823
S2B		10.5 <u>+</u> 3.4	9.5±7	10.2±3.8	23.8±1.7	1.02
S3B		8.6 <u>+</u> 3.8	12.3±6.3	5.6±4.8	177.3±33.5	1.53
S4B		7.3 <u>+</u> 4.1	8.2±7.3	8.6±4.12	272.6±55.4	0.84
S5B		12.9 <u>+</u> 2.8	14±5.9	19.9±1.5	197.8±38.3	0.64
S6B		14.7 <u>+</u> 2.4	13.5±6.0	23.9±0.61	362.9±76.13	0.61
S7B		15.2 <u>+</u> 2.3	14.8±5.7	15.1±2.6	380.3±80.1	1.006
S8B		17.5 <u>+</u> 1.7	16.5±5.3	23.7	315.8±65.3	0.73
S9B		5.8 <u>+</u> 4.4	4.7±8.1	4.6±5.0	64.5±7.7	1.26
S10B		98.7 <u>+</u> 16.9	87.7±11	101.4±17.2	N.D	0.97
Average	96.16±29.7	25.12±4.0	25.21±4.86	26.58±4.8	31.05±67.6	1.103
Max	231±28.8	103.8±18.5	237±45.2	118.3±21.0	1910.7±431.2	2.25
Min	14.4±10	5.8 <u>+</u> 4.4	4.7±8.1	4±5.2	23.8±1.7	0.57

Table.1 The activities concentrations of the ²³⁵U, ²³⁸U, ²²⁶Ra, ²³²Th and ⁴⁰K in Bq/kg for the measured samples.

N.D = Not detected

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²²⁶Ra activity

In soil samples the activity concentrations of ²²⁶Ra were found in the range of4.7±1.7-237±45.24 Bq/kg, with an average value 39.81±9.28 Bq/kg. This result is slightly higher than the world wide average value of 35Bq/kg for the same radionuclides in soils reported by UNSCEAR^[11]

3.2 ²³²Th Activity

The concentration of ²³²Th is found in the range $4\pm0.32-118\pm21.04$ Bq/kg with mean value 26.58±4.82 Bq/kg this result is lower than the world average of 30 Bq/kg

3.3 ⁴⁰K Activity

The activity ⁴⁰K is found in the range23.8 \pm 1.7-1910 \pm 431.2Bq/kg with the average value of 31.05 \pm 67.6 Bq/kg. This result is mostly lower than the world wide average of 400Bq/kg but S1A is very high for the same kind of nuclide ^[3,11]

3.4 ²³⁵U Activity

The concentration was detected in 6 samples in the range14.4±10 -231±60 Bq/kg with average 96.16±29.69Bq/kg. For sample SA9 (Miserata) was found energy line 187 keV between 185.7 keV ²³⁵U(I γr 0.572) and 186.2 keV ²²⁶Ra(I_{γr}0.0036), there is difficulty to detect these peaks, so that the concentration of ²³⁵U was calculated according this relation^[15]:

$$A_{235_{U}} = \frac{\left[\frac{(CR_{187}M)}{\varepsilon_{Peak}} - A_{226_{Ra}} \times I_{226_{Ra}}\right]}{I_{235_{U}}}$$

Where CR_{187} is the count rate of the peak centered at 187 keV, ϵ_{Peak} is the detector efficiency at that energy ,M is the mass of sample (kg). From data : CR_{187} =0.013 count/s, M=0.53kg

 $I_{\gamma(235}_{U)} = 0.572$, $\epsilon_{Peak} = 0.014081$, $I_{\gamma(226}_{Ra}) = 0.056$ and $A_{(226}_{Ra}) = 3.14$ Bq/kg the result is very small 0.66 Bq/kg can be neglected

3.5 Radiological indices

To estimate the health effects ,the radiation hazards such as radium equivalent (Ra_{eq}),external hazard index (H_{ex}) ,absorbed dose Rate (D) ,effective dose rate (ED) and representative level index ($I_{\gamma r}$) have been calculated from the activity of nuclides ²²⁶Ra ,²³²Th and ⁴⁰K using the equations (3-7)respectively and the values have shown in Table 2



Table.2 Radium equivalent activity Ra_{eq} ,Absorbed dose rate D, Effective dose rate , External hazard index H_{ex} andRepresentative levelindex I_{yr} .

Sample.ID	Ra _{eq} activity	Absorbed dose	Effective dose	External	Representative	
-	(Bq/kg)	D(nGy/h)	rate	hazard	Level index $I_{\gamma r}$	
		-	$ED(\mu Sv/y)$	index H _{ex}	Bq/kg	
S1A	200.15±26.99	153.76±24.50	188.57±30.03	0.85±0.13	2.41±0.41	
S2A	41.15±9.49	25.78±4.87	31.61±5.98	0.15±0.16	0.41±0.05	
S3A	14.62±15.57	11.69±8.10	14.34±9.94	0.06±004	0.21±0.1	
S4A	97.32±3.40	52.81±1.33	64.76±1.63	0.30±0.01	0.37±0.06	
S5A	48.27±7.85	30.36±3.82	37.23±4.69	0.17±0.04	0.48±0.03	
S6A	285.73±46.62	138.26±20.93	169.57±25.67	0.81±0.04	0.59 ± 0.01	
S7A	35.26±10.83	24.26±5.22	29.75±6.40	0.14±0.03	0.42 ± 0.05	
S8A	77.92±1.05	47.27±0.06	57.97±0.07	0.27±0.06	0.66 ± 0.05	
S9A	11.35±16.32	7.55±9.05	9.26±11.10	0.043±0.01	0.14±0.11	
S10A	293.17±48.33	130.86±19.24	160.49±23.59	0.79±0.01	1.9±0.23	
S1B	31.79±11.64	27.24±4.54	33.41±5.56	0.15±0.03	0.41±0.05	
S2B	24.22±13.37	11.72±8.09	14.37±9.93	0.07±0.02	0.18±0.10	
S3B	22.89±13.68	16.50±7.00	20.23±8.59	0.09±0.02	0.23±0.09	
S4B	24.76±13.25	20.42±6.10	25.05±7.48	0.11±0.03	0.32±0.07	
S5B	45.40±8.51	27.04±4.58	33.16±5.62	0.16±0.04	0.42 ± 0.05	
S6B	53.54±6.65	36.14±2.50	44.32±3.06	0.20±0.05	0.59 ± 0.01	
S7B	42.56±9.16	31.97±3.45	39.21±4.23	0.18±0.04	0.51±0.03	
S8B	55.42±6.21	35.45±2.66	43.47±3.25	0.20±0.05	0.56±0.01	
S9B	11.87±16.20	7.70±2.66	9.45±11.06	0.04±0.01	0.13±0.11	
S10B	232.70±34.46	103.60±12.98	127.06±15.91	0.63±0.14	1.67±0.24	
Average	82.50±16.00	47.02±7.90	57.66±6.69	0.27±0.06	0.63±0.09	
Max	293.17±48.33	153.76±24.50	188.57±30.03	0.85±0.16	2.41±0.41	
Min	11.35±1.05	7.55±0.06	9.26±0.07	0.043 ± 0.004	0.13±0.006	

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From the **Table2**, it shows that the radium equivalent (Ra_{eq}) is found in the range 11.35±1.05-293±17Bq/kg, an average value of 82.50±16) Bq/kg .The average value of radium equivalent is less than the safe limits 370Bq/kg^[13]. The mean value of external radiation hazard index is (0.85) which is less than 1 and confirm it is safe to carry out the activities for the human in that region . The outdoor air absorbed dose rate due terrestrial gamma rays at 1m above the ground were calculated for²²⁶Ra ,²³²Th and ⁴⁰K and the range is 7.55±0.06-153.76±24.50 nGY/ with an average 47.02±7.90)nGy/h which is lower than the world average of 60 nGy/h^{[1,16.13].}

The annual effective dose rate equivalent is calculated using a conversion factor of 0.7 Sv/Gy to convert the absorbed dose rate to to the effective dose equivalent and 0.2 for the outdoor occupancy factor. The annual effective dose rates are found in the range of $9.26\pm0.07-188.57\pm30.03$) µSv/y with an average 57.66 ± 6.69)µSv/y which is lower than the world average of 80μ Sv/y^[16].

The representative level index I_{yr} equation (7) must be less than 1.for the investigated samples this index is in average 0.63±0.09 Bq/kg ,but higher than unity in three samples S1A,S10A and S10B 2.41±0.41, 1.9±0.23 and 1.67±0.24 respectively.

Country	²³⁸ U	²³² Th	⁴⁰ K	References
Malizia	39±0.7	52±1	61±15	[8]
Egypt	79±2	44±1	586±18	[6]
Libya	7.5±2.5	6.7±1.9	4.5±1.3	[13]
Iran	74±4	69±4	1130±32	[9]
Turky	70±0.8	83±1	1234±7	[10]
Kenya	93.36±00	150±00	732.64±00	[7]
Nigeria	74.74±5.67	199.23±43.30	1021.27±7.14	[1]
This work	25.12±4.0	25.21±4.86	26.58±4.8	
World wide	33	45	412	[13]

Table.3 Average activity concentration in this work and others (Bq/kg)

4. Conclusions

In this study, the results indicate that the natural radioactivity concentration of ²²⁶Ra is lower than that of ²³²Th at the S10A. The values of mean absorbed dose rate, annual effective dose rate and the Representative level index activity were higher than worldwide average values in regions (S1A,S1B and S6A ,but representative level index of SA6 is also higher than unity); whereas the external hazard index less than unity in all samples.

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