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Determination of Radionuclide's Levels and Absorbed Dose for Soil, Rock, Plant and Water in Gondola – Libya

*Amany T. Sroor, *Nadia Walley EL-Dine, *Samia.M. EL-Bahi,**Hamad. M. Adress Hasan, ***Jemila Mussa Ali

*Faculty of Women for Arts, Science and Education. Physics Department, Ain Shams University**
***Chemistry department, College of science Omar AL-mokhtar University*
****physics department, College of science Omar AL-mokhtar University*
Corresponding Author: Amany T. Sroor

***Abstract**: The levels of natural radioactivity in some soil, rock, plant and water samples collected from different locations of Gandula-Libya were measured. The radioactivity levels of ^{226}Ra, ^{238}U, ^{232}Th and ^{40}K were measured by direct γ-ray spectrometry using HPGe detector. Based low background gamma-ray counting system with specially designed shield. The average radioactivity concentration in soil samples are 66.32, 59.52, 56.07 and 517.92 $Bq.kg^{-1}$ for ^{226}Ra, ^{238}U, ^{232}Th and ^{40}K, respectively. For rocks the average activity concentrations of same radionuclides are 26.01, 23.39, 25.13 and 100.97 $Bq.kg^{-1}$ respectively. In addition the average activity of ^{226}Ra, ^{238}U, ^{232}Th and ^{40}K for plants are 106.22, 95.49, 143.22 and 1255.82 $Bq.kg^{-1}$, respectively. The activity concentrations in water of previous radionuclides 19.24, 16.78, 19.32 and 102.78$Bq.L^{-1}$, respectively. The absorbed dose rate **D**, the annual outdoor effective dose $\boldsymbol{E_{out}}$ and excess lifetime cancer risk **ELCR** in this study are calculated. The results from this study have been compared with different countries of the world and the world average radioactivity in the same samples.*

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I. Introduction

Natural radionuclides of uranium ^{238}U, thorium ^{232}Th and potassium ^{40}K are present in the earth's crust. When these radionuclides and their daughters in the series undergo decays gamma rays, beta and alpha radiations are released to the environment **[1]**. Therefore, radionuclides can present very harmful biological effect to human cells and tissues as a result of continuous ingestion. Exposure to ionizing radiation causes damage to living tissue, and can result in mutation, cancer, and death **[2]**. Various research works have been carried out by scientists on the measurement of radionuclide concentration in both surface and underground waters of different areas of the world **[3]**. The knowledge of radiation levels in the environment is an important for assessing the effects of radiation exposure. So, the aim of the present study is to measure the natural radioactivity levels for estimating the radiological hazard indices in soils, rocks, plants and water in Gandula area. This work can be used as a baseline guideline for assessing the exposure of the natural radiation in the study region, especially, in this area, the rocks are used as building materials and the soils for agricultural purposes.

Geological Description of Study Area

Al-Gabale Al-akhdar is a high forested area in northeastern Libya is characterized by its high altitude from most of Libya and high rainfall rates and the availability of fertile land suitable for agriculture. Most of the rocks in this area are calcareous rocks. The population census in this area is considered large and divided over several cities, including the Gandula area. Its geographical coordinates are ($21^{o}\,34^{\backslash}\,25.176"$ E , $32^{o}\,32^{\backslash}\,28.0608"$ N). Gandula is a rural area, and the agriculture is the major economic activity with livestock farming contributing by good ratio of the country's gross domestic product, so it was important to focus on the environment of this region like : soil, rock, plant and water which was conducted by this study. Fig. (1) shows Gandula area.

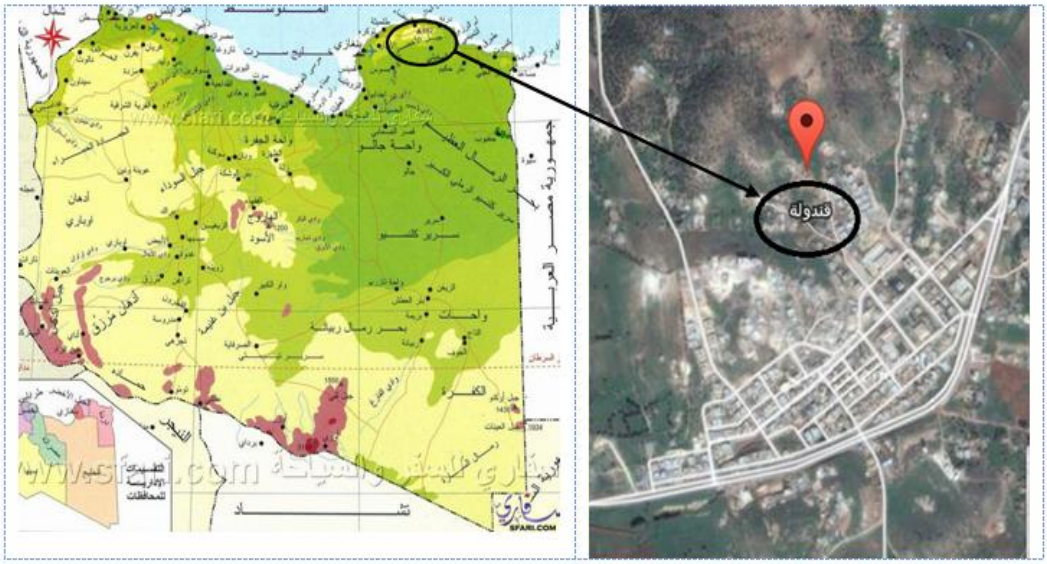

Fig. (1) : The geographical map of Libya and samples location.

Sampling Preparation

The samples collected from different location of Gandula–Libya. we selected a well for ground water used for drinking. Various samples (soil, rock, plant and water) were taken from around the well and in all directions (north, south, west and east) in Gandula. The distance between each point were a different set of samples and another 1km (the distance between the samples in each direction 1km, start from around well, then 1km, then 2km distance for all directions)**.** For soil 12 samples were collected from different locations and all directions (SN, SS, SW and SE), and for rock 12 samples were selected (RN, RS, RW and RE), also for plant samples were taken (PN, PS, PW and PE). The water samples were collected during 5 weeks (W1, W2, W3, W4 and W5) , which is 1 liter per week. The soil, rock and plant samples were oven dried at the temperature of $95C^{o}$ for 3 and 4 hours until the moisture was completely removed **[4]**. The samples were then ground into fine particles and thoroughly mixed and pass through a fine mesh sieve 200 mesh size to obtain composite representative samples **[4]**. Finally, the sample were placed in polyethylene bottles of 250 cm^3 volume and weighted **[4]**. which were well sealed using silicon and plastic tapes for air tight for about 30 days in order to allow secular equilibrium between ^{226}Ra and its short-lived decay products in the ^{238}U series and ^{232}Th. For gamma analysis, these samples were placed directly over the detector. The counting time for each sample was 70000 sec, except the water samples measured for a period time 140000 sec. The measured activity concentrations were presented as $Bq.kg^{-1}$.

Instrumentation

The activity concentrations of radionuclides in the samples were determined by a nondestructive analysis using a computerized gamma ray spectrometry system with coaxial high-purity germanium (HPGe) detector. (ORTEC572A) of sensitive volume of 76.11 cm^3. The energy resolution, expressed in terms of FWHM, was 1.9 keV at 1332.5 keV gamma ray ^{60}Co transitions. The gamma spectrometer was coupled to conventional electronics connected to a multichannel analyzer card (MCA) installed computer. A software program called MAESTRO-32 was used to accumulate and analyze the data and to calculate the natural radioactivity concentrations in the samples. To reduce the gamma ray background and noises for the hyper pure germanium. A cylindrical lead shield with a fixed bottom and movable cover were shielded the detector . The shield contained two inner concentric cylinders of copper and aluminum in order to absorb X-rays generated in the lead . The leakage current can be further reduced from its room temperature value by cryogenic cooling of solid-state medium, by used liquid nitrogen temperature (77 K^{o}) **[5]**. All gamma measurements were taken often calibrating the multi-channel analyzers (MCA) using ^{241}Am, ^{60}Co and ^{226}Ra sources. The absolute efficiency calibration of HPGe detector, using the KCl solution has been done through two stages the relative photo peak efficiency curve of the detector was obtained using ^{226}Ra point source (unknown activity). In the second stage, the relative efficiency curve was normalized to an absorbed volume efficiency curve using KCl solution **[6, 7]**. The activity concentration of ^{226}Ra determined from photo peak 186.2 keV. The activity concentration of ^{238}U series determined ^{214} Pb photo peak (295.2 and 351.9) keV and ^{214}Bi (609.3, 768.4, 1120.3, 1238.1, 1377.7 and 1764.5) keV. similarly for ^{232}Th series, the activity concentration determined from ^{228}Ac (92, 209.5, 338.5, 911.1 and 968.9) keV and ^{208} Tl at 583 keV, ^{212}Bi at 727.2 keV. ^{40}K was determined

from photo peak 1460 keV **[8]**. The concentration of ^{226}Ra was determined by measuring is 186 keV photopeak and 352 and 609 keV photopeaks of ^{214}Pb and ^{214}Bi. It was clear that the concentration of radium at the 186 keV photopeak was very high because of the contribution of ^{238}U which emits gamma rays from the same energy. The photopeaks of 352 and 609 keV gave an apparent radionuclide concentration that was very low due to the emission of radon (^{214}Pb and ^{214} Bi are radon product). The results of these phenomena were corrected. For the 186 keV phtopeak, it was assumed that 58% of the gamma ray activity was due to ^{226}Ra and its concentrations were divided by 1.72. For ^{214}Pb and ^{214}Bi, it was equal 8.6 % and concentrations calculated from these photopeaks were multiplies by 1.094. The results obtained from the 186 keV photopeak and the 352 and 609 keV photopeaks were generally in go mutual agreement **[9].**

Activity Concentration

The activity concentrations (A) of the radionuclides in the samples determined using the following equation **[10].**

$$\mathbf{A} = \frac{\mathbf{N}}{\boldsymbol{\varepsilon}\, \mathbf{I_\gamma t\, m}} \qquad \textbf{(1)}$$

Where , A is the activity concentration (Bq.kg^{-1}) in the sample, N is the corrected net photo-peak area at energy peak ($N=N_S-N_B$), $\mathbf{N_s}$ is the net photo peak area in the sample and $\mathbf{N_B}$ is the corresponding net photo peak area in the background spectrum), $\boldsymbol{\varepsilon}$ is the absolute efficiency at photopeak energy. $\mathbf{I_\gamma}$ is the gamma-ray emission probability corresponding to the peak energy, m is the mass **(kg)** of the measured sample, **t** : is the time of the sample spectrum collection in seconds **[10].**

Absorbed Dose

The absorbed dose in the samples **(nGy.h^{-1})** are calculated. The gamma absorbed dose rate **(D)** in air at 1 m above the ground level is for natural radionuclide ^{226}Ra,^{232}Th, and ^{40}K are estimated by using the relation:

$$\mathbf{D = 0.427\, A_U + 0.662\, A_{Th} + 0.043\, A_K \ (nGy\ .h^{-1})} \qquad \textbf{(6)}$$

Where : 0.462, 0.604 and 0.0417 nGy.h^{-1} per Bq.kg^{-1} are the conversion factors, **D**: is the dose rate in **nGy.h^{-1}**, $\mathbf{A_{Ra}}$, $\mathbf{A_{Th}}$ and $\mathbf{A_K}$ the activities in **(Bq.kg^{-1})** of ^{226}Ra, ^{232}Th and ^{40}K, are respectively **[11]**.

Annual Effective Dose Equivalent

The annual effective dose equivalent **(E_{out})** is calculated from the absorbed dose rate (D), by multiplying of conversion factor 0.7 Sv.Gy^{-1} and the occupancy factor of 0.2 (20 % of 8760 h in a year) to convert the absorbed dose in air to effective dose. During the present study, the E_{out} was calculated using the following equations **[12]**:

$$\mathbf{E_{out}\ (nSv.y^{-1}) = D\ n\ (Gyh^{-1}) \times 24\ h \times 365.24\ d \times 0.2 \times 0.7\times 10^{-3}\ \ Sv.Gy^{-1}} \qquad \textbf{(7)}$$

Excess lifetime cancer risk

The excess lifetime cancer risk **(ELCR)** was estimated by using the annual effective dose and the lifetime of continuous exposure of population as following equation.

$$\mathbf{ELCR = E_{out} \times D_L \times R_F} \qquad \textbf{(8)}$$

Where : $\mathbf{D_L}$ is the duration of lifetime (approximately 66 years) and $\mathbf{D_F}$ is the risk factor (Sv^{-1}) , which reflects the fatal cancer risk per Sievert **[13]**. For stochastic effects, ICRP 60 uses values of 0.05 for the public **[14]**.

II. Result and Desiccation

Activity Concentrations

The result of activity concentrations of ^{226}Ra, ^{238} U, ^{232}Th and ^{40}K in all samples investigated were presented in Table (1). The activity concentrations of ^{226}Ra, ^{238} U, ^{232}Th and ^{40}K in soil samples were ranged between (43.14-104.08, 36.44-93.39, 36.75-69.35 and 64.59-661.28) Bq.kg^{-1} for ^{226}Ra, ^{238} U, ^{232}Th and ^{40}K respectively, with average (66.32, 59.52, 56.07 and 517.92) Bq.kg^{-1}, respectively. For rock samples the activity concentrations of ^{226}Ra, ^{238} U, ^{322}Th and ^{40}K were ranged (21.44-33.94, 17.34-30.91, 19.14-41.18 and 47.48-170.46) Bq.kg^{-1}, respectively, with average activity concentrations are (26.01, 23.39, 25.13 and 100.97) Bq.kg^{-1} respectively. We found the activities of previous radionuclides in plant samples in ranged (56.48-191.45, 51.34-160.24, 98.31-275.12 and 610.21-3133.42) Bq.kg^{-1}. The average values in plant samples are (106.22, 95.49, 143.22 and 1255.82) Bq.kg^{-1}, respectively. The activity concentrations in water samples for the above radionuclides were presented (15.50-23.63, 14.65-22.63, 15.37-22.43 and 84.95-116.20) Bq.kg^{-1}, respectively, with average are (19.24, 16.78, 19.32 and 102.78) Bq.kg^{-1}, respectively. The average activity concentrations of ^{226}Ra for soil, plant and water samples are higher than the world's population with average of 32, 50 and 0.5 Bq.kg^{-1}, respectively **[16, 17]**. Also for ^{238}U the average activity concentrations for all samples higher than permissible level 33, 20 and 1 Bq.kg^{-1} **[15, 16**].

Table (1) : The activity concentrations (Bq.kg-1 and Bq.L-1) of the radionuclides 226Ra,238U, 232Th and 40K of the investigated sample.

Type of Samples	Samples	^{226}Ra	^{238}U	^{232}Th	^{40}K
Soil	SN0	70.00	65.20	65.09	572.86
	SS0	75.48	67.21	61.41	615.22
	SW0	63.64	58.86	60.60	600.01
	SE0	68.14	65.31	59.65	64.59*
	SN1	88.75	76.39	65.98	654.74
	SS1	53.14	45.26	51.02	484.76
	SW1	56.56	51.57	49.42	585.124
	SE1	55.94	53.45	58.44	486.28
	SN2	104.08*	93.39**	69.35**	645.06
	SS2	73.20	60.72	57.81	661.28**
	SW2	43.14*	36.44*	37.33	464.62
	SE2	43.79	40.43	36.75*	380.53
	Average	66.32	59.52	56.07	517.92
	P.L	32	33	45	412
Rock	RN0	31.43	27.00	21.39	87.99
	RS0	27.01	24.42	19.89	95.88
	RW0	24.92	21.18	21.2	89.12
	RE0	22.49	21.12	29.94	106.04
	RN1	32.54**	30.91**	23.18	170.46**
	RS1	27.63	24.26	26.93	47.48*
	RW1	21.51	20.63	29.83	113.50
	RE1	24.75	22.63	21.49	100.22
	RN2	33.94	29.77	23.46	118.024
	RS2	22.22	17.34*	19.14*	89.58
	RW2	21.444*	18.01	41.18**	116.38
	RE2	22.187	20.35	23.77	77.00
	Average	26.01	23.39	25.13	100.97
	P.L	50	33	50	500
Plant	PN0	169.01	154.08	176.11	1354.92
	PS0	118.19	101.24	165.85	794.16
	PW0	99.08	91.53	116.20	1057.88
	PE0	103.79	82.42	88.39	884.17
	PN1	56.48	51.34*	98.31*	610.21*
	PS1	69.79	68.13	114.52	782.85
	PW1	115.35	109.06	99.20	1204.09
	PE1	89.73	87.77	111.36	1231.04
	PN2	191.45*	160.24**	138.85	1850.38
	PS2	61.021*	60.633	117.55	956.68
	PW2	81.43	78.15	217.18	1210.02
	PE2	119.319	101.177	275.121**	3133.416**
	Average	106.22	95.49	143.22	1255.82
	P.L	50	20	15	420
Water	W1	18.71	16.06	19.80	90.80
	W2	19.14	16.26	21.96	111.56
	W3	19.23	14.90	22.43**	116.20**
	W4	23.63**	22.63**	17.05	110.38
	W5	15.50*	14.05*	15.37*	84.95*
	Average	19.24	16.78	19.32	102.78
	P.L	0.5	1	0.1	10
* Lowest value		** highest value		P.L : Permissible	

The average activity concentrations of radionuclides (^{232}Th and ^{40}K) in soil, plant and water samples are higher than the permissible level (45, 15 Bq.kg^{-1} and 0.1 Bq.L^{-1} for ^{232}Th) and (412, 420 Bq.kg^{-1} and 10 Bq.L^{-1} for ^{40}K), but in rock samples the average activity concentrations of same radionuclides are lower than the permissible level (50 for ^{226}Ra, 33 for ^{238}U, 50 for ^{232}Th and 500 for ^{40}K) Bq.kg^{-1}, respectively **[15, 16]**. As shown in Figs. (2), (3), (4), (5) and (6)**.**

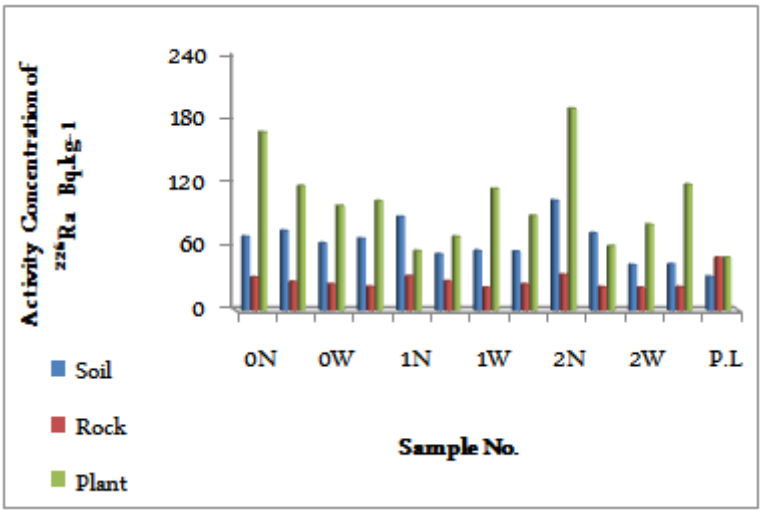

Fig. (2) : Activity concentrations of ^{226}Ra for soil, rock and plant samples.

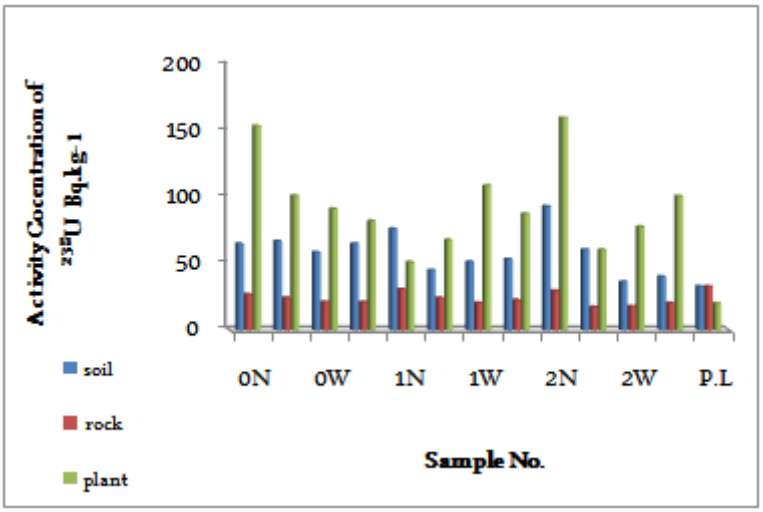

Fig. (3) : Activity concentrations of ^{238}U soil, rock and plant samples

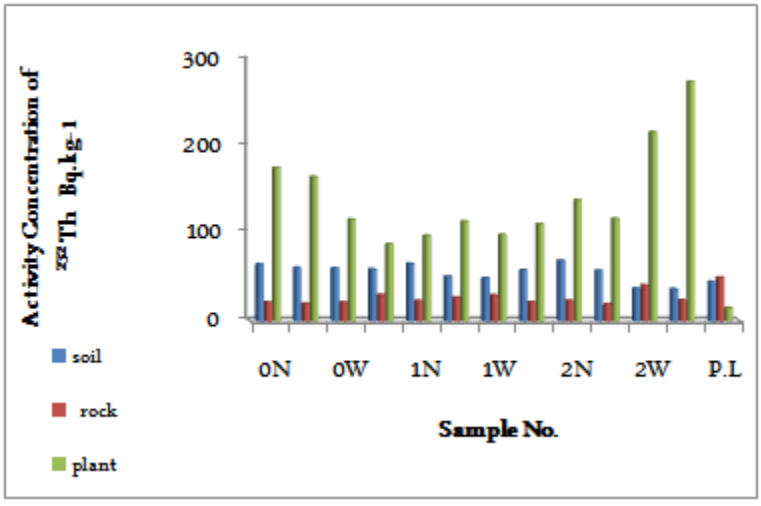

Fig. (4) : Activity concentrations of ^{232}Th for soil, rock and plant samples.

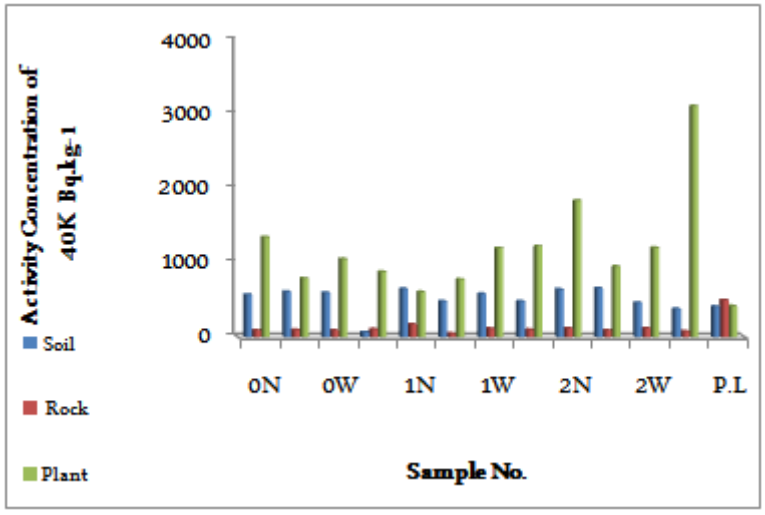

Fig. (5) : Activity concentrations of ^{40}K for soil, rock and plant samples.

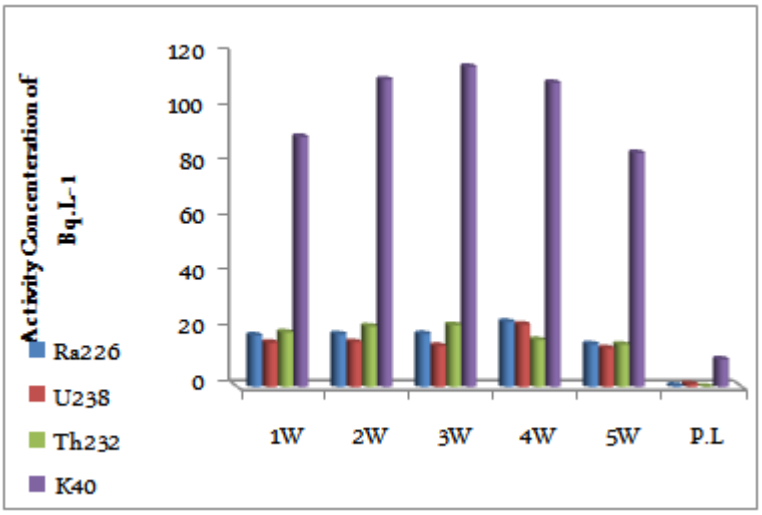

Fig. (6) : Activity concentrations of ^{226}Ra, ^{238}U, ^{232}Th and ^{40}K water samples.

This study is compared with the results from different countries of the world and the recommended values as given in Table(2). Table (2) shows that the values of average activity concentrations obtained in this study of ^{226}Ra are comparable with those obtained in Libya which lower than the others, while that average value obtained for ^{238}U and ^{232}Th in all other countries are lower than the value obtained in study. The average presents value of ^{40}K activity concentrations is higher than the obtained values in all countries. As shown in this Table, the radioactivity in all studied samples varied from one country to another, The variations in the activity concentrations in the soil of the various locations of the world, depend on the geological and geographical conditions of the area and the extent of fertilizer applied to the agricultural lands. In general, the radioactivity in samples varied from one country to another.

Table (2) : Comparison of natural radioactivity concentrations (Bq.kg^{-1} and Bq.L^{-1}) in all samples in present study and different countries of the world.

Type of samples	Country [Ref]	Average activity concentrations			
		^{226}R	^{238}U	^{232}Th	^{40}K
Soil	Present study (Bq.kg^{-1})	66.32	59.52	56.07	517.92
	Japan[15]	33	29	28	310
	Poland[15]	26	26	21	410
	World average	32	33	45	412
Rock	Present study (Bq.kg^{-1})	26.01	23.395	25.13	100.97
	Egypt[17]	410	-	7.9	37.6
	Japan[18]	35.8		20.7	139.4

	World average	**50**	**33**	**50**	**500**
Plant	**Present study ($Bq.kg^{-1}$)**	**106.22**	**95.49**	**143.22**	**1255.82**
	Ghana[19]	**-**	**31.8**	**56.2**	**839.8**
	Italy[20]	**23.2**	**0.4**	**-**	**654.7**
	World average	**50**	**20**	**15**	**420**
Water	**Present study ($Bq.L^{-1}$)**	**19.24**	**16.78**	**19.32**	**102.78**
	Nigeria[21]	**0.48**	**48.2**	**0.04**	**0.36**
	Saudi Arabia[22]	**2.06**	**0.03**	**-**	**73.31**
	World average	**0.5**	**1**	**0.05**	**10**

Radiation hazards

In this study area the radiological hazard index parameters in comparison with the world average are presented in Table (3). The average values of absorbed dose rate **D** of most soil and all plant samples are higher than the international values, except some rock and water samples are lower than the international values 59 $nGy.h^{-1}$ **[16]**. The annual outdoor effective dose $\mathbf{E_{out}}$ of most soil and all plant samples in studied area are higher than the international average of normal background dose received from radionuclides of terrestrial origin, and also the values of rock and water samples are lower than the permissible limit 0.07 $mSv.y^{-1}$ **[16].** The values cancer risk factor **ELCR** for most soil and all plant samples are higher than the international values. But the values of rock and water samples are lower than the international values 0.29×10^{-3} **[16]**, as shown in Figs.(6), (7), (8), (9) (10) and (11) .

Table (3) : The value of absorbed dose rate, annual outdoor effective dose and cancer risk factor for all samples.

Type of Samples	Samples	Dose rate D(out) ($nGy.h^{-1}$)	Annual effective dose E(out) ($mSv.y^{-1}$)	Cancer risk factor ELCR(out)$x10^{-3}$
Soil	**SN0**	**95.56**	**0.12**	**0.39**
	SS0	**95.81**	**0.12**	**0.39**
	SW0	**91.05**	**0.11**	**0.37**
	SE0	**70.16**	**0.09**	**0.28**
	SN1	**104.45**	**0.13**	**0.42**
	SS1	**73.94**	**0.09**	**0.30**
	SW1	**79.89**	**0.10**	**0.32**
	SE1	**82.42**	**0.10**	**0.33**
	SN2	**113.52****	**0.14****	**0.46****
	SS2	**92.63**	**0.11**	**0.37**
	SW2	**60.25**	**0.07**	**0.24**
	SE2	**57.96***	**0.07***	**0.23***
Rock	**RN0**	**29.47**	**0.04**	**0.12**
	RS0	**27.72**	**0.03**	**0.11**
	RW0	**26.99**	**0.03**	**0.11**
	RE0	**33.39**	**0.04**	**0.13**
	RN1	**35.82**	**0.04**	**0.14**
	RS1	**30.23**	**0.04**	**0.12**
	RW1	**33.44**	**0.04**	**0.13**
	RE1	**28.20**	**0.03**	**0.11**
	RN2	**33.32**	**0.04**	**0.13**
	RS2	**23.93**	**0.03***	**0.10**
	RW2	**39.96****	**0.05****	**0.16****
	RE2	**27.73***	**0.034**	**0.112***
Plant	**PN0**	**240.64**	**0.29**	**0.97**
	PS0	**187.23**	**0.23**	**0.76**
	PW0	**161.50**	**0.20**	**0.65**
	PE0	**131.73**	**0.16**	**0.53**
	PN1	**113.24***	**0.14***	**0.46***
	PS1	**138.17**	**0.17**	**0.56**
	PW1	**164.13**	**0.20**	**0.66**
	PE1	**164.13**	**0.20**	**0.66**
	PN2	**239.91**	**0.29**	**0.97**
	PS2	**144.85**	**0.18**	**0.59**

	PW2	229.17	0.28	0.93
	PE2	360.07**	0.44**	1.46**
Water	W1	23.87	0.03	0.10
	W2	26.28**	0.03**	0.11**
	W3	26.21	0.032**	0.11**
	W4	25.70	0.031	0.10
	W5	19.82*	0.02*	0.08*
	P.L	59	0.07	0.29
* Lowest value		** highest value		P.L : Permissible Level

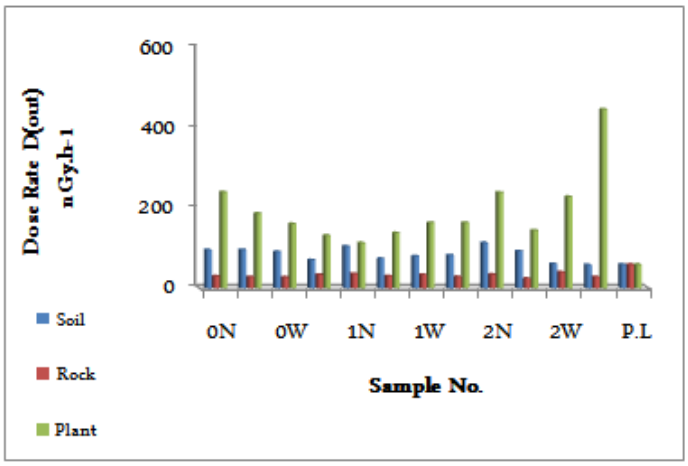

Fig. (5) : The absorbed dose of soil, rock and plant samples.

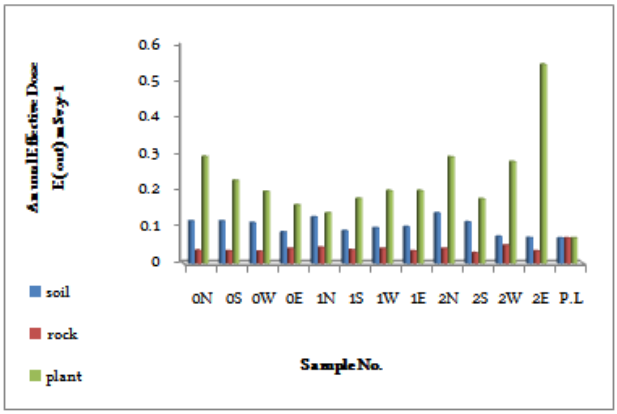

Fig. (7) : The annual effective dose of soil, rock and plant sampl

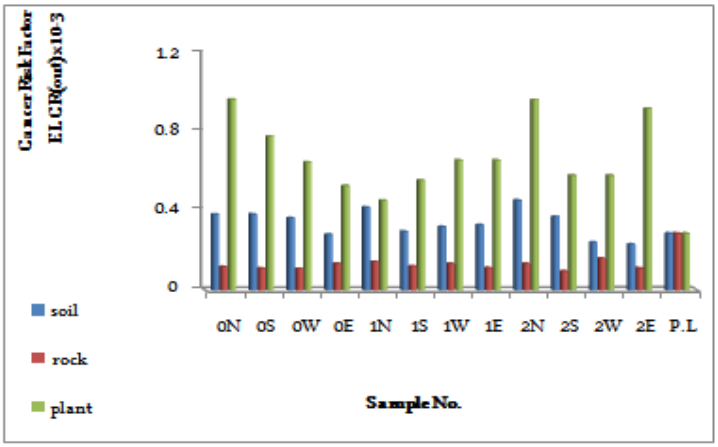

Fig. (8) : The cancer risk factor of soil, rock and plant samples.

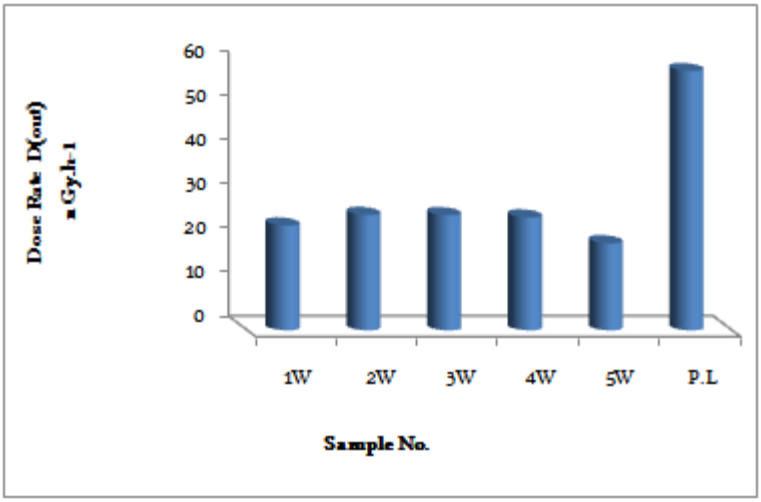

Fig. (9) : The absorbed dose of water samples.

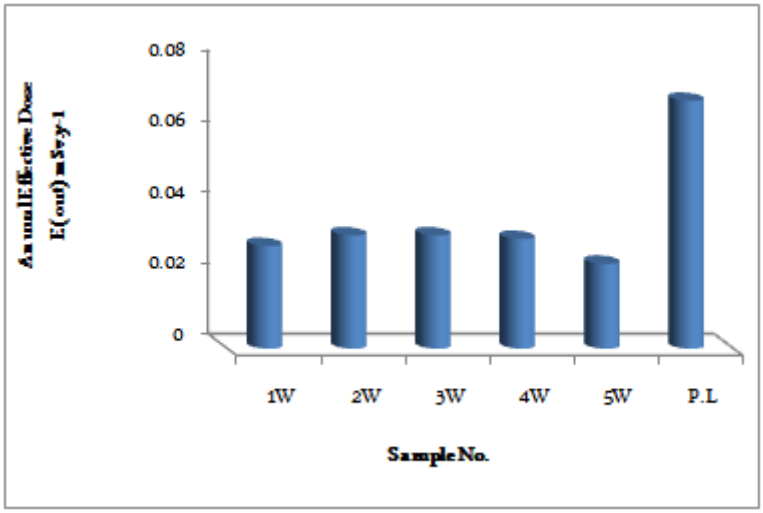

Fig. (10) : The annual effective dose of water samples.

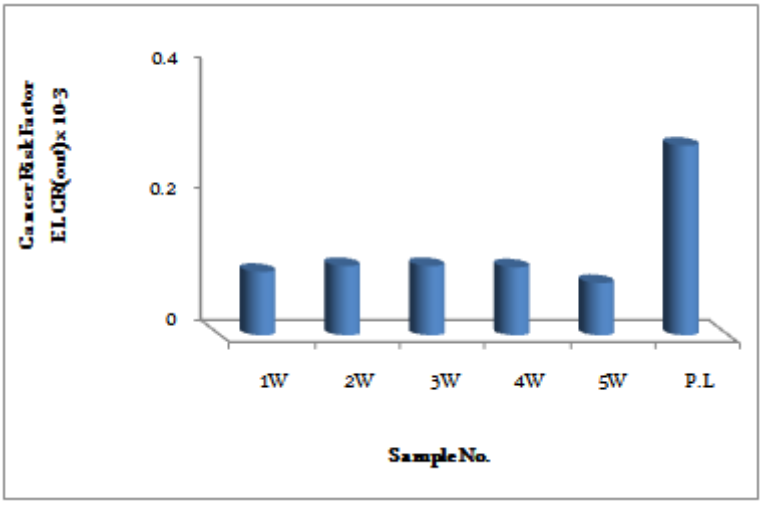

Fig. (11) : The cancer risk factor of water samples.

III. Conclusion

The natural radionuclides have been measured by using HPGe detector with a specially designed shield. This study appeared that the soil samples of the three locations contain high activity concentrations of ^{226}Ra and ^{238}U, and its progenies, also ^{232}Th and ^{40}K occur in high activity. So natural radioactivity are higher than the world's average as UNSCEAR (2010). The activity concentrations of ^{226}Ra, ^{238}U, ^{232}Th and ^{40}K in all samples of plant are higher than the world's average. Also natural radioactivity of all samples of water are higher than permissible level. The natural radioactivity in all rock samples are lower than the world's average.
The absorbed dose rate, the annual effective dose in outdoor environment and the excess lifetime concern risk of soil and plant samples were higher than the world's average which represent radiological risk for people life in these location to protect against higher radioactivity.

Finally this study can be used as a baseline for facture investigations and the data obtained in the study may be useful for natural radioactivity mapping.

This indicated that the study area was radiologically not safe relatively for human being. Recommend the work of study on the soil must be made in different place. Also study must be made on the natural radioactivity on the around waters to be healthy on human.

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