



ISSN 1110-0451

Arab Journal of Nuclear Sciences and Applications

Web site: ajnsa.journals.ekb.eg



(E S N S A)

Radiological Impacts of Petroleum Exploration Activities in Ras Qattara Area, North Western Desert, Egypt

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ARTICLE INFO

Article history:

Received: 28th Oct. 2021

Accepted: 5th Feb. 2022

Keywords:

Radioactivity;

Radiological;

NORM;

Spectrometer;

Hazard indices.

ABSTRACT

The activity concentrations and the associated with hazard indices of primordial radioactive nuclides ^{238}U , ^{232}Th and ^{40}K in soil samples of different depths from petroleum well in Ras Qattara area north western desert, Egypt, were estimated using gamma-ray spectroscopy system using a closed end-coaxial Canberra N-type HPGe detector of vertical configuration, with 40% relative efficiency and 2.0 KeV energy resolution at 1.33 MeV photons of ^{60}Co . The average soil activities for ^{238}U , ^{232}Th , and ^{40}K were 32.07 ± 2.75 , 14.04 ± 3.35 and 297.44 ± 14.05 Bq/kg, respectively. The average values of radium equivalent (R_{eq}), absorbed dose rate (D_{out}), annual effective dose equivalent (AEDE), annual gonadal dose equivalent (AGDE) and excess lifetime cancer risk (ELCR) were 75.04 Bq/Kg, 35.70 nGyh⁻¹, 0.04 mSvyr⁻¹, 251.16 μSvyr^{-1} and 0.15×10^{-3} respectively. Some other radiological hazard indices such as external hazard index (H_{ex}), internal hazard index (H_{in}), and representative level index (I_r), were estimated and compared with the world average values. The average values for all radiological hazard indices in the soil sample of the present study were lower than the limit average values recommended by UNSCEAR 2000.

INTRODUCTION

Naturally occurring radioactive materials NORM are characterized by wide distribution and having radiological effects more than those produced in nuclear industries [1]. NORM is responsible for approximately 80% of the total radiation exposure of the world's population and 57% of this exposure comes from ^{226}Ra a member of ^{238}U decay series [2]. NORM is much related to the geological and geographical conditions and appears at different levels in soil of different regions of the world [3]. Naturally occurring radioactive materials may be enhanced above NORM levels due to a number of human activities such as petroleum explorations, mining, fertilizer production, iron and steel production, extraction of coal, building materials industry and (uranium and thorium) mining etc [4]. Petroleum explorations cause NORM to be accumulated and elevated up NORM levels during drilling and processing of petroleum [5, 6, 7]. The uncontrolled release of radioactive material due to petroleum exploration may

pollute the environment represents potential environmental and health risks to the worker if not properly handled [4]. Hazardous radiations may enter the human body by internal or external exposure as inhalation and ingestion. The more radiation dose a person gets, the more chance of developing cancer, leukaemia, eye cataracts, hematological depression and etc occurs [8 and 9].

In present study, soil samples of different depths from petroleum well in Rase El-Qattara area, north western desert, Egypt, were collected in order to estimate the activity concentrations of primordial radionuclides ^{238}U , ^{232}Th and ^{40}K and radiological hazard indices due to petroleum exploration activity.

Geographical and Geological aspects

Qattara Depression is considered the largest natural closed depression of the north western desert, Egypt, which covers a surface area of about 19,500 Km² [10, 11] with triangular shape headed in Assuit Barrage and a base end at the Mediterranean Sea. From geological

and geomorphologic view, Qattara Depression can be typed as a polygenetic depression returned back to the Middle and lower Miocene age [12, 13]. Petroleum source rocks which contain the precursors of hydrocarbon formation and organimatter in the northern Qattara Depression occurred in various Jurassic and Cretaceous deposits and in Paleozoic and Tertiary rocks where sandstone and carbonate reservoirs rocks of Jurassic and Cretaceous age have retained the immigration oil from the source rocks. Various shales, compact limestone and dolomite beds of Jurassic, Cretaceous, Eocene, and Oligocene age have made efficient seals which restrict hydrocarbons to escape from reservoir and Structural, Local fault-block structures and Stratigraphic traps of Cretaceous to Early Tertiary have ensured a fixed and firm position of seal and reservoir [14, 15].

Sampling and preparation

Soil samples were collected from different depth from petroleum well in Ras Qattara, North Western Desert, Egypt. The samples were packaged in polyethylene bags, systematically labelled. The samples were homogenized and kept under normal conditions in the laboratory environment. The collected samples were pulverized to a fine dust then, sieved using a 1 mm mesh size to remove the large size and homogenized. Then, the samples were dried to completely remove the moisture. All samples were kept tightly closed and stored for 30 days to form a radioactive equilibrium [16, 17, 18].

Gamma spectrometric analysis

Radiometric analysis of all the samples was done at the Radiation Protection Department in the Nuclear and Radiological Regulatory Authority, Egypt. The activity concentrations assessment of primordial radionuclides ^{238}U , ^{232}Th and ^{40}K in the samples was done using a closed end-coaxial Canberra N-type HPGe detector of vertical configuration, with 40% relative efficiency and 2.0 KeV energy resolution at 1.33 MeV photons of ^{60}Co was used. The detector is shielded by a detector lead shield with Outer Jacket: 9.5 mm thick low carbon steel, Bulk Shield: 10 cm thick low background and Graded Lining: 1 mm tin and 1.6 mm copper. The spectra were analyzed using CANBERRA (Genie 2000) program. The detector was calibrated using gamma mixed sources with different radionuclides. The background was estimated using the same standard measurement conditions. The background levels in the laboratory were subtracted from the net count for all measured samples. A validation of the accuracy of the calibration by using Lab SOCS software for mathematical efficiency calibration of Ge

detectors for laboratory samples with the same geometry shape was verified. Quality Assurance was carried out by analysis of IAEA-381 and SOIL 6 reference materials with a known concentration of natural radioactivity.

Sample analysis and activity concentration

By considering the secular equilibrium, ^{238}U and ^{232}Th activity concentrations were estimated from their daughters where ^{238}U was estimated from gamma-ray transitions of ^{214}Bi (609.3, 1120.3 and 1764 KeV) and ^{214}Pb (351 KeV). ^{208}Tl (583.19 and 2614.53 KeV), ^{212}Pb (238.63 and 300.09 KeV), ^{228}Ac (911 KeV) and ^{212}Bi (727.3 KeV) peaks were used for estimating the activity concentrations of ^{232}Th . The activity concentrations of ^{40}K were estimated from 1460.83 KeV (10.7%) peak [16, 17, 19 and 20]. The activity concentrations of ^{238}U , ^{232}Th and ^{40}K in samples were calculated by using the following equation

$$A_S = \frac{N_C}{M} \times \frac{1}{\tau} \times \frac{1}{\eta} (Bq/Kg). \quad (1)$$

Where N_C is the net counting of γ -ray (counts per second) corrected for background, τ is the detector efficiency of the specific γ -ray, η is the absolute transition probability of γ -decay and M is the mass of the sample in Kg. The activity concentrations of ^{238}U and ^{232}Th were expressed in terms of ppm while ^{40}K was expressed in % as the specific activity of a sample containing 1 ppm by weight of ^{238}U is 12.35 Bq/Kg and of ^{232}Th is 4.06 Bq/Kg and 1% of weight of ^{40}K is 313 Bq/Kg [16, 21, 22].

Radiation hazard indices

Radium equivalent (Ra_{eq})

To compare the activity levels of samples containing different concentrations of ^{238}U , ^{232}Th and ^{40}K , the radioactivity has been defined in terms of radium equivalent (Ra_{eq}) in Bq/Kg which is calculated as the follows [16, 23, 24].

$$Ra_{eq} (Bq/Kg) = A_U + 1.43 A_{Th} + 0.077 A_K \quad (2)$$

Where A_U , A_{Th} and A_K are the activity concentrations of ^{238}U , ^{232}Th and ^{40}K in Bq/Kg respectively.

The absorbed dose rates (D_{out})

Radiological hazard risk from external exposure to radiation due to gamma radiations at 1m above the ground surface for the uniform distribution of the primordial radionuclides ^{238}U , ^{232}Th and ^{40}K was calculated from the following equations [25 and 26].

$$D_{out} (nGyh^{-1}) = 0.462A_U + 0.621A_{Th} + 0.0417A_K \quad (3)$$

Where 0.462, 0.621 and 0.0417 are conversions factors in $nGyh^{-1}$ per unit activity concentration in Bq/Kg of dry weight for ^{238}U , ^{232}Th and ^{40}K respectively.

Gamma radiation representative level index (I_γ)

Gamma radiation hazard representative level I_γ associated with natural radionuclides in the soil was calculated by the following equation [23, 24]

$$I_\gamma = A_U / 150 + A_{Th} / 100 + A_K / 1500 \quad (4)$$

Where the safety value of this index is ≤ 1 .

The annual effective dose equivalent (AEDE)

Conversion coefficient of $0.7 SvGy^{-1}$, outdoor occupancy of 0.2 and total time in hour per year of 8760 are used to estimate the annual effective dose equivalent received outdoor ($mSvyr^{-1}$) as follows [16].

$$AEDE (mSvyr^{-1}) = D (nGyh^{-1}) \times 8760 h \times 0.7 \times (10^3 mSv/10^9 nGy) \times 0.2,$$

$$AEDE (mSvyr^{-1}) = D (nGyh^{-1}) \times 1.21 \times 10^{-3} (mSvyr^{-1}) \quad (5)$$

External and Internal hazard Indices (H_{ex} & H_{in})

The external and internal hazard indices are calculated as follows [25].

$$H_{ex} = A_U / 370 + A_{Th} / 259 + A_K / 4810 \quad (6)$$

$$H_{in} = A_U / 185 + A_{Th} / 259 + A_K / 4810 \quad (7)$$

The values of H_{ex} and H_{in} must be less than unity to keep the radiation hazard insignificant.

Excess lifetime cancer risk (ELCR)

The expected number of cancer cases or diagnosed cases with cancer in a given number of people due to exposure to radiation over a lifetime at a given exposure level, considering average age of 70 years, was calculated from the following [16]

$$ELCR = AEDE (mSvyr^{-1}) \times DL \times RF \quad (8)$$

Where $AEDE$ is the outdoors annual effective dose, DL is the average lifetime of 70 years and Rf is the risk factor (Sv) assigned by ICRP as 0.05/ Sv for the public [27 and 28].

Annual gonadal equivalent dose (AGED)

Measuring the possible threat resulting from the specific activities of ^{238}U , ^{232}Th and ^{40}K on the bone

marrow activities and bone surface cells called annual gonadal equivalent dose is calculated as follows [16].

$$AGED (mSvyr^{-1}) = 3.09A_U + 4.18A_{Th} + 0.314 A_K \quad (9)$$

RESULTS AND DISCUSSION

The activity concentrations of the primordial radionuclides ^{238}U , ^{232}Th and ^{40}K in the samples were calculated by equation 1. The measured activity concentrations and the uncertainty of ^{238}U , ^{232}Th and ^{40}K in Bq/Kg for the samples were listed in Table 1 and fig. 1. From results, the activity concentration for ^{238}U , ^{232}Th and ^{40}K radionuclides were with the ranges of 11.54 ± 1.90 to $56.19 \pm 3.95 Bq/kg$, 3.99 ± 2.01 to 42.13 ± 1.80 and 88.68 ± 6.92 to $762.49 \pm 14.14 Bq/kg$ respectively. The corresponding average values of the measured activity concentrations were 32.07 ± 2.75 , 14.04 ± 3.35 , and $297.44 \pm 14.05 Bq/kg$ for ^{238}U , ^{232}Th and ^{40}K respectively. The values in ppm were varied from 0.93 to 4.55 ppm with an average value of 2.60 ppm and 0.98 to 10.38 ppm with average value of 3.46 ppm for ^{238}U and ^{232}Th respectively. The percentage values of ^{40}K were varied from 0.28% to 2.44% with average value of 0.95%. From results, the ^{40}K concentrations in all samples are higher than that of ^{238}U and ^{232}Th and the concentration of radionuclide activities had the following order $^{40}K > ^{238}U > ^{232}Th$.

Upon the recorded values, the average activity concentration of ^{238}U is within the world average value of 35 Bq/Kg for ^{238}U and is lower compared to the world average values of 30 Bq/Kg and 400 Bq/Kg for ^{232}Th and ^{40}K respectively [16]. The overall results showed that the activity concentrations for ^{238}U , ^{232}Th and ^{40}K increased by increasing the depth as it reaches its maximum values of 56.19 ± 3.95 , 42.13 ± 1.80 and $762.49 \pm 14.14 Bq/Kg$ at depths 10340, 10520 and 10160 ft for ^{238}U , ^{232}Th and ^{40}K respectively. The difference in values may because of the difference in radionuclides concentration of ^{238}U , ^{232}Th and ^{40}K and their daughters in soil and rocks depending on the difference of geological structures and geophysical conditions.

From results listed in Table 1 and fig. 2, Ra_{eq} in Bq/Kg was varied from 28.20 to 151.65 Bq/Kg for soil samples in all depths with an average value of 75.04 Bq/Kg which is lower than the accepted safety limit value of 370 Bq/Kg given by UNSCEAR 2000 [16, 3].

Table 2 and fig. 3 showed that the D_{out} in $nGyh^{-1}$ for the samples was varied from 13.45 to 70.82 $nGyh^{-1}$ with an average value of 35.70 $nGyh^{-1}$ which is lower than the world limit value of 59 $nGyh^{-1}$ given by UNSCEAR 2000 [16, 3].

From table 2 and fig. 4, The estimated $AEDE$ in $mSvy^{-1}$ by depth difference depending on the value of the D_{out} in $nGyh^{-1}$ was between 0.02 and 0.09 $mSvy^{-1}$ with an average value of 0.04 $mSvy^{-1}$ while the worldwide average value given by UNSCEAR 2000 is 0.7 $mSvy^{-1}$ [3].

Table 2 and fig. 5 showed that, the calculated values of H_{ex} were varied from 0.10 to 0.41, with an average value of 0.21 while, the internal exposure by radon and its progeny expressed by H_{in} was varied from 0.11 to 0.55 with an average value of 0.29. The calculated values of H_{ex} and H_{in} for all samples did not exceed the maximum limit, which is 1 [16].

The calculated values of I_y in Bq/Kg of all samples copied in Table 2 and fig. 6, The values were varied from 0.21 to 1.11 Bq/Kg with an average of 0.55 Bq/Kg which is below the permissible limit of 1.00 Bq/Kg [3].

The values of $AGDE$ in $\mu Svyr^{-1}$ copied in table 2 and fig. 7 showed that, the values were varied from 94.87 to 497.57 $\mu Svyr^{-1}$ with average value of 251.16 $\mu Svyr^{-1}$ which is less than the average world limit of 300 $\mu Svyr^{-1}$ [29].

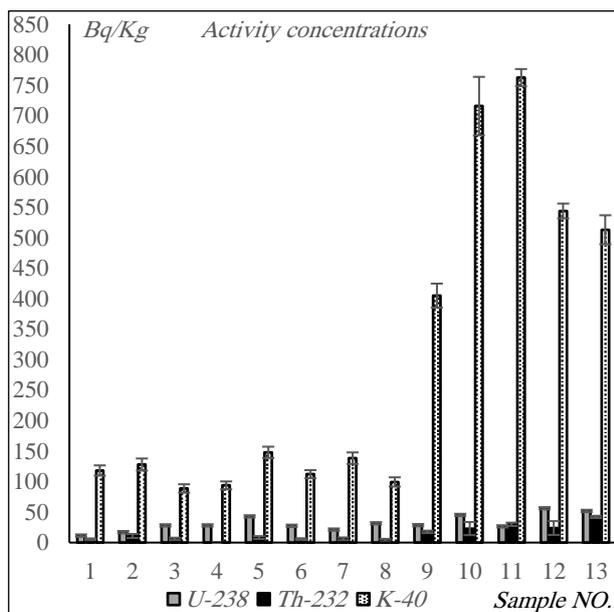
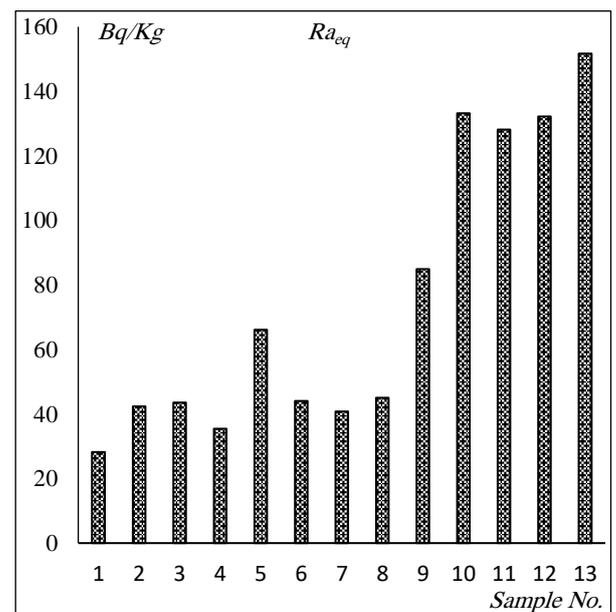
Table 3 and fig. 8 showed that, the values of $ELCR$ were varied from 0.06×10^{-3} to 0.30×10^{-3} with an average value of 0.15×10^{-3} which is less than the world average value of 0.29×10^{-3} [16].

Table (1): The activity concentration of ^{238}U , ^{232}Th and ^{40}K and ^{238}U and ^{232}Th content and ^{40}K percentage for soil samples

Sample NO.	Depth in (ft)	^{238}U (Bq/Kg)	^{232}Th (Bq/Kg)	^{40}K (Bq/Kg)	^{238}U (ppm)	^{232}Th (ppm)	^{40}K (%)
1	70	11.54 ± 1.90	5.29 ± 1.43	118.20 ± 8.39	0.93	1.30	0.38
2	1110	17.08 ± 4.83	10.77 ± 3.32	128.00 ± 9.91	1.38	2.65	0.41
3	2400	28.18 ± 3.10	5.99 ± 2.00	88.68 ± 6.92	2.28	1.48	0.28
4	3000	28.18 ± 2.40	----	93.82 ± 6.47	2.28	---	0.30
5	3600	42.94 ± 3.18	8.22 ± 2.38	148.00 ± 9.30	3.48	2.02	0.47
6	4140	27.52 ± 4.44	5.53 ± 1.52	112.10 ± 6.71	2.23	1.36	0.36
7	6930	21.28 ± 3.88	6.16 ± 2.30	138.41 ± 9.44	1.72	1.52	0.44
8	7950	31.64 ± 2.75	3.99 ± 2.10	99.40 ± 7.60	2.56	0.98	0.32
9	8950	28.60 ± 1.61	17.57 ± 2.01	405.00 ± 19.70	2.32	4.33	1.29
10	9920	45.30 ± 3.65	22.95 ± 10.70	715.80 ± 48.00	3.67	5.65	2.29
11	10160	26.56 ± 1.87	29.99 ± 2.68	762.49 ± 14.14	2.15	7.39	2.44
12	10340	56.19 ± 3.95	23.87 ± 11.32	543.83 ± 12.23	4.55	5.88	1.74
13	10520	51.90 ± 3.55	42.13 ± 1.80	513.00 ± 23.80	4.20	10.38	1.64
MAX		56.19 ± 3.95	42.13 ± 1.80	762.49 ± 14.14	4.55	10.38	2.44
MIN		11.54 ± 1.90	3.99 ± 2.01	88.68 ± 6.92	0.93	0.98	0.28
AVRGE		32.07 ± 2.75	14.04 ± 3.35	297.44 ± 14.05	2.60	3.46	0.95

Table (2): Radiation doses and radiation hazard indices for soil samples.

Sample NO.	Depth in (ft)	H_{ex}	H_{in}	D_{out} ($nGyh^{-1}$)	AEDE ($mSvyr^{-1}$)	Ra_{eq} Bq/Kg	ELCR $\times 10^{-3}$	I_{γ} Bq/Kg	AGED ($\mu Svyr^{-1}$)
1	70	0.08	0.11	13.45	0.016	28.20	0.06	0.21	94.87
2	1110	0.11	0.16	19.74	0.024	42.34	0.08	0.31	138.00
3	2400	0.12	0.19	20.33	0.025	43.57	0.09	0.31	139.95
4	3000	0.10	0.17	16.93	0.021	35.40	0.07	0.25	116.52
5	3600	0.18	0.29	30.97	0.038	66.09	0.13	0.47	213.51
6	4140	0.12	0.19	20.73	0.025	44.06	0.09	0.31	143.36
7	6930	0.11	0.17	19.32	0.024	40.75	0.08	0.30	134.96
8	7950	0.12	0.21	21.17	0.026	45.00	0.09	0.32	145.67
9	8950	0.23	0.31	40.71	0.050	84.91	0.17	0.64	288.97
10	9920	0.36	0.48	64.64	0.079	133.24	0.28	1.01	460.68
11	10160	0.35	0.42	62.18	0.076	128.15	0.27	0.98	446.84
12	10340	0.36	0.51	63.06	0.077	132.20	0.27	0.98	444.18
13	10520	0.41	0.55	70.82	0.087	151.65	0.30	1.11	497.57
MAX		0.41	0.55	70.82	0.09	151.65	0.30	1.11	497.57
MIN		0.10	0.11	13.45	0.02	28.20	0.06	0.21	94.87
AVRGE		0.21	0.29	35.70	0.04	75.04	0.15	0.55	251.16

**Fig. (1): The activity concentrations in Bq/Kg for all soil sample.****Fig. (2): The radium equivalent in Bq/Kg for all soil samples**

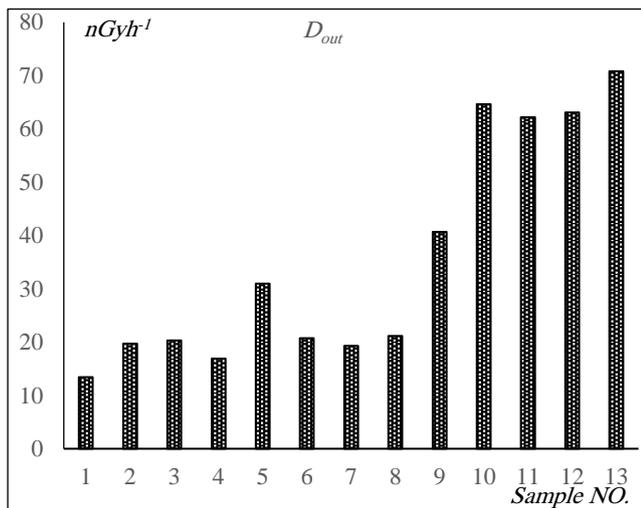


Fig. (3): The outdoor absorbed dose in $nGyh^{-1}$ for all soil samples

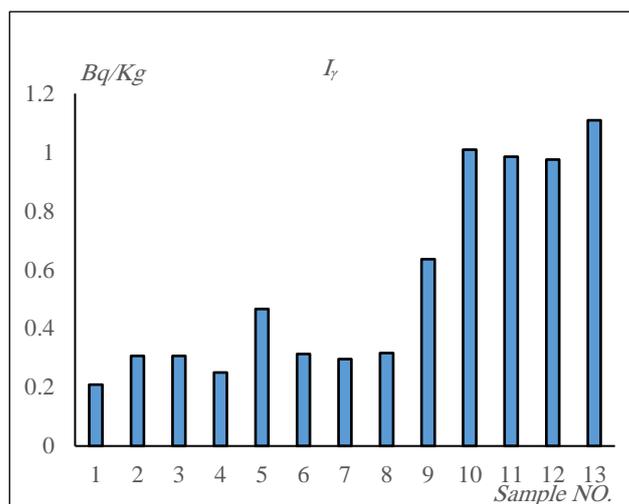


Fig. (6): The representative level index in Bq/Kg for all soil samples

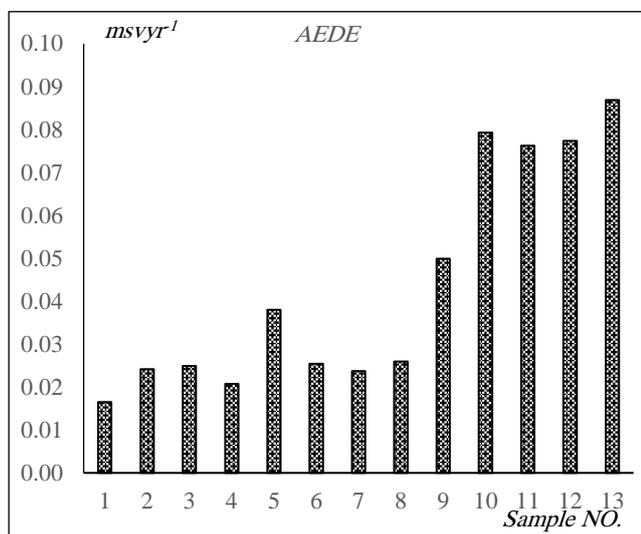


Fig. (4): The annual effective dose in $mSvyr^{-1}$ for all soil samples

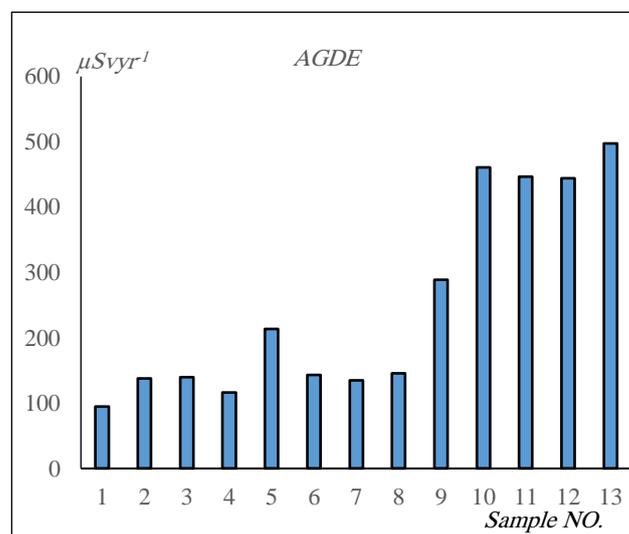


Fig. (7): The annual gonadal dose equivalent in $\mu Svyr^{-1}$ for all soil samples

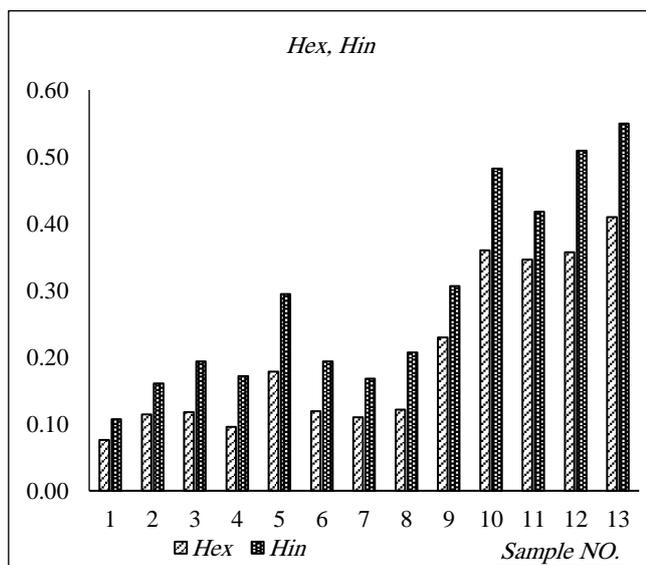


Fig. (5): The external and internal hazard indices for all soil samples

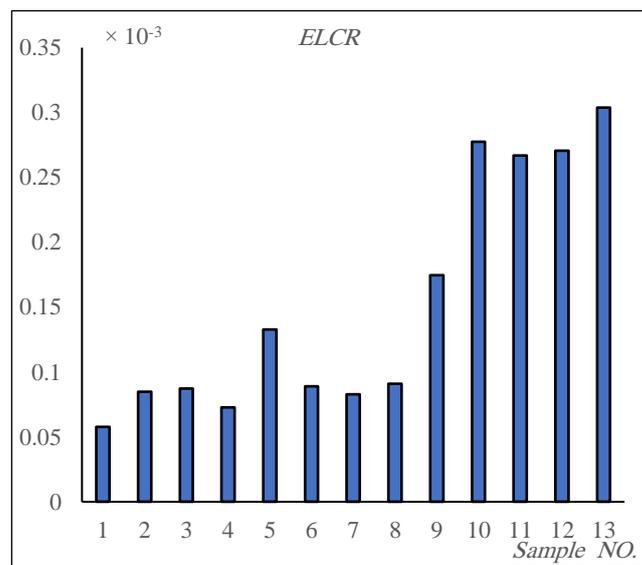


Fig. (8): The excess lifetime cancer risk for all soil samples

From fig. 9, The XRD pattern recorded at room temperature using X-ray diffraction instrument (model Bruker D8, CuK α radiation, $\lambda = 1.5418 \text{ \AA}$) and broadening elimination using a standard Al $_2$ O $_3$ sample

with a corundum structure has showed that potassium element existed in different depths of soil samples with the essential components of it for different diffraction peaks in different orientations.

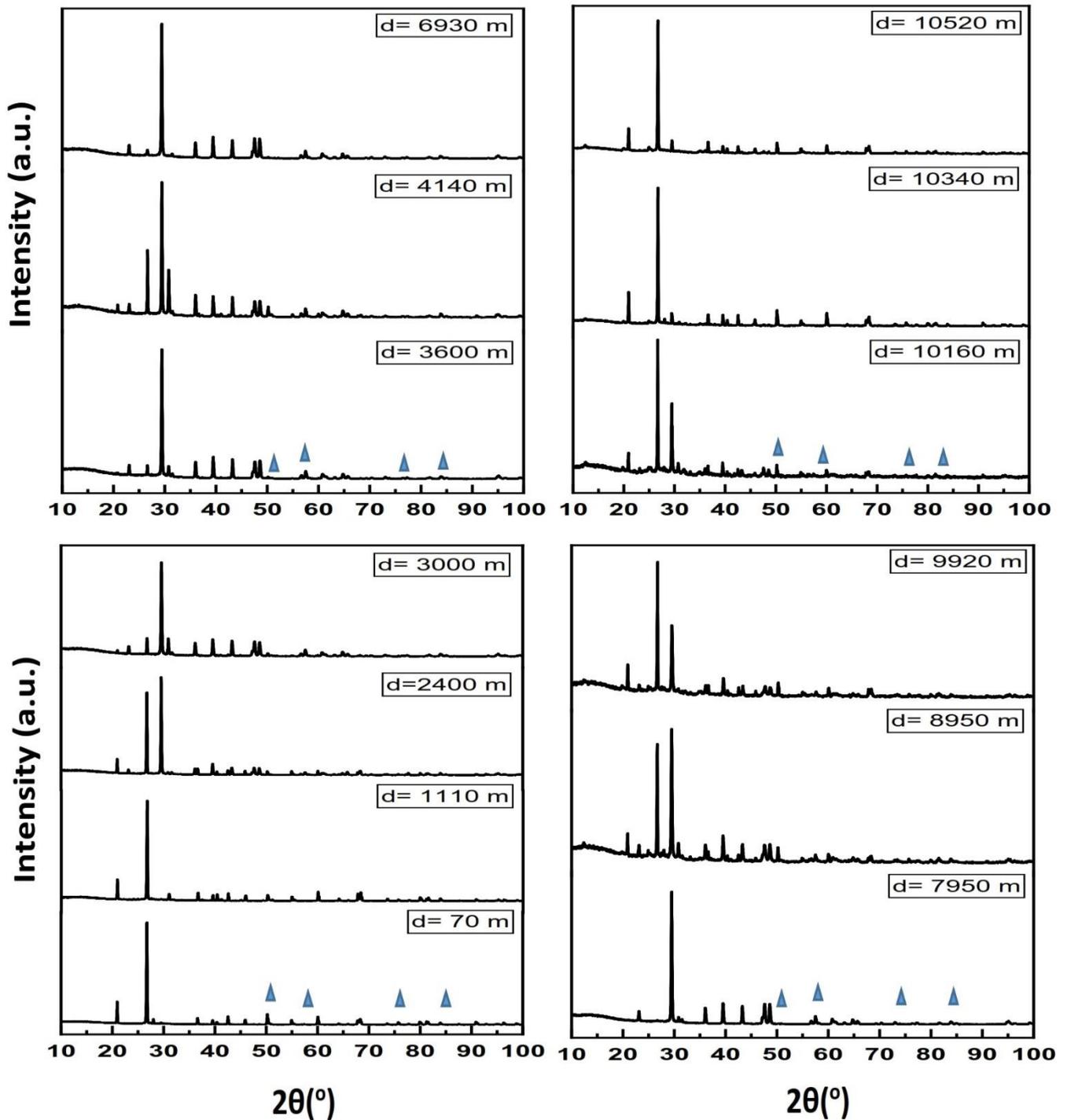


Fig. (9): The XRD pattern for all soil samples

CONCLUSION

In the present study, the primordial radionuclides ^{238}U , ^{232}Th , and ^{40}K in soil samples at different depths from petroleum well in Ras Qattara area, north western desert, Egypt have been estimated using gamma-ray spectroscopy system using N-type HPGe detector. The measured average values of ^{238}U , ^{232}Th and ^{40}K were lower than the average world values reported by UNSCEAR [3]. Radium equivalent (Raeq), external and internal hazard indices, representative index (I_r), the absorbed dose rates (Dout), the annual effective dose (AEDE), excess lifetime cancer risk (ELCR) and annual gonadal dose equivalent (AGDE) were estimated, analyzed and discussed. From the obtained data, the estimated hazard indices have no significant health risk. This study has figured out the baseline information of the radioactivity levels related to petroleum exploration activity in Ras Qattara area, north western desert, Egypt which could be utilized as a reference for future studies.

ACKNOWLEDGEMENTS

The authors are greatly grateful to Assistant Prof. A. M. Abdel Hakeem, Physics department, Faculty of Science, Sohag University for the valuable constructive discussion and comments. They are also greatly grateful to Mr. Mahmoud Hussien Abedel-hakim, Radiation safety department, Atomic energy Authority for the valuable help during measuring sample

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