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Determination and Distribution Map for Radionuclides in Soil Samples from Different Location by Gamma Spectrometry Using Software Analysis

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ABSTRACT

The fundamental goal of the current study is to determine the mean activity concentrations of natural and artificial radionuclides of ^{226}Ra, ^{232}Th, ^{40}K, and ^{137}Cs using gamma spectrometry for three locations, in Egypt, Saudi Arabia, and Iraq, which are significant and vital countries in the Middle East. The mean absorbed dose rate equals 22.35, 28.96, and 43.34 nGy h^{-1} for Egypt, Saudi Arabia, and Iraq. The results are consistent with international reports. The dose contribution percentages for investigated locations are 24 %, 30 %, and 46 % for Egypt, Saudi Arabia, and Iraq, respectively. The obtained results were clarified by statistical measurements using one-way ANOVA test to determine the distribution and differences between the averages of the three groups under study, as they may be influenced by geological variations and human intervention. It was found that the Iraq samples followed a symmetrical, standard normal distribution, while samples from Egypt and Saudi Arabia did not. Statistically significant differences were found between the data from the three countries.

INTRODUCTION

Radioactivity is the emission of radiation from a nuclear reaction or the decay of an unstable nucleus by loss of energy and elementary particles.

Humans on Earth are always subject to various types of radiation. Natural radioactivity sources, or naturally-occurring radioactive materials (NORM), are one of the leading causes of radiation exposure to humans and contribute 81 % of the total. The remaining 19 % comes from sources that can be considered artificial to varying degrees. The development of new technologies has resulted in by-products or waste contributions, both through the creation of artificial radiation sources and radionuclides and through enhancement of concentrations of radionuclides obtained from nature. The later kind of radiation source is known as technologically-enhanced naturally-occurring radioactive materials (TENORM). Secondary production of TENORM wastes from industry or nuclear weapons may generate radiation exposure, not only to the persons directly involved in these activities but also to the local population or even the entire population. As a result, constant attention and monitoring is required during routine operations in such industries. This exposure is mainly caused by external radiation emitted by radionuclides and their decay products. Several occupational radiation exposure assessments in the industry have been recently reported by radiation protection specialists [1-4].

Generally, radionuclides with long half-lives are hazardous to humans as they can enter the human body through the food chain and, in this way, increases radiation burden for a long time [5]. Estimating normal radioactivity in the soil is

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imperative since it makes a difference in checking changes in foundation action with time due to any radioactive decay.

The variety of topography plays an enormous portion in the arrangement of soils by providing the parent fabric. Thus, the radioactivity in the soil is similar to that within the from which it is determined. Variations in concentrations might be related to the differing characteristics of geographical arrangements and the differences in the radioactive material contents within the parent rocks [6].

Environment radioactivity and other sources of external exposure to gamma radiation depend primarily on the topographical conditions, and occur at exceptional levels within the soil of each locale in the world. Radionuclides are present in soil and are incorporated metabolically into plants. Radionuclides, for the most part, are exchanged from the soil to the plants. The physical-chemical parameters of soil play a crucial part in the statement and distribution of radionuclides [6,7].

The standard radionuclide action concentrations of soils and plants are higher in developed ranges due to the impact of composts [6]. Common radionuclides enter the human body through the nourishment chain and the inward breath. They tend to accumulate in different tissues of the human body. Natural radionuclides accumulate in the lungs, liver, skeleton tissues, and kidneys.

Radionuclides are considered dangerous for human beings because they may accumulate in such organs such as the bone, where baneful effects occur. About 66 % of uranium intake is rapidly excreted through urine, while the rest is deposited in the kidneys and several other organs [7].

The current study reviews the distribution map for TENORM and NORM of several soil samples from three locations in Egypt, Saudi Arabia, and Iraq. In addition, statistical analysis using one-way ANOVA was performed to determine the relationship between the three locations and the effect of geological formation elements on dose exposure. Statistical analysis was used to track future developments in the concentration of radioactive materials in these regions and determine their relationship.

MATERIAL AND METHODS

Geological setting

The Middle East is a geographical region that includes southwest Asia and North Africa, as shown in Fig. 1. Geographically, it overlooks the Red Sea, the Arabian Gulf, the Mediterranean, and the Arabian Sea. The Middle East is the cradle of human civilizations. It is also part of the area often called "the ancient world". Many tombs of ancient kings are found in the region. Many of those tombs exhibit high concentrations of radon gas.

Three of the most important countries in this region are: Iraq, where many wars have occurred; Egypt; and Saudi Arabia. Therefore, this study focused on those countries due to the developments they have witnessed and statistically significant comparisons of the concentration of natural and industrial radioactive materials in those countries. Egypt is located around the latitude of approximately 30° 06' N and longitude of approximately 31° 25' E, Saudi Arabia 24° 16' 0.86" N and 45° 06' 28.26" E, and Iraq 33° 00' N and 44° 00' E. The samples were collected from a city in the northern coast of Egypt, El Dammam city in Saudi Arabia, and Mosul city in Iraq.

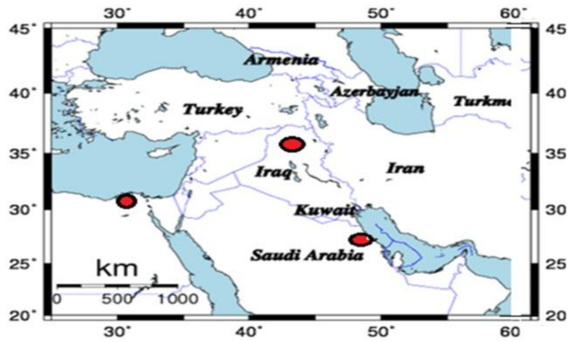

Fig. 1. Geological map of the study locations, including distribution.

Preparation of samples

The current study focused on three Middle Eastern regions, namely, a northern part of Egypt, an eastern region of Saudi Arabia, and Mosul in Iraq. Thirty-seven samples were collected from the three regions and prepared by drying and sifting using a 200-mesh sieve to reach homogeneity. Each sample of nearly 750 g was put in a 500-ml capacity beaker, sealed, and stored for one month to allow a secular equilibrium in the decay chain to be reached. The measurement duration for each sample was nearly 24 hours. A gamma spectroscopy system was used to assess the radionuclides contents of ^{226}Ra, ^{232}Th, ^{40}K, and ^{137}Cs. The authors used SPSS software package version 26 to perform one-way ANOVA test for clarification.

THEORY OF COUNTING PROCESS

Activity concentration

The nuclides' activity concentrations were calculated by using the Eq. (1) [8].

$$AC = \frac{C_S - C_{BG}}{\varepsilon P M} \quad (1)$$

In (1), AC is the activity concentration of each radionuclide in Bq kg^{-1}, C_S is the total count rate of the sample in cps, C_{BG} is the total background count rate in cps, ε is photo-peak efficiency, P is the probability of emission, and M is sample mass in kg.

Absorbed dose rate

The absorbed dose rate was calculated using Eq. (2) [9,10].

$$\mathrm{D} = 0.462AC(Ra) + 0.604AC(Th) + 0.0417AC(K) + 0.030AC(Cs) \quad (2)$$

In (2), D represents the dose rate in nGy h^{-1}, while AC(Ra), AC(Th), AC(K), and AC(Cs) are activity concentrations of radionuclides ^{226}Ra, ^{232}Th, ^{40}K, and ^{137}Cs, respectively, and the unit of multiplication factors in the right-hand side is nGy h^{-1} Bq^{-1} kg. The obtained results are recorded in Table 1.

Table 1. Radiation activity of ^{226}Ra, ^{232}Th, ^{40}K, and ^{137}Cs in Bq kg^{-1} and statistical measurements of radiological parameters hazard from Egypt, Saudi Arabia, and Iraq.

Study area	Minimum	Maximum	Mean	Standard Deviation
The activity of ^{226}Ra (Bq/kg)				
Egypt	3.67	87.24	20.35	23.68
Saudi Arabia	7.24	58.37	21.66	26.79
Iraq	16.21	38.83	32.52	6.49
The activity of ^{232}Th (Bq/kg)				
Egypt	2.61	53.43	10.52	14.14
Saudi Arabia	5.68	82.89	18.80	22.17
Iraq	8.53	16.21	26.31	38.83
The activity of ^{40}K (Bq/kg)				
Egypt	69.68	408.06	158.16	88.58
Saudi Arabia	86.55	326.37	202.15	98.83
Iraq	236.63	613.11	378.94	123.29
The activity of ^{137}Cs (Bq/kg)				
Egypt	0.05	0.41	0.13	0.09
Saudi Arabia	0.05	0.31	0.16	0.11
Iraq	2.18	17.92	8.18	5.55
Absorbed Dose Rate (nGy/h)				
Egypt	7.21	77.49	22.35	18.79
Saudi Arabia	12.39	82.44	28.96	24.46
Iraq	25.06	59.96	43.34	9.81
Outdoor Annual Effective Dose (mSv)				
Egypt	0.01	0.1	0.03	0.02
Saudi Arabia	0.02	0.1	0.04	0.03
Iraq	0.03	0.07	0.05	0.01

RESULTS AND DISCUSSION

Fifteen samples were collected from Egypt, twelve from Saudi Arabia, and ten from Iraq; after they were analyzed using a high-purity germanium detector, the following results were found:

The activity concentrations of ^{226}Ra, ^{232}Th, ^{40}K, and ^{137}Cs in the thirty-seven samples from the three countries in the Middle East region were calculated using Eq. (1). The obtained findings are available in Table 1 and Fig. 6. The activity concentrations of ^{226}Ra and ^{137}Cs were determined at their photopeak energies of 186.1 keV and 661.7 keV, respectively. The primary sources of ^{137}Cs as an artificial radionuclide are the Chornobyl accident and nuclear weapon tests. The findings for Egypt showed that the average activity concentrations of ^{226}Ra ranged from 3.67 Bq/kg to 87.24 Bq/kg with a mean of 20.35 Bq/kg. For Saudi Arabia, they ranged from 7.24 Bq/kg to 58.37 Bq/kg with a mean of 21.66 Bq/kg, while for Iraq, they ranged from 16.21 Bq/kg to 38.83 Bq/kg with mean 32.52 Bq/kg. These findings agree with recent international publications [9,17].

The average activity concentrations of ^{232}Th were recorded as 10.52, 18.80, and 26.31 Bq/kg for Egypt, Saudi Arabia, and Iraq, respectively. The obtained values are lower than those reported by international publications [9,17].

The average activity concentrations of ^{40}K were recorded as 158.16, 202.15, and 378.94 Bq/kg for Egypt, Saudi Arabia, and Iraq, respectively. The obtained values do not reach the values considered risky for health ($\geq$ 400 Bq/kg) and agree with global reports [9,17].

The arithmetic means of activity concentrations of ^{137}Cs were recorded as 0.13, 0.16, and 8.18 Bq/kg for Egypt, Saudi Arabia, and Iraq, respectively. The measured values for Egypt and Saudi Arabia regions are nearly equal due to the high similarity in a geological formation. However, Iraq samples recorded high values, and this is due to the wars that Iraq experienced recently [18,23].

The absorbed dose was calculated and summarized in Table 1. The Egypt samples were in the 7.21-77.49 nGy/h range, with an average of 22.35 nGy/h and a standard deviation of 18.79 nGy/h. The range of Saudi Arabia samples was 12.39- 82.44 nGy/h, with an average of 28.96 nGy/h and a standard deviation of 24.46 nGy/h. The range of Iraq samples was between 25.06 and 59.96 nGy/h with an average of 43.34 nGy/h and a standard deviation of 9.81 nGy/h. Although the results are consistent with international reports [9,11], the Iraqi samples represent higher dose rates than those of Egypt and Saudi Arabia samples due to the high concentration of ^{137}Cs, which is considered an artificial source, and this increase relative to adjacent countries is due to the Iraq wars [18].

From Table 1, it is clear that the outdoor annual effective dose is less than 1 mSv for investigated regions. These results are in line with the previous studies [10,12].

The frequency distribution curve of absorbed dose is shown in Fig. 2 for investigated regions. This curve was evaluated for its excess kurtosis and skewness; the curve follows a standard normal distribution if excess kurtosis equals zero [13-15]. The excess kurtosis for Egypt and Saudi Arabia samples equals 5.04 and 5.03, and the skewness equals 2.20 and 2.13, respectively. Thus, the frequency distribution curve is right-skewed. It is not a regular or symmetric distribution. For Iraq samples, the excess kurtosis equals 0.44 and skewness equals -0.16. This result is near zero; thus, the curve follows a nearly standard normal and symmetric distribution.

The dose contribution percentages for investigated locations are 24 %, 30 %, and 46 % for Egypt, Saudi Arabia, and Iraq, respectively, as shown in Fig. 3. To verify the presence of statistically significant differences in the means of the data for the three countries under study, one-way ANOVA test was used to determine the differences between the averages of the degrees of agreement as influenced by the geological variable between the three groups under study. The geological variables depend on the chemical and physical formation occurring in soil and rock type. They include, for example, mineral or elemental composition, pH, moisture content, and grain size. These variables help increase the activity concentration at the land level [13,20,21, 23,24,25]. In the author's opinion, the Middle East region is nearly similar in its geological formations; thus, the radionuclides contents are close in magnitude.

Table 2 presents the results Kolmogorov-Smirnov and Shapiro-Wilk tests of normality. The tests clarify if the data are normally distributed; the null hypothesis is rejected if the value of P is lower than 0.05. We can assume that our data are average except for Egypt samples [16]. Also, it clears from the standard Q-Q plot of dose rate (in nGy/h) and box plots, and it indicates that the data for the investigated regions are approximately normal, as shown in Fig. 4 and Fig. 5.

Table 2. Results of Kolmogorov-Smirnov and Shapiro-Wilk normality tests.

Locations	Kolmogorov-Smirnov			Shapiro-Wilk		
	Statistic	df	Sig.	Statistic	df	Sig.
Egypt	0.287	15	0.002	0.729	15	0.001
Saudi Arabia	0.235	12	0.067	0.729	12	0.003
Iraq	0.124	10	0.200	0.991	10	0.998

It is clear from homogeneity test that the value of significance is higher than 0.05. Therefore, we will accept the null hypothesis, which is the homogeneity of samples, and thus the variance analysis test can be completed. Table 3 shows that there is homogeneity in the descriptive statistics. It can be noticed that Iraq has the highest value, while Egypt has the lowest value of the 95 % confidence interval.

Table 3. Descriptive statistics of the homogeneity test for absorbed dose rate (Bq/kg).

	N	Std. Error	95 % Confidence Interval for Mean	
			Lower Bound	Upper Bound
Egypt	15	4.85137	11.9475	32.7578
Saudi Arabia	12	5.65979	16.5029	41.4171
Iraq	10	3.10283	36.3189	50.3571
Total	37	3.08848	23.9036	36.431

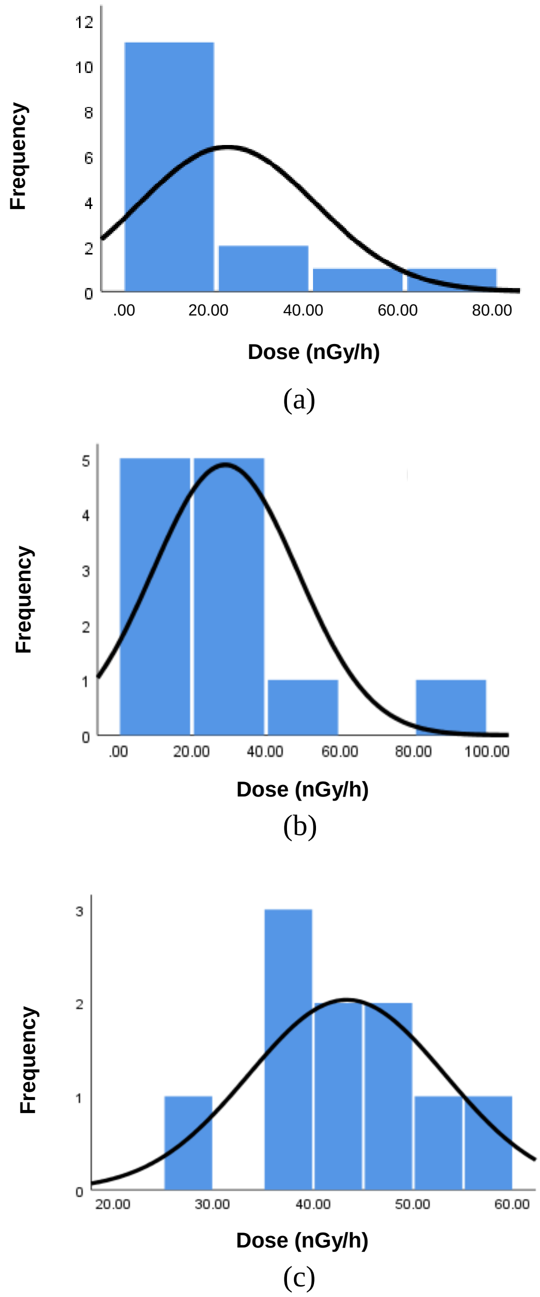

Fig. 2. The frequency distribution curve of absorbed dose for (a) Egypt, (b) Saudi Arabia, and (c) Iraq, respectively.

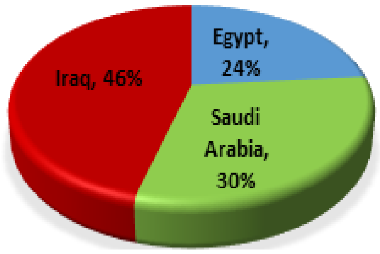

Fig. 3. The contribution percentage of dose for investigated locations.

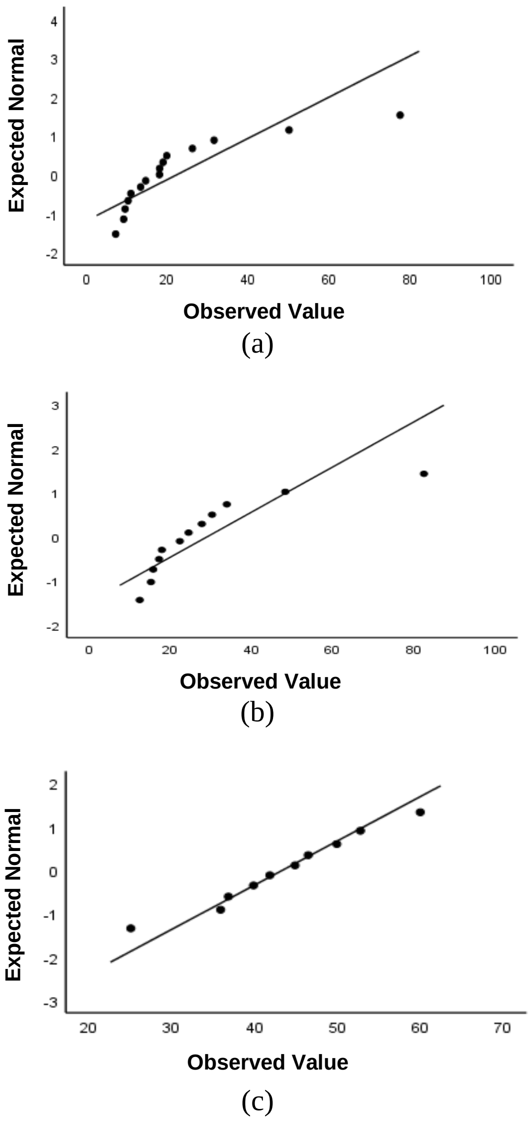

Fig. 4. Normal Q-Q plots of dose (nGy/h) for (a) Egypt, (b) Saudi Arabia, and (c) Iraq, respectively.

Table 4 shows the evidence that there are statistically significant differences between the averages of the radiation dose values for the three countries under study, according to the difference of the geological characteristics, where the *F* was equal to 4.52 with a significance value of 0.018, which is a statistical function at the level of significance equal to 0.05. Moreover, Table 4 finds differences between the three countries with data for Iraq showing highest values. It means that the arithmetic mean of Iraq is 43.34, the largest in agreement with the maximum value (59.96) of the score for the arithmetic means of the other two groups, which were recorded as 22.35 for Egypt and 28.96 for Saudi Arabia.

Table 4. Results of one-way ANOVA test.

Dose (nGy/h)	Sum of Squares	df	Mean Square	F	Sig.
Between Groups	2668.192	2	1334.096	4.519	.018
Within Groups	10037.372	34	295.217		
Total	12705.563	36			

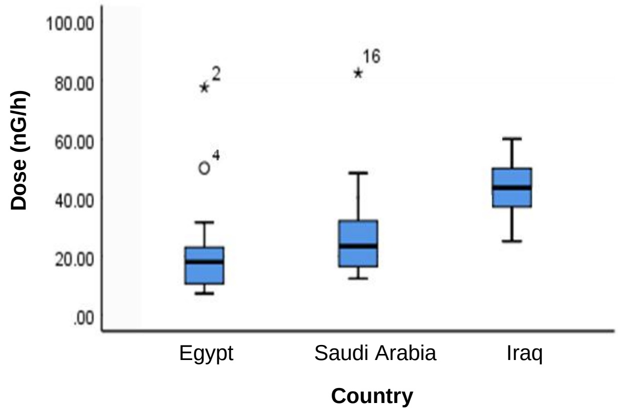

Fig. 5. Box plot for investigated locations.

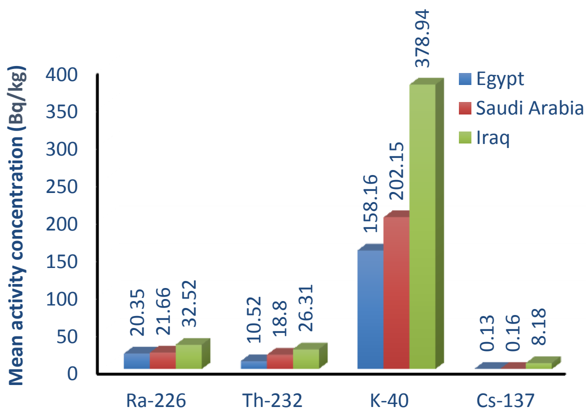

Fig. 6. Mean activity concentration (Bq/kg) for ^{226}Ra, ^{232}Th, ^{40}K, and ^{137}Cs of the studied country.

Table 5. Comparison of activity concentrations for radionuclides ^{226}Ra, ^{232}Th, ^{40}K, and ^{137}Cs of the soil sample from several countries in the region.

Country	Activity concentration (Bq/kg)				Reference
	Ra-226	Th-232	K-40	Cs-137	
Jordan	42	23	309	3.7	[13]
Turkey	37.9	7.1	274.6	7.2	[19]
Kuwait	23.9	14.1	368	---	[20]
Egypt	35.53	23.59	266.41	---	[21]
Saudi Arabia	23.2	7.73	278	1.42	[22]
Qatar	10	17	201	4	[23]

Table 5 compares activity concentrations for ^{226}Ra, ^{232}Th, ^{40}K, and ^{137}Cs. The data for the adjacent countries was in agreement with the current study and global report except for ^{137}Cs which exceeds the internationally permissible safety limit in most countries studied, namely as Turkey, Qatar, and Jordan. The researchers interpreted the presence of Cs-137 due to fallout and geological variables for its soil [13].

CONCLUSION

The current study assessed the average activity concentrations of natural and artificial radionuclides ^{238}U, ^{232}Th, ^{40}K, and ^{137}Cs from Egypt, Saudi Arabia, and Iraq. The obtained results show that the average concentration of radionuclides in Egypt and Saudi Arabia were acceptable; however, samples from Iraq represent a high concentration of Cs-137. The absorbed dose for Iraq samples is considerably higher than for other countries under investigation. Also, the results were analyzed statistically to clarify the difference between the three countries under study using a one-way ANOVA test. Based on the statistical calculations, the null hypothesis was rejected, and the alternative hypothesis was accepted; the alternative hypothesis states that there are statistically significant differences among the three countries under study due to the geological nature variable and the human intervention for each region. These differences indicate that the samples from Iraq exhibit higher activity concentrations. The current results can be considered as a database for future studies.

AUTHOR CONTRIBUTION

Several of the authors collected and prepared the samples. Others calculated and analyzed the data. All of them wrote and reviewed the final manuscript.

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