RA-4

Determination of natural radioactivity and estimation of radiation hazard in cultivated soil samples in El-Minia governorate (Upper Egypt)

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Abstract

Natural radioactivity is a source of continuous exposure to human beings. It originates from both extraterrestrial sources and radioactive elements in the earth crust. Assessment of radioactive elements in the study area is very important from different point of view especially for human health. The natural radioactivity of soil samples were measured by Gamma spectrometry using $3''×3''$ NaI (Tl) well detector. Cultivated soil samples were collected from eight regions in El-Minia governorate including; Maghagha, BaniMazar, Mattay, Samallot, El-Minya, Abo Qurqas, Mallawy, and Deir Mawas district. The activity concentration ranged from $12.43±0.62$ Bq.kg^{-1} to $30.15±1.51$ Bq.kg^{-1} with average $20.58±1.04$ Bq.kg^{-1}for ^{226}Ra, $7.27±0.35$ Bq.kg^{-1} to $25.74±1.28$ Bq.kg^{-1} with average $14.37±0.75$Bq.kg^{-1} for ^{232}Th and $149.24±7.45$ Bq.kg^{-1}to $270.94±13.59$ Bq.kg^{-1} with average $210.20±10.55$ Bq.kg^{-1}for ^{40}K. The measured values are comparable with other worldwide radioactivity and are within the range specified by UNSCEAR 2000 .Also radiological hazard indices were evaluated. All hazard indices are below the permissible limit. Therefore, the radiation hazard and cancer risk are insignificant in the investigated area.

Keywords: activity concentration, radiological hazard, cultivated soil and γ-spectroscopy

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1. Introduction

Natural radioactivity is a main source of continuous exposure to humans where natural sources still contribute almost 80% of the collective radiation exposure of the world's population [1]. Human beings are exposed to ionizing radiation from natural sources throughout their lifetime, and sometime from man-made sources [2]. The two significant natural sources of external radiation to which human are exposed are cosmic rays and terrestrial gamma rays. Terrestrial gamma rays are essentially due to radionuclides belonging to uranium-238 (^{238}U) and thorium-232 (^{232}Th) series and singly occurring potassium-40 (^{40}K) that are present in the earth's crust [3]. Artificial radionuclides can also be present like Caesium-137 (^{137}Cs) is a fission product which is formed through nuclear weapon tests and nuclear power plant accidents [4]. Distributions of ^{238}U, ^{226}Ra, ^{232}Th and ^{40}K in soils depend on the radionuclide distribution in rocks from which they originate and on the processes through which the soils are concentrated [5]. The high natural radiation levels commonly are associated with igneous rocks, such as granite, and lower levels commonly with sedimentary rocks [6]. Soil is the most important source of terrestrial gamma radiation levels, containing trace quantities of terrestrial radionuclides, whose concentrations depend on the local geology of each region in the world [7, 8]. Measurement of natural radioactivity in rocks and soils is vital in determing and monitoring the amount of change of the natural background activity with time for environmental protection [9]. The aim of this work is to estimate the activity concentrations of naturally occurring radionuclides ^{226}Ra, ^{232}Th, and ^{40}K in different soils and to determine the radiation hazard of these soil samples.

2. Materials and methods

2.1. Study area

The present study covered El-Minia governorate about 139 km, from Deir Mawas, in the south (38° 37′ 34″ N ; 30° 98′ 03″ E) to Maghagha, in the north (38° 28′ 39″ N ; 30° 83′ 32″ E) including eight regions: Deir Mawas (11 samples), Mallawy (11 samples), Abu-Qurqas (19 samples), Minya (9 samples), Samallot] (10 samples), Matay (10 samples), Bani Mazar (10 samples), and Maghagha (9 samples). El-Minia governorate is one of the important agricultural and industrial regions in Upper Egypt. El-Minia is mainly an agricultural governorate, as it has 35000 acres distributed in centers of governorate where the highest in Mallawy, Samallot, and Bani Mazar and lowest in El-Edwa and Deir Mawas. El-Minia governorate is famous for cultivation of wheat, corn, sugar cane, sugar beets. In addition to several industrial activities including paking and freezing vegetables,

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fish farming, also there is cement factory in Samalott, and sugar factory in Abo-Quraqas. Fig (1) shows the locations map of the studied area.

2.2. Sampling collection and preparation

A total of 89 soil samples were collected from El-Minia governorate, extended from Deir Mawas city to Maghagha city. Samples have been collected at depth 30 cm from the bottom of soil because the natural terrestrial radiation originates predominantly from the upper layer 30 cm of the soil only. After collection, samples were dried in oven at 110°C for 48 hours to ensure that moisture is completely removed. The samples were crushed, homogenized, and sieved through a 200μm mesh, which is the optimum size enriched of higher heavy minerals content [10].

About 150 gram of each sample were collected in tight plastic containers of 8 cm diameter and 5 cm height. The containers were closed by screw caps and plastic tape was wrapped over the caps[10]. Finally soil samples were sealed for 4 weak to reach secular equilibrium when the rate of decay of the daughters becomes equal to that of the parent [11,12]
.

2.3. Gamma spectrometric analysis

Under the assumption that secular equilibrium was reached between ^{226}Ra and^{232}Th with their decay products, the activity concentration of ^{226}Ra was determined from the average concentration of ^{214}Pb (352 keV) and ^{214}Bi (609,1120 and 1765 keV) and that of ^{232}Th was determined from the average concentration of ^{212}Pb (238 keV), ^{208}Tl (2615 keV), and ^{228}Ac (911 keV) in each sample under study [13]. Since ^{40}K is directly γ-emitter, so its activity concentration could be determined from its single photopeak at 1460 keV.

The activity concentration has been determined by using gamma ray spectrometer which consists of 3'×3' NaI(Tl) scintillation well detector with multichannel analyzer MCA and its electronic circuits. The detector had a photopeak efficiency of about 1.2×10^{-5} at 1332 keV for ^{60}Co and an energy resolution of 7.5 at 662 keV for ^{137}Cs and operation bias voltage 1000 V dc. The detector is shielded with a 6 cm lead castle that is lined inside with a Cu sheet. The measurement time of activity was 43200 s. the measured γ-ray spectrum were analyzed by software program Maestro 32. ^{60}Co source (1173.2 and 1332.5 KeV) and ^{137}Cs source (662 KeV) are used for energy calibration.

The efficiency calibration was performed using standard source sample which contain a known activity of one or more gamma ray emitters of the radionuclides ²²⁶Ra (351.99, 609.32, and 1764.51 KeV) and ²³²Th (238.63 KeV).

The efficiency calibration curve was made using different energy peaks covering the range up to ≈ 2000 kev and fig(2) shows that Experimental and theoretical efficiency curves for 3″ x 3″ NaI(Tl) well detector. Eq(1) is used for calculating the absolute efficiency[11].

$$\eta_{Exp} = \frac{N_p \cdot 100}{I_\gamma . TOC . A_{BOC}} \qquad \textbf{Eq(1)}$$

Where
N_p is the net peak area (Count/S), I_γ the intensity of emitted γ-ray (%), T_{OC} the time of counting (s), and A_{BOC} the activity (Bq) of the standard source.

3. Results and Discussion

3.1. Activity analysis

By using gamma-ray spectrometer, activity concentrations of the natural radionuclides were investigated in the soil samples. The activity concentrations of the radionuclides in the meaured samples were computed using the following relation [15]:

$$A = \frac{N_p}{e \times \eta \times m} \qquad \textbf{Eq(2)}$$

Where
N_P is the count per second, e is abundance of the γ- peak in a radionuclide, η is the measured efficiency for each gamma-ray peak observed for the same number of channels either for the sample or standard source, and m is sample mass in kilograms. The three most important primordial radionuclides investigated in the area of interest were ²²⁶Ra, ²³²Th and ⁴⁰K [16].

The activity concentrations of ²²⁶Ra, ²³²Th and ⁴⁰K radionuclides in soil samples are presented in Table 1 and illustrated in Fig(3).

The activity concentration ranged from 12.43±0.62 Bq.kg⁻¹ to 30.15±1.51 Bq.kg⁻¹ with average 20.58±1.04 Bq.kg⁻¹ for ²²⁶Ra, 7.27±0.35 Bq.kg⁻¹ to 25.74±1.28 Bq.kg⁻¹ with

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average14.37±0.75Bq.kg⁻¹ for ²³²Th , and149.24±7.45 Bq.kg⁻¹ to 270.94±13.59 with average 210.20±10.55 Bq.kg⁻¹ for ⁴⁰K.

The obtained results were compared with other studies in Egypt and some other countries as listed in table (2). The overall results show that the activity concentration of ²²⁶Ra, ²³²Th and ⁴⁰K are not uniformly distributed in soil. The variations of the activity concentration depends on the geochemical nature of the soil, the obtained mean values of the activity concentrations are lower than the recommended value of the world average(30 Bq.kg⁻¹ for ²²⁶Ra, 35 Bq.kg⁻¹ for ²³²Th, and 370 Bq.kg⁻¹ for ⁴⁰K). It is also observed that the measured activity concentration of ⁴⁰K exceeds markedly the values of both Uranium and Thorium, as it is the most abundant radioactive element under consideration. Moreover the excessive use of the Potassium containing fertilizers in the area adjacent to the sampling sites may contribute to the higher values of ⁴⁰K activity [17].

3.2. The radiological hazards:

Radiation hazard due to specified radionuclide ²²⁶Ra, ²³²Th, and ⁴⁰K were assessed by different indices according to UNSCEAR, 2000 to arrive at a safe conclusion on the health status of an exposed person or environment. The most widely used radiation indices are presented in table 3.

3.2.1. The Radium Equivalent Activity (Raeq):

The Raeq index represents a weighted sum of activities of the above mentioned natural radionuclides and is based on the estimation that 1 Bq·kg⁻¹ of ²²⁶Ra, 0.7 Bq·kg⁻¹ of ²³²Th, and 13 Bq·kg⁻¹ of ⁴⁰K produces the same gamma radiation dose rates. The index is given as
:

$$\text{Raeq} = C_{Ra} + (1.43\ C_{Th}) + (\ 0.077\ C_K\ ) \qquad \text{Eq(3)}$$

Where
C_{Ra}, C_{Th} and C_K are the average activity concentration in the sample in Bq.kg⁻¹ of ²²⁶Ra, ²³²Th, and ⁴⁰K respectively [18].

3.2.2. The Absorbed Dose Rate

The absorbed dose rate (Dr) in air at average gonad height of one meter above the surface of ground due to the natural radionuclides ^{226}Ra, ^{232}Th and ^{40}K was estimated using the formula given as [19,20]:

$$D = 0.427 \, C_{Ra} + 0.662 C_{Th} + 0.0423 \, C_{K} \qquad\qquad Eq(4)$$

Where
C_{Ra} is the average of the activity concentration of ^{226}Ra in the sample , C_{Th} is the average of the activity concentration of ^{232}Th in the sample, and C_{K} is the average of the activity concentration of ^{40}K in the sample, in Bq kg^{-1}.

3.2.3. The Annual Effective Dose Equivalent (AEDE):

The annual effective dose equivalent (AEDE) to the population can be calculated using the conversion coefficient from absorbed dose in air to effective dose (0.7 Sv·Gy^{-1}) the indoor to outdoor ratio (1.4), the outdoor occupancy factor 0.2 and the indoor occupancy factor 0.8. Therefore, the annual effective doses outdoors and indoors equivalent are calculated by using the relations [18,21].

$D_{outdoor}$ (mSv/yr) = [Dr. (mGy/hr) × 24 hr × 365.25 d × 0.2 × 0.7 Sv/Gy] × 10^{-6}
$$Eq(5)$$

D_{indoor} (mSv/yr) = [Dr. (mGy/hr) × 24 hr × 365.25 d × 1.4 × 0.8 × 0.7 Sv/Gy] ×10^{-6}
$$Eq(6)$$

The corresponding worldwide values of D_{out} and D_{in} and D_{tot} are 0.08, 0.42 and 0.50 mSv·y^{-1}, respectively UNSCEAR 2000

3.2.4 The External and Internal Hazard Index (H$_{ex}$, H$_{in}$):

The external (H$_{ex}$) and internal (H$_{in}$) hazard index due to the emitted γ-rays of the soil samples were calculated and examined according to the following formula:

$$H_{ex} = \frac{C_{Ra}}{370} + \frac{C_{Th}}{259} + \frac{C_{K}}{4810} \leqslant 1 \qquad\qquad Eq(7)$$

$$H_{in} = \frac{C_{Ra}}{185} + \frac{C_{Th}}{259} + \frac{C_{K}}{4810} \qquad\qquad Eq(8)$$

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The value of H_{ex} must be lower than unity in order to keep the radiation hazard insignificant. This is the radiation exposure due to the radioactivity from a construction material, limited to 1.5 mGy·y^{-1}. The maximum values of H_{ex} equal to unity correspond to the upper limit of Ra_{eq} (370 Bq·kg^{-1}) [22].

3.2.5. Representative level index($I_{γr}$)

An additional hazard index so called representative level index was calculated by using the formula [23,24]:

$$I_{γr} = \frac{C_{Ra}}{150} + \frac{C_{Th}}{100} + \frac{C_K}{1500} \qquad\qquad Eq(9)$$

Where
C_{Ra}, C_{Th} and C_K are the activities of ^{226}Ra, ^{232}Th and ^{40}K, respectively, in Bq kg$^{–1}$. The value of $I_{γr}$ must be less than unity in order to keep the radiation hazard insignificant.
As shown in table (3) the radium equivalent Ra_{eq} ranged from 34.17 to 70.93 Bq.kg^{-1} with average 56.38 Bq.kg^{-1}.It is inferred that for all the soil samples analyzed, the radium equivalent activity value is well within and less the permissible limits of 370 Bq·kg^{-1}.

The absorbed dose rate (D_r) ranged from 16.96 to 33.44 with average 24.96 nGyh^{-1} with average 26.75 nGyh^{-1}, these values of dose rates are less than the allowed limit 59 nGyh^{-1}. Radioactivity level index ($I_γ$) ranged from 0.26 to 0.51 with average 0.41. The calculated values for most samples were lower than the international values (Iγ < 1).
External hazards (H_{ex}) ranged from 0.14 to 0.19 with average 0.15 where the calculated average values were less than the acceptable value (1.5 mGy·y^{-1}). The maximum values of H_{ex} equal to unity correspond to the upper limit of Ra_{eq} (370 Bq·kg^{-1}).
The annual effective dose rate (AEDE$_{out}$) ranged from 20.58 to 41.33μSvy^{-1} with average 31.54μSvy^{-1}values were lower than the corresponding worldwide values of 80 μSvy^{-1}

The measured results are lower than the recommended international limits. Therefore,the study area is still in the limits of normal radiation level.

4. Conclusion:

The activity concentrations of naturally occurring radionuclides ^{226}Ra, ^{232}Th, and ^{40}K in 131 soil samples from El-Minya governorate have been determined by gamma spectroscopy. The activity concentrations were found to be from12.43±0.62 Bq.kg^{-1} to

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30.15±1.51 Bq.kg^{-1} with average 20.58±1.04 Bq.kg^{-1}for ^{226}Ra, 7.27±0.35 Bq.kg^{-1} to 25.74±1.28 Bq.kg^{-1} with average14.37±0.75Bq.kg^{-1} for ^{232}Th and149.24±7.45 Bq.kg^{-1}to 270.94±13.59 Bq.kg^{-1} with average 210.20±10.55 Bq.kg^{-1}for ^{40}K. The radiation hazard indices [The radium equivalent (*Raeq*), the absorbed dose rate (*D$_r$*), radioactivity level index (*I$_\gamma$*) ,external hazards (*H$_{ex}$*) , and the annual effective dose rate (AEDE$_{out}$)] are calculated. The measured results are lower than the recommended international limits.

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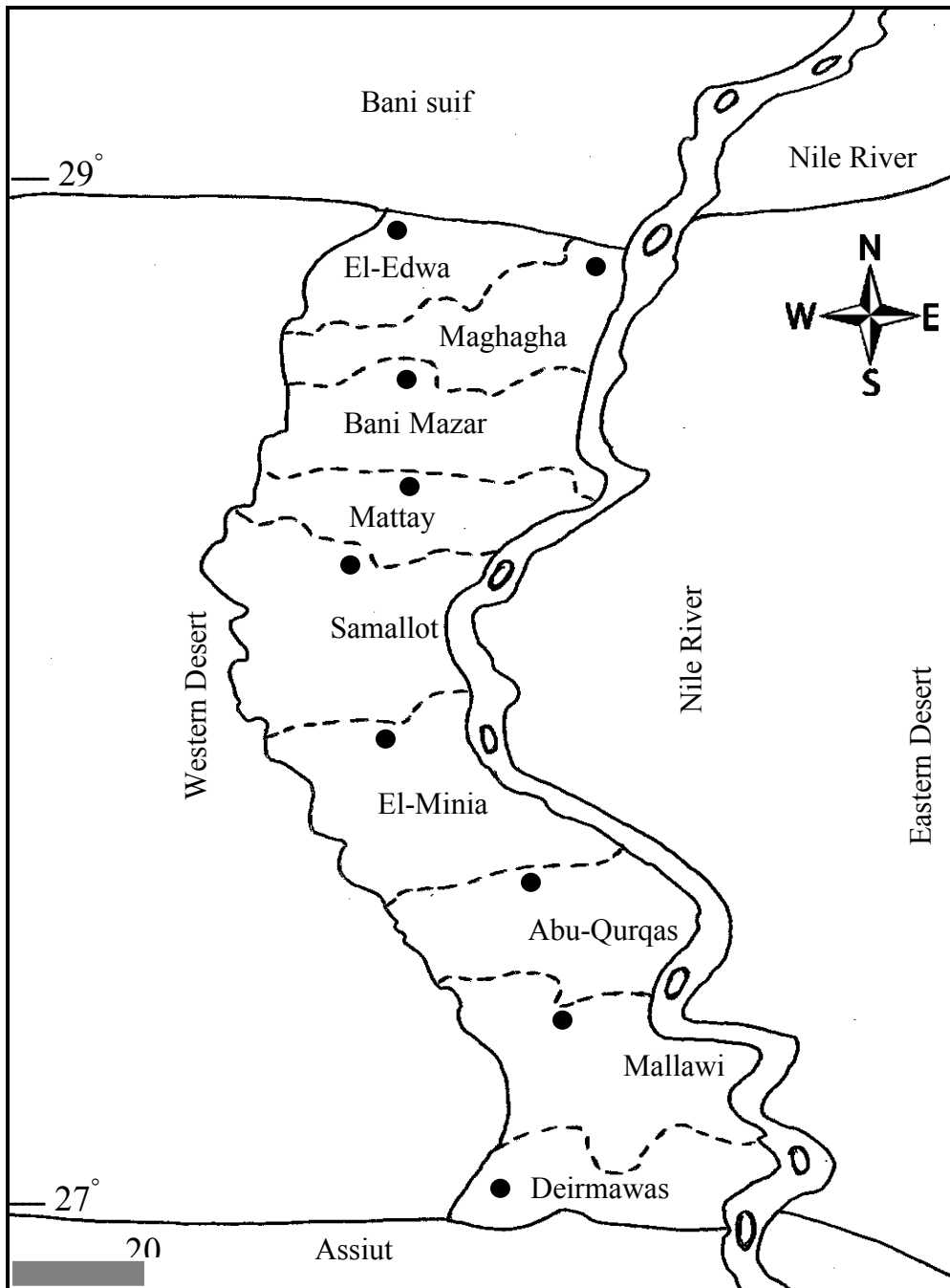
Fig(1): locations map of studied samples.

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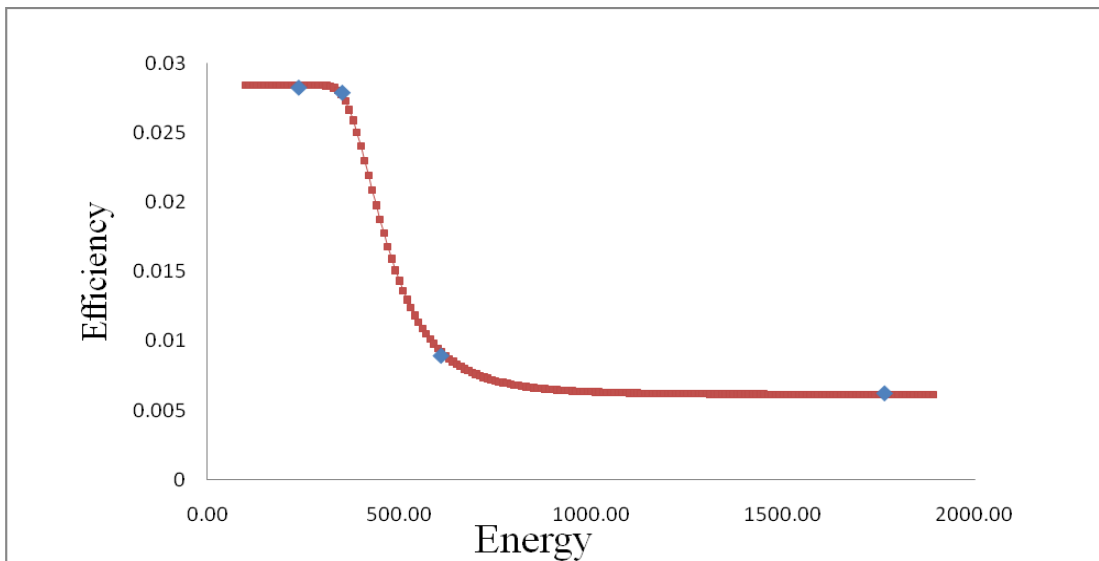Engineering**
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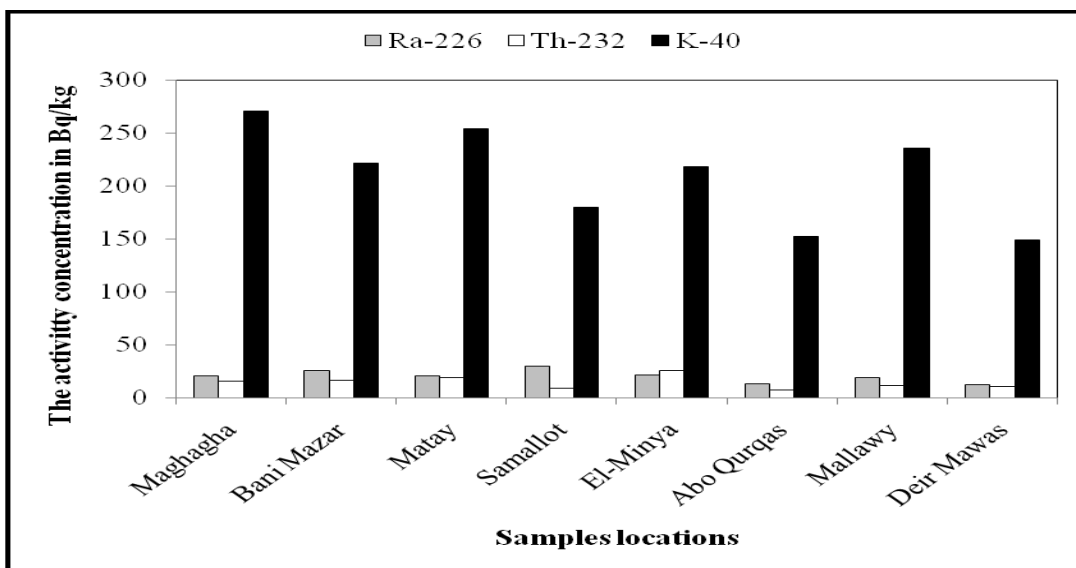**Fig 2. Experimental and theoretical efficiency curves for 3″ x 3″ NaI(Tl) well detector.**

Fig 3. The activity concentrations of radionuclides in soil samples collected from El-Minia Governorate

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Table 1. The activity concentrations of radionuclides in soil samples collected from El-Minia Governorate

Samples locations	Number of samples	activity concentration (Bq.kg^{-1})		
		^{226}Ra	^{232}Th	^{40}K
Maghagha	9	20.67±1.14	15.52±0.77	270.94±13.59
BaniMazar	10	26.24±1.32	16.54±0.83	221.27±10.99
Matay	10	20.84±1.04	18.82±0.94	254.32±13.15
Samallot	10	30.15±1.51	9.33±0.46	179.80±8.98
El-Minya	9	21.85±1.08	25.74±1.28	218.13±10.87
Abo Qurqas	19	13.35±0.66	7.27±0.35	152.16±7.61
Mallawy	11	19.15±0.96	11.29±0.8	235.75±11.82
DeirMawas	11	12.43±0.62	10.5±0.53	149.24±7.45
Min		12.43±0.62	7.27±0.35	149.24±7.45
Max		30.15±1.51	25.74±1.28	270.94±13.59
Average		20.58±1.04	14.37±0.75	210.20±10.55

Table (2): comparison between activities of ^{226}Ra, ^{232}Th and ^{40}K for all soil samples under present study and other countries.

Country	Note	Activity (Bq/kg)			References
		^{226}Ra	^{232}Th	^{40}K	
Egypt (Qena)	Farm soil	13.7	12.3	1233	24
Egypt (The Nile Delta	Farm soil	16.6	18.1	316.0	25
and Middle Egypt)					
Nigeria	Farm soil	55.3	26.4	505.1	26
Palestine	Farm soil	34.5	23.8	120.0	27
Saudi Arabia	Farm soil	44.87	54.59	2652.3	28
Malaysia	Farm soil	127	304	302	29
Turkey	Farm soil	48.35	20.48	744.76	30
Sultanate of Oman	Farm soil	14.42	9.95	158.21	31
Yemen	Farm soil	30.41	36.26	358.12	18
Brazil	Farm soil	47	29	678	19
UNSCEAR 2000	Soil	30	35	370	1
Present work		20.58	14.37	210.20	

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Table 3. The radium equivalent (*Raeq*), the absorbed dose rate (*D_r*), radioactivity level index (*I_γ*) ,external hazards (*H_{ex}*) , and the annual effective dose rate (AEDE$_{out}$) of the soil samples collected from El-Minia governorate.

Samples location	Number of samples	Ra_{eq} $(Bq.kg^{-1})$	D_r $(n.Gy^{-1})$	I_γ	H_{ex}	$AEDE_{out}$ $(\mu Sv.h^{-1})$
Maghagha	9	65.81456	31.4571	0.487657	0.177763	27.92
Bani Mazar	10	62.29549	29.37685	0.456333	0.168283	36.05
Matay	10	68.05218	32.50655	0.502843	0.183797	39.89
Samallot	10	57.36833	26.67033	0.4144	0.155	33
El-Minya	9	70.93133	33.44333	0.514667	0.191533	41.33
Abo Qurqas	19	34.1748	16.96141	0.263352	0.14249	20.58
Mallawy	11	53.47232	25.07725	0.397907	0.144427	30.77
Deir Mawas	11	38.95748	18.58113	0.287512	0.105219	22.8
average		56.38331	26.75924	0.415584	0.158564	31.54