ENVIRONMENTAL GAMMA-RAY MEASUREMENTS OF NATURALLY RADIOACTIVE SAMPLES IN THE REGION OF FERTILIZER FACTORIES

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ABSTRACT

AHPGe high-resolution gamma-ray spectrometer was used to measure the terrestrial gamma radioactivity of plant samples collected from the surrounding area of Abu Zabal Company for fertilizers and chemical industries in Egypt.

Each plant sample was dried, meshed, sealed in 250 ml cylindrical vials and measured in the laboratory for 48 hours in order to determine the radioactivity concentrations of naturally occurring isotopes ^{226}Ra, ^{232}Th and ^{40}K in them.

The activity concentration of ^{226}Ra was determined from the intensity of the gamma-lines of ^{214}Pb (352 keV) and ^{214}Bi (609 keV), while that of ^{232}Th - from gamma-lines of ^{228}Ac (911 keV) and ^{212}Bi (727 keV). In addition, ^{40}K activity concentration was determined from the intensity of the characteristic gamma-line at ~1460 keV.

The activity concentration of ^{226}Ra was found to vary from ~1.26Bq/kg to ~32.43 Bq/kg that of ^{232}Th ranged from ~1.20Bq/kg to ~16.67Bq/kg.

Finally, the gamma radioactivity concentration of ^{40}K was found to lie in the interval from ~397.71 Bq/kg to ~1729.68 Bq/kg.

The specific radioactivity mean-values of ^{226}Ra, ^{232}Th and ^{40}K in plant samples were found to be (6.50 ± 8.94)Bq/kg, (7. 80± 5.39)Bq/kg and (729.85 ± 344.93)Bq/kg, respectively.

INTRODUCTION

Most of the living organisms on the earth are showing natural radiation, which is typically due to the activity concentration of primordial radionuclides ^{232}Th,^{238}U and their product of decay, in addition to the other naturally radionuclide^{40}K present in the earth shell[1]. In various geological formations like soils, rocks, plants, sand, water and air the natural radioactivity is wide spread in the earth's environment. Therefore, humans should be careful of their natural environment with regard to the radiation health effects. Some of the radiation health effects are chronic lung diseases, acute leucopoenia, anemia and necrosis of the mouth. Thorium exposure can cause lung, pancreas, hepatic, bone, kidney cancers and leukemia [2]. Nowadays, the scientific subjects that attract public attention are the human exposure to ionizing radiation, since radiation of natural origin is mainly responsible about the most of the whole radiation exposure of the human population.

In the recent years, there are many studies done on the high background radiation areas in the world to show the risk estimation due to long term low-level whole body exposures to the public, such as Australia, Brazil, China, India, Iran, Japan, etc., possess high levels of natural radiation. The high radiation levels are due to the presence of large quantities of naturally occurring radioactive minerals in the rocks, soils, sediments, etc. [3]. In order to have environmental protection, the monitoring of any release of radioactivity to the environment is important. Fast and accurate methods for examine of radioactivity are necessary for the monitoring. Low level gamma spectrometry is suitable for both qualitative and quantitative determinations of gamma emitting nuclides in the environment [4].

This study used to determine the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in the plant samples from the surrounding area of Fertilizer Company. In order to understand the occurrence and distribution of natural radionuclides of plant samples in the area under investigation and evaluate potential health hazards; the representative level index, $I_{\gamma r}$, the radium equivalent activity (Ra_{eq}), and the annual effective dose equivalent (AEDE) in the air for all plant samples were estimated to assess the contribution of this radionuclide to public exposure.

The factory was established in 1974 to manufacture all kinds of phosphate fertilizers and chemicals. The fertilizer production facility produces Single Super phosphate, Triple Super phosphate, Phosphoric Acid, Ammonium nitrate and Sulphuric Acid, the ores which used in these productions are Phosphate Stone, Sulphur, Ammonium Sulphate and Potassium Sulphate. The choice of that area to study the contamination level caused due to that company lie in the agricultural land of Abu Zabal city and for quality control for the hazards of fertilizer companies in Egypt.

EXPERIMENETAL WORK

Samples location

The Plant samples collected along 2000 m, with separation 200 m for each one, surrounding the site under investigation as shown in the map in figure (1). P0 at the center of the factory and from P1L to P5L at the left side of it and from P1R to P5R at the right side, and also collect 2 samples away from the both sides of the company by 6000 m P6L at the left and P6R on the right side of the factory.

Figure 1: The map of the Abu Zabal company for fertilizers and chemical industries and the cites of the collected samples of the plants.

Samples Preparation

All the 13 plant samples (500 gm) each were collected and then washed to remove possible contaminants. The samples were dried in air for 72 hour, then dried in an oven for 72 hours at 70°C to constant weight, ground into a powder using a mortar and pestle and meshed perfectly to pass 2 mm mesh for homogeneity. Finally, each sample was packed, sealed in polyethylene containers (250 ml) and left for 4 weeks before counting, in order to allow the in-growth of Uranium and Thorium decay products and achievement of secular equilibrium ^{226}Ra and ^{222}Rn with their respective progenies[5,6].

Detection System Setup

The energy and intensity of various gamma ray lines have been measured using CANBERRA coaxial (HPGe) detector Model number GC4018 of relative efficiency 40% coupled to a 16384 channel analyzer. The full width at half maximum (FWHM) was found to be 1.8keV for ^{60}Co –1.33MeV gamma ray line.

In these measurements, the standard point sources ^{241}Am, ^{133}Ba, ^{207}Bi, ^{139}Ce, ^{134}Cs, ^{137}Cs, ^{152}Eu, ^{153}Gd, ^{54}Mn, ^{22}Na, ^{88}Y, ^{65}Zn and ^{60}Co were used for energy and efficiency calibration for the detector. The certificates of the sources activities and their uncertainties and confidence levels for sources are listed in table (1). The data sheet states the values of the half-lives, the photon energies and the photon emission probabilities per decay for the all radionuclides used in the calibration process are listed in table (2), which available from the National Nuclear Data Center Web Page or on the IAEA website.

Table 1. Point sources activities and their uncertainties.

Nuclide	Activity(KBq)	Reference Date	Uncertainty %	Confidence Level
^{241}Am	102.7	1 October 2014	10	0.95
^{133}Ba	58.9			
^{207}Bi	53.9			
^{139}Ce	172.8			
^{60}Co	105.3			
^{134}Cs	106.4			
^{137}Cs	110			
^{152}Eu	53.6			
^{153}Gd	124			
^{54}Mn	122.7			
^{22}Na	125.1			
^{88}Y	236.7			
^{65}Zn	107			

The spectrum acquired with the Genie 2000 data acquisition and analysis software made by Canberra using its automatic peak search and peak area calculations, along with changes in the peak fit using the interactive peak fit interface when necessary to reduce the residuals and error in the peak area values. The live time, the run time and the start time for each spectrum were entered into the spreadsheets. These sheets were used to perform the calculations necessary to generate the experimental full energy peak efficiency (FEPE) curves with their associated uncertainties as a function of the photon energy for cylindrical HPGe detectors.

Table 2. Half-lives, photon energies and photon emission probabilities per decay for the all radionuclide's used in this work.

Nuclide	Energy (Kev)	Emission Probability %	Half Life (Year)
^{241}Am	59.53	35.78	432.200
^{133}Ba	80.99	32.9	10.520
	276.39	7.164	10.520
	302.85	18.34	10.520
	356.01	62.05	10.520
	383.85	8.94	10.520
^{207}Bi	569.7	97.76	32.200
	1063.66	74.58	32.200
	1770.23	6.87	32.200
^{139}Ce	165.85	79.9	0.377
^{60}Co	1173.228	99.85	5.270
	1332.492	99.982	5.270
^{134}Cs	569.32	15.38	2.067
	604.72	97.65	2.067
	795.83	85.5	2.067
	801.945	8.7	2.067
^{137}Cs	661.657	84.99	30.000
^{152}Eu	121.78	28.41	13.542
	244.69	7.5	13.534
	344.28	26.58	13.534
	411.11	2.237	13.534
	778.9	12.96	13.542
	867.38	4.241	13.542
	964.13	14.62	13.542
	1112.11	13.4	13.542
	1408.01	20.85	13.542
^{153}Gd	97.43	30.2	0.662
	103.18	21.4	0.662
^{54}Mn	834.838	99.976	0.855
^{22}Na	511	179.79	2.602
	1274.53	99.944	2.602
^{88}Y	898.036	93.9	0.292
	1836.052	99.38	0.292
^{65}Zn	1115.539	50.6	0.668

The ETNA program (an acronym standing for Efficiency Transfer for Nuclide Activity measurements) used to convert the Full Energy Peak Efficiency (FEPE) curve for using point sources to the (FEPE) for using volumetric source. This program developed in the Laboratories National Henri Becquerel (BNM/LNHB) CEA/ Saclay, France by Marie Christine [7].

Sample Counting

Each sample was put into the shielded HPGe detector and measured for 48 hours, the background gamma radiation was determined in the laboratory by an empty cylindrical vial under identical conditions also for 48 hours. The obtained spectrum of the background gamma radiation was subtracted from the measured gamma ray spectra of each sample. A selected one of the obtained spectrum is shown in figure (2).

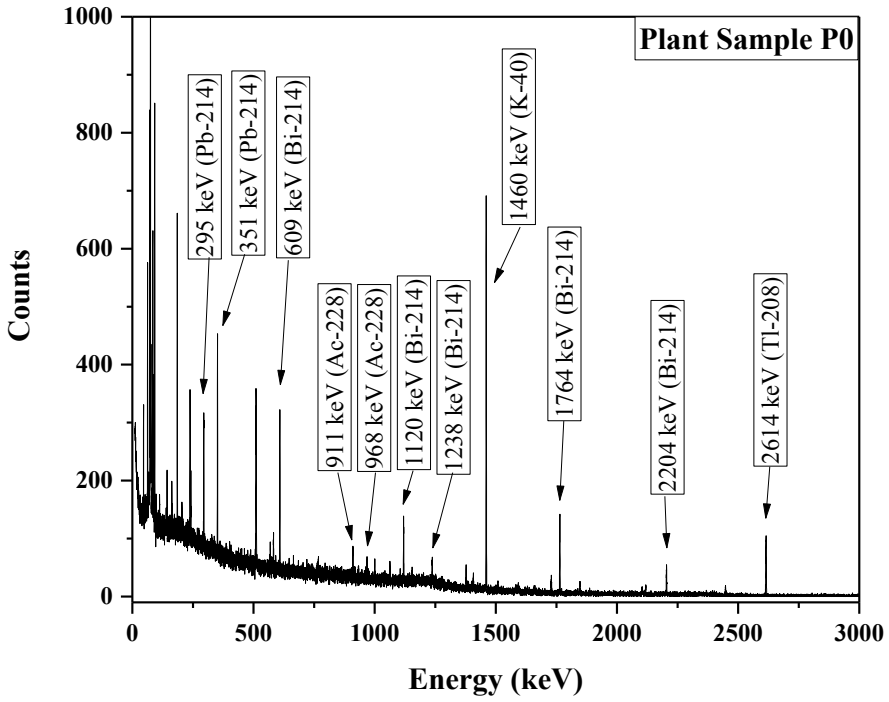

Figure 2: The main part of a gamma-ray spectrum measured for sample P0.

The spectra of all samples were perfectly analyzed using Genie 2000 data acquisition and analysis software to calculate the activity concentrations of the ^{40}K , ^{226}Ra and ^{232}Th.

Evaluation of Activity Concentration

The activity concentrations of the natural radionuclides in the measured samples were calculated using the following relation [8]:

$$A_s\ (Bq / kg) = C_a / \varepsilon \times P_r \times M_s \qquad (1)$$

where, C_a is the net gamma counting rate (counts per second), ε the detector efficiency of the specific gamma-ray, P_r the absolute probability of gamma decay and M_s the mass of the sample (kg). Activity concentrations of ^{226}Ra were determined using the gamma energy lines of ^{214}Pb (352 keV) and ^{214}Bi (609 keV). While that of ^{232}Th was obtained from gamma lines of ^{228}Ac (911 keV) and ^{212}Bi (727 keV). The ^{40}K activity concentration was finally determined using the 1460 keV gamma line [9].

Radium equivalent activities (Ra_{eq})

To assess the radiological hazard of the plant samples, it is useful to calculate anindex called the radium equivalent activity, Ra_{eq}, it is proposed that activity concentration of 259 Bq/ kg of ^{232}Th and 4810 Bq/kg of ^{40}K are equivalent to 370 Bq/kg of ^{226}Ra, and in these quantities, each of these radionuclide gives an effective dose of 1.5 mGy/ year. On the basis of these values Ra_{eq} is defined as the following [10]:

$$Ra_{eq} = A_{Ra} + 1.43\ A_{Th} + 0.077\ A_K \quad (2)$$

where A_{Ra}, A_{Th} and A_K are the activity concentration in (Bq/kg) of ^{226}Ra (^{238}U-series), ^{232}Th and ^{40}K, respectively.

Radiation Hazard Index (H_{ex}, H_{in})

In order to estimate the level of gamma radiation hazard associated with natural radionuclides in specific plant samples, this factor is used. The external hazard index is obtained from Ra_{eq} expression during the assumption that its maximum value acceptable corresponds to the upper limit of Ra_{eq} (370 $Bq{\cdot}kg^{-1}$) according to [11]. This index value must be less than unity in order to keep the radiation hazard insignificant; then, the external hazard index (H_{ex}) can be defined as the potential radiological hazard posed by the different samples was calculated using the following equation [12].

$$H_{ex} = \frac{1}{370}A_{Ra} + \frac{1}{259}A_{Th} + \frac{1}{4810}A_K \quad (4)$$

In addition, the external hazard, radon and its short-lived products are also hazardous to the respiratory organs. The internal exposure to radon and its daughter products is quantified by the internal hazard index (H_{in}) which was given by the following equation

$$H_{in} = \frac{1}{185}A_{Ra} + \frac{1}{259}A_{Th} + \frac{1}{4810}A_K \quad (5)$$

Representative level index ($I_{\gamma r}$)

Radiation hazards due to the specified radionuclides of ^{226}Ra, ^{232}Th and ^{40}K were assessed by another index called representative level index, $I_{\gamma r}$. The following equation was applied to calculate $I_{\gamma r}$ for plant samples under investigation [13].

$$I\gamma r = = \frac{1}{150}A_{Ra} + \frac{1}{100}A_{Th} + \frac{1}{1500}A_K \quad (6)$$

Absorbed Dose Rate (D_R)

The absorbed dose rate D_R (nGy/h) in air at a height of about 1m above the ground surface for uniform distribution of radionuclides were calculated using the following equation [1]:

$$D_R\ (nGy/h) = 0.472A_{Ra} + 0.662A_{Th} + 0.0432\ A_K \quad (7)$$

The absorbed dose rate expresses the received dose in the open air from the radiation emitted from radionuclides concentration in environmental material. Also, it is the first major step for evaluation of the health risk.

It is well known that the acceptable total absorbed dose rate due to gamma radiations and absorbed by the staff operators surrounded by materials containing radionuclides of ^{226}Ra, ^{232}Th and ^{40}K should not exceed the average world value of 59nGy/h [11].

Annual Effective Dose Equivalent (AEDE)

The Annual Effective Dose Equivalent (AEDE) was calculated from the absorbed dose by applying the dose conversion factor of 0.7 Sv/Gy with an outdoor occupancy factor of 0.2 and 0.8 for indoor [1].

$$(AEDE)_{Outdoor}(mSv/y) = D(nGy/h) \times 8760\ (h/y) \times 0.7 \times 10^{-6}\ (mSv/nGy) \times 0.2 \quad (8)$$

$$(AEDE)_{Indoor}(mSv/y) = D(nGy/h) \times 8760\ (h/y) \times 0.7 \times 10^{-6}(mSv/nGy) \times 0.8 \quad (9)$$

To estimate annual effective doses, account must be taken of (a) the conversion coefficient from absorbed dose in air to effective dose and (b) the indoor occupancy factor. Annual estimated average effective dose equivalent received by a member is calculated using a conversion factor of 0.7 Sv/Gy, which is used to convert the absorbed rate to human effective dose equivalent with an outdoor occupancy of 20% and 80% for indoors [11].

RESULTS AND DISCUSSION

As shown in table 3, the activity concentrations of ^{226}Ra from 1.26±0.08 to 32.43±1.71 $Bq{\cdot}kg^{-1}$, of ^{232}Th ranged from 1.20±0.05 to 16.67±1.99 $Bq{\cdot}kg^{-1}$ and of ^{40}K from 397.71±6.02 to 1729.68±24.57 $Bq{\cdot}kg^{-1}$ for all the 13 samples measured in this study. Figures (3) and (4) shows the variations of the activity concentration of ^{226}Ra, ^{232}Th and ^{40}K at different positions, where point 0 refer to P0 and from 2 to 10 refer to P1L (at 200 m from left side of the factory) to P5L (after 1000 m) respectively with step 200 m, and from -2 to -10 refer to P1R (at 200 m from right side of the factory) to P5R (after 1000 m at right side) respectively with step 200 m, and 12 and -12 refer to P6L (after 6000 m at left side) and P6R (after 6000 m at right side) respectively.

In table 4, the resultant activity concentration mean values of (6.50±8.94), (7.80± 5.39) and (729.85± 344.93) Bq/kg are found for ^{226}Ra, ^{232}Th and ^{40}K, respectively in plants samples. The radiological hazard (Ra_{eq}, D_R, $AEDE_{(indoor)}$, $AEDE_{(outdoor)}$, $I_{\gamma r}$, H_{ex} and H_{in}) in the plants samples of the areas under investigation tabulated in table 5.

From table (5), the radium equivalent activity values are lower than the permissible maximum value of 370 Bq/kg [14]. The calculated values of H_{in} and H_{ex} are less than the critical value of unity. The absorbed dose rate, which corresponding population-weight (world average), have a value of 59nGy/h [11]. Position P1L is higher than the permissible maximum value, but for the rest position is lower than it. The annual effective dose equivalent the corresponding world average value is 0.41 mSv/y of which 0.07 mSv/y is from outdoor and 0.34 mSv/y from indoor exposure [15]. Therefore, the study area is still in the zones of normal radiation level except position P1L which are higher than the permissible maximum value (0.46 mSv/y for indoor and 0.11 mSv/y for outdoor).

Table 3. Activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in all plants samples studied.

Sample	Activityconcentrations ± Stat. ErrorBq/Kg		
	Ra-226	Th-232	K-40
P0	32.43±1.71	16.67±1.99	493.15±7.23
P1L	14.77±0.81	15.68±3.29	1729.68±24.57
P2L	2.25±0.15	14.90±0.07	397.71±6.02
P3L	1.89±0.12	9.37±0.91	641.82±8.38
P4L	1.74±0.13	10.96±1.30	539.04±7.33
P5L	1.40±0.09	8.14±0.88	602.71±7.89
P6L	1.36±0.14	6.14±1.00	530.46±3.76
P1R	9.33±0.59	6.28±1.63	939.13±10.69
P2R	10.77±1.40	4.23±0.48	574.00±8.21
P3R	1.87±0.52	3.74±0.72	502.09±6.93
P4R	1.26±0.08	2.87±0.41	853.79±9.69
P5R	2.76±0.18	1.20±0.05	819.61±9.11
P6R	2.72±0.15	1.25±0.08	864.90±10.02

Table 4. Average activity concentration and standard deviations of naturally occurring of ^{226}Ra, ^{232}Th and ^{40}K radionuclides in plant samples of the areas under investigation.

Average activity concentration Bq/kg						
Number of samples	Ra-226		Th-232		K-40	
	Mean	S. D.	Mean	S. D.	Mean	S. D.
13	6.50	8.94	7. 80	5.39	729.85	344.93

Table 5. Average radiological hazard (Ra_{eq}, $I_{\gamma r}$, H_{ex},H_{in},D_R, $AEDE_{(indoor)}$ and $AEDE_{(outdoor)}$) in the plant samples of the areas under investigation.

Sample	Ra_{eq}	$I_{\gamma r}$	H_{ex}	H_{in}	D_R	$AEDE_{(indoor)}$	$AEDE_{(outdoor)}$
	Bq/Kg	Bq/Kg	Bq/Kg	Bq/Kg	nGy/h	mSv/y	mSv/y
P0	94.24	0.712	0.255	0.342	47.65	0.234	0.058
P1L	170.37	1.41	0.460	0.500	92.07	0.452	0.113
P2L	54.18	0.43	0.146	0.152	28.11	0.138	0.034
P3L	64.70	0.53	0.175	0.180	34.82	0.171	0.043
P4L	58.91	0.48	0.159	0.164	31.36	0.154	0.038
P5L	59.46	0.49	0.161	0.164	32.09	0.157	0.039
P6L	50.99	0.42	0.138	0.141	27.62	0.136	0.034
P1R	90.62	0.75	0.245	0.270	49.13	0.241	0.060
P2R	61.03	0.50	0.165	0.194	32.69	0.160	0.040
P3R	45.88	0.38	0.124	0.129	25.05	0.123	0.031
P4R	71.11	0.61	0.192	0.195	39.38	0.193	0.048
P5R	67.58	0.58	0.182	0.190	37.50	0.184	0.046
P6R	71.10	0.61	0.192	0.199	39.47	0.194	0.048
Mean	**73.859**	**0.608**	**0.199**	**0.217**	**39.764**	**0.195**	**0.049**

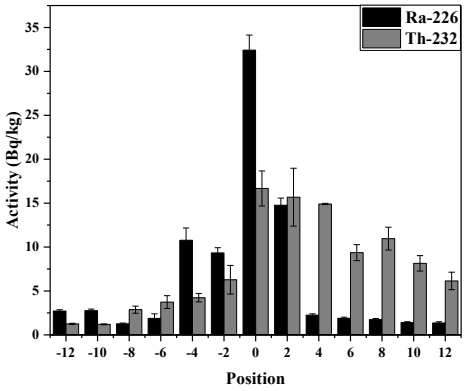

Figure 3: Variation of activity concentration of ^{226}Ra and ^{232}Th at different positions.

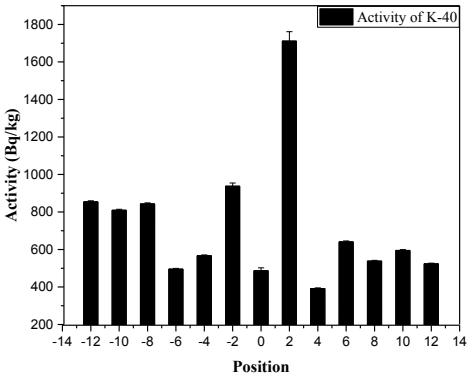

Figure 4: Variation of activity concentration of ^{40}K at different positions.

As shown from figures (3) and (4), it is clear that, the high concentration of the ^{226}Ra, ^{232}Th and ^{40}K were at the center of the factory and it decreases by increasing the distance from the factory.

Table (6) gives a comparison of the activity concentration of ^{226}Ra, ^{232}Th and ^{40}K in the plants from this study with those from other studies in Egypt. And it is clear from the table that the range of this study is higher than the ranges in the other studies except the northwestern area of the Delta which is higher than all the studies. Table 7 shows the activity concentration of ^{226}Ra, ^{232}Th and ^{40}K in the plants from other countries and some of these studies higher than this study and some of them lower it.

Table 6. Comparison of the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in the plants from this study with those from other studies in Egypt.

Studied Area	**Average radionuclide activity in plants (Bq/kg)**		
	Ra-226	**Th-232**	**K-40**
This Study	(1.2-32.4)	(1.2-16.6)	(398-1730)
Burullus lake [16]	(1.0-3.8)	(0.9-3.2)	(233-498)
Burullus lake [17]	<1.5	<4.9	(235-507)
Northeast Sinai [18]	<3.1	(0.6-2.6)	(102-624)
Suez Gulf [19]	(0.8−3.5)	<2.0	(119−443)
Northwestern area of the Delta [20]	41.7	14.1	2416

Table 7. The activity concentration of ^{226}Ra, ^{232}Th and ^{40}K in the plants from other countries.

Studied Area	Average radionuclide activity in plants (Bq/kg)		
	Ra-226	**Th-232**	**K-40**
Ghana [21]	31.8 (20.4–46.9)	56.2 (42.0–70.6)	839 (566–1093)
Brazil [22]	-	21.7 (<11.0–43.0)	976 (666–1216)
Italy [23]	0.4 (<0.1–7.3)	-	654 (5–3582)
Nigeria [24]	15.6 (14.7–16.2)	8.5 (7.0–11.4)	68 (67–70)
Serbia [25]	2.6 (0.6–8.2)	7.4 (1.7– 15.1)	590 (126–1243)

CONCLUSIONS

It is important to determine the background radiation level, in order to evaluate the health hazards. The method of gamma spectrometry had been used to measure the radioactivity concentration of 13 plant samples collected from the surrounding area of Abu Zabal Company for fertilizers and chemical industries in Egypt. The activity concentrations of ^{226}Ra from 1.26±0.08 to 32.43±1.71 Bq·kg^{-1}, of ^{232}Th ranged from 1.20±0.05 to 16.67±1.99 Bq·kg^{-1} and of ^{40}K from 397.71±6.02 to 1729.68±24.57 Bq·kg^{-1} of all the 13 samples measured in this study, with mean activity concentrations are found to be 6.50, 7.80 and 729.85 Bq/kg, respectively. The high concentration of the ^{226}Ra, ^{232}Th and ^{40}K were at the center of the factory and it decreases with increasing the distance from the factory, which mean that the factory effects on the surrounding area and also it is clear from the values of the absorbed dose rate and the annual effective dose equivalent, which exceed the permissible maximum value for them. The value of Raeq activity was found to be less than 370 Bq/kg, the external hazard indices were found to be less than the acceptable limit of unity.

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