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# Assessment of radioactivity levels and radiological-hazard indices in plant fertilizers used in Iraqi Kurdistan Region

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**Abstract**
The present study is to determine the activity concentration of radioactive nuclide in plant fertilizers used in agriculture in Iraqi Kurdistan region using high- purity germanium detector (HPGe) gamma spectrometer. The results showed that the range of activity concentrations for $^{226}$Ra, $^{232}$Th, $^{40}$K and $^{137}$Cs in chemicals and organic fertilizers are (0.1–134), (0.1–74), (1–12,000) and (0–1) Bq/kg respectively, based on the measured activities that were used to assess the radiological hazards. Radium equivalent activity in some samples exceeds the value (370 Bq/kg) which recommended by the OECD.

**Keywords** Radioactivity · Organic fertilizer · Chemical fertilizer · Radiological hazards · HPGe detector

## Introduction

In latest years, agricultural activities have extended widely, resulting in the applications of the different types of fertilizers. Chemical fertilizers are chemical compounds that provide necessary chemical elements and nutrients to the plants. Fertilizers have become important to the agricultural field all over the world [1]. It has been known that phosphate rocks contain high concentrations of uranium, thorium, radium and their decay products [2]. Phosphate rock is a main raw material used for manufacturing different types of phosphate fertilizers. Therefore, when this rock is processed into phosphates' fertilizers, most of the radioactive nuclides come into the fertilizers. It has been estimated that phosphate fertilizers that applied to the fields could increase the radioactivity level in soils [3].

Fertilizers reorganize naturally occurring radioactive nuclides at trace levels through the environment and become a source of radiation. This phenomenon may result in potential radiological-hazard due to possible migration of elements of the plant fertilizers to soil and plants, and via the food chain, to human beings where this may lead to internal exposure through ingestion of food grown on fertilized soils [4].

The abnormal content of uranium, thorium and its decay products in ore rocks and fertilizer are the main sources of high radioactivity background parts that have been recognized in several areas of the world [5]. From the point of view of the international atomic energy agency of radioactive nuclides in food and the environment, it is necessary to measure the natural environmental radiation levels provided by ground, air, water, fertilizer etc. and to assess the dose limits of exposure to radiation from these naturally occurring radioactive nuclides [6, 7]. Human activities which involve the presence of natural radiation sources that lead to a significant increase in the exposure of workers, Regulation (EU) No 305/2011 requires information to be made available when products are placed on the market, and the current annual effective dose limits for occupational and public exposure should be maintained [8].

The aims of the present study are to determine the content of radioactivity in fifty different types of plant fertilizers that were commonly used in agriculture in Iraqi Kurdistan Region and to estimate their radiological risk to farmers and workers working in the a fertilizer factories and stores of the long-term exposure due to their application. Comparisons of the presented results were obtained with those of national and the world averages.

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## Experimental

To determine the activity concentration of radioactive nuclides, fifty samples of plant fertilizers were collected from markets and companies in Iraqi Kurdistan Region. Properties of these samples were listed in Table 1. Kurdistan is located in the Middle East; it is situated in the north of Iraq, southeast of Turkey, northwest of Iran, and northeast of Syria, as shown in Fig. 1.

The samples were crushed and meshed perfectly to pass through 0.2 mm mesh. One kilogram from each sample was packed in a Marinelli beaker (one-liter size) for gamma spectrometry and sealed for 6 weeks to reach secular equilibrium between the radium contents of the sample and their daughter radionuclides [9, 10].

Each sample from fertilizers was subjected to a gamma-ray spectrometer with HPGe detector setup and the multi-channel analyzer. The high- purity germanium detector is p-type of vertical closed-end coaxial, manufactured by PGT (Princeton Gamma Tec-PGT Company-USA) with the following specifications: Crystal diameter is 70.6 mm, crystal length 70.7 mm, resolution (FWHM) at 122 keV of $^{57}$Co is 1.18 keV and at 1332 keV of $^{60}$Co is 1.97 keV with relative efficiency 73.8% for this energy. The instrument calibrated for energy by using the standard point gamma-ray sources, such as $^{60}$Co (peaks 1173.2 and 1332.5 keV), $^{137}$Cs (peak 661.7 keV) and $^{226}$Ra (peaks 186.1, 295, 351.9, 609, 665, 1120 and 1764 keV). The efficiency calibration was achieved using the same three standard sources, and the relative efficiency curve was normalized to absolute volume efficiency curve, using two radioactive samples prepared [potassium chloride KCl powder and of Uranylace ($UO_2(OCOCH_3)_2{\cdot}2H_2O$) solution] to satisfy the geometrical conditions of the tested samples.

The detector was placed in a wall of lead with thickness (10 cm) to shield the measuring station against background radioactivity. The samples were placed over the detector for at least 10 h. The spectra were evaluated by Thermo Scientific System 8000 multi-channel analyzer and the computer software program (Quantum Gold 2001 for PGT Company). In order to determine the background distribution in the environment around the detector, an empty sealed beaker was counted in the same manner and in the same geometry as the samples. The measurement time of activity or background was 10 h. (The background spectra were used to correct the net peak area of gamma rays of measured isotopes). After measurement and subtraction of the background, the activity concentration was calculated from following gamma-ray photopeak lines [11, 12].

(a) $^{226}$Ra activity concentration was calculated as the weighted average of the activity determined, using the gamma-ray lines 351.9 (35.8%) keV gamma-rays from $^{214}$Pb decay, 609.3 (44.8%), 1120 (14.8%) and 1764.5 keV (15.36%) gamma-rays from $^{214}$Bi decay.

(b) The gamma-ray photo peaks used for the determination the activity concentration of the $^{232}$Th were 238.6 keV (43%) from $^{212}$Pb decay, 583 keV (84.5%) and 2614.5 keV (99.16%) from $^{208}$Tl decay and 911.2 keV (26.6%) from $^{228}$Ac decay.

(c) The activity concentration of $^{40}$K was directly determined using 1460.8 (10.7%) gamma-rays line.

(d) The activity concentration of $^{137}$Cs was directly determined using 661.7 (85.21%) gamma-rays line.

## Theory

### Activity concentration

Activity concentration of the isotopes calculated using the following formula [13, 14]:

$$\text{Activity concentration} = [\text{Net count}/(\varepsilon \times I_\gamma \times T \times M)) \pm (\text{SD}/(\varepsilon \times I_\gamma \times T \times M)] \quad (1)$$

where $\varepsilon$ is the absolute gamma peak efficiency of the detector to its particular gamma-ray energy, $I_\gamma$ is the decay intensity of the specific energy peak (including the decay branching ratio information), $T$ is the counting time of the measurement in second, $M$ is the mass of the sample in kg and SD is the standard deviation of the net count rate per second.

### Radium equivalent activity ($Ra_{eq}$)

The total exposure to radiation from these naturally occurring radioactive nuclides was defined in terms of radium equivalent activity ($Ra_{eq}$) in (Bq/kg) and used to assess the gamma-ray radiation hazards due to specified radionuclides $^{226}$Ra, $^{232}$Th and $^{40}$K. The $Ra_{eq}$ of a sample in (Bq/kg) can be achieved using the following relation [15, 16]:

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

where $A_{Ra}$, $A_{Th}$, $A_K$ are the activity concentrations of $^{226}$Ra, $^{232}$Th and $^{40}$K in a unit (Bq/kg) respectively. The published maximal permissible $Ra_{eq}$ is 370 Bq/kg [17, 18].

### The absorbed gamma dose rates ($D_R$)

The absorbed gamma dose rates in air at 1 m above the ground surface for the uniform distribution of naturally occurring radioactive nuclides were calculated based on guidelines provided by [19]:

**Table 1** Type, a chemical component and form of plant fertilizer samples under study

| No. | Code of fertilizer samples | Type of fertilizer | Chemical components of fertilizer | | | Form of fertilizer | Made in |
|---|---|---|---|---|---|---|---|
| | | | N (%) | $P_2O_5$ (%) | $K_2O$ (%) | | |
| 1 | K 1 | Potassium sulfate | 0 | 0 | 52 | Powder | Belgium |
| 2 | K 2 | Potassium nitrate | 0 | 0 | 12 | Foliar spray | Turkey |
| 3 | N 1 | Urea | 46 | 0 | 0 | Granular | Russia |
| 4 | N 2 | Urea | 46 | 0 | 0 | Granular | Iran |
| 5 | N + Ca (NCa 1) | Calcium nitrate | 13 | 0 | 0 | Powder | Belgium |
| 6 | N + Ca (NCa 2) | Calcium nitrate | 13.5 | 0 | 0 | Powder | Belgium |
| 7 | NP 1 | Urea ammonium phosphate | 20 | 20 | 0 | Granular | Russia |
| 8 | NP 2 | Diammonium phosphate | 18 | 46 | 0 | Granular | Turkey |
| 9 | NP 3 | Diammonium phosphate | 18 | 46 | 0 | Granular | Jordan |
| 10 | NP 4 | Diammonium phosphate | 18 | 46 | 0 | Granular | Tunisia |
| 11 | NP 5 | Urea ammonium phosphate | 18 | 44 | 0 | Powder | Belgium |
| 12 | NP 6 | Urea ammonium phosphate | 18 | 44 | 0 | Powder | Belgium |
| 13 | NP 7 | Urea ammonium phosphate | 17 | 44 | 0 | Powder | Netherlands |
| 14 | NP 8 | Mono ammonium phosphate | 17 | 62 | 0 | Powder | Netherlands |
| 15 | NPK 1 | N.P.K. complex | 12 | 6 | 18 | Granular | Italy |
| 16 | NPK 2 | N.P.K. complex | 18 | 18 | 5 | Granular | UAE |
| 17 | NPK 3 | N.P.K. complex | 15 | 30 | 10 | Powder | Netherlands |
| 18 | NPK 4 | N.P.K. complex | 20 | 5 | 5 | Powder | Spain |
| 19 | NPK 5 | N.P.K. complex | 13 | 2 | 44 | Powder | Belgium |
| 20 | NPK 6 | N.P.K. complex | 12 | 8 | 40 | Powder | Belgium |
| 21 | NPK 7 | N.P.K. complex | 15 | 8 | 26 | Powder | Belgium |
| 22 | NPK 8 | N.P.K. complex | 20 | 20 | 20 | Powder | Belgium |
| 23 | NPK 9 | N.P.K. complex | 20 | 20 | 20 | Powder | Belgium |
| 24 | NPK 10 | N.P.K. complex | 12 | 8 | 16 | Powder | Germany |
| 25 | NPK 11 | N.P.K. complex | 20 | 20 | 20 | Powder | Germany |
| 26 | NPK 12 | N.P.K. complex | 12 | 8 | 40 | Powder | Turkey |
| 27 | NPK 13 | N.P.K. complex | 20 | 20 | 20 | Powder | Turkey |
| 28 | NPK 14 | N.P.K. complex | 15 | 30 | 15 | Powder | Turkey |
| 29 | NPK 15 | N.P.K. complex | 10 | 5 | 30 | Powder | Jordan |
| 30 | NPK 16 | N.P.K. complex | 18 | 18 | 18 | Powder | Jordan |
| 31 | NPK 17 | N.P.K. complex | 12 | 47 | 10 | Powder | Israel |
| 32 | NPK 18 | N.P.K. complex | 12 | 6 | 40 | Powder | Israel |
| 33 | NPK 19 | N.P.K. complex | 20 | 20 | 20 | Powder | Israel |
| 34 | NPK 20 | N.P.K. complex | 16 | 8 | 32 | Foliar spray | UAE |
| 35 | NPK 21 | N.P.K. complex | 13 | 11 | 11 | Foliar spray | UAE |
| 36 | NPK 22 | N.P.K. complex | 10 | 52 | 8 | Foliar spray | UAE |
| 37 | NPK 23 | N.P.K. complex | 30 | 10 | 10 | Foliar spray | USA |
| 38 | NPK 24 | N.P.K. complex | 10 | 20 | 30 | Foliar spray | USA |
| 39 | NPK 25 | N.P.K. complex | 10 | 52 | 10 | Foliar spray | USA |
| 40 | PK (L 1) | Potassium phosphate | 0 | 25 | 28 | Liquid | USA |
| 41 | K (L 2) | Potassium nitrate | 0 | 0 | 26 | Liquid | Spain |
| 42 | N (L 3) | Urea ammonium nitrate | 31 | 0 | 0 | Liquid | KSA |
| 43 | P (L 4) | Super phosphate | 0 | 62 | 0 | Liquid | KSA |
| 44 | Organic + NPK (L5) | Organic + complex | 7 | 21 | 0 | Liquid | UAE |
| 45 | Organic + NPK (OR1) | Organic + complex | 6 | 12 | 10 | Powder | Spain |
| 46 | Organic (OR 2) | Organic | 2.03 | 0.52 | 0.43 | Granular | Germany |
| 47 | Organic (OR 3) | Organic | 1.12 | 0.61 | 0.61 | Granular | Turkey |

**Table 1** (continued)

| No. | Code of fertilizer samples | Type of fertilizer | Chemical components of fertilizer | | | Form of fertilizer | Made in |
|---|---|---|---|---|---|---|---|
| | | | N (%) | $P_2O_5$ (%) | $K_2O$ (%) | | |
| 48 | Organic (OR 4) | Organic | 4 | 0.11 | 0.55 | Powder | Turkey |
| 49 | Organic (OR 5) | Organic | 3.29 | 0.5 | 0.59 | Powder | Iraq |
| 50 | Organic (OR 6) | Organic | 1.82 | 0.3 | 0.52 | Powder | Iraq |


**Fig. 1** Map of Iraqi Kurdistan Region

$$D_R(\text{nGy/h}) = (A_{Ra} \times 0.462) + (A_{Th} \times 0.604) + (A_K \times 0.0417) \quad (3)$$

## Annual effective dose (AED)

In order to estimate the annual effective dose rate in the air, the conversion coefficient from the absorbed dose in the air to the effective dose received by an adult must be considered. Annual estimated average effective dose (AED) received by an individual was calculated using a conversion factor of 0.7 Sv/Gy, which was used to convert the absorbed rate to the human effective dose. The annual effective dose is determined using the following equations [15, 16]:

$$\text{AED} = D_R \times T \times xF \quad (4)$$

where $D_R$ is the calculated dose rate (in nGy/h), $T$ is the occupancy time and $F$ is the conversion factor, this value is published in UNSCEAR 1993 and UNSCEAR 2000 [20, 21], to be 0.7 Sv/Gy for environmental exposure to gamma rays of moderate energy. The outdoor occupancy factor $T$ is about (20% of 8760 h/y). The outdoor annual effective dose is given by the following equation:

$$AED_{\text{outdoor}}(\mu\text{Sv/y}) = D_R(\text{nGy/h}) \times (0.2 \times 8760\ \text{h/y}) \times 0.7\ (\text{Sv/Gy}) \quad (5)$$

The world average annual effective dose (AED) from outdoor or indoor terrestrial gamma radiation only is 70 μSv/y [21].

## External hazard index ($H_{ex}$)

The external hazard index is an evaluation of the hazard caused by gamma-rays emitted from natural radioactive nuclides. It was calculated by assuming that 370 Bq/kg of $^{226}$Ra or 259 Bq/kg of $^{232}$Th or 4810 Bq/kg of $^{40}$K produces the same gamma-ray dose rate. The prime objective

of this index is to limit the radiation dose to the admissible permissible dose equivalent limit around 1 mSv/y. In order to evaluate this index, one can use the following relation [15, 18]:

$$H_{ex} = (A_{Ra}/370) + (A_{Th}/259) + (A_K/4810) \leq 1 \quad (6)$$

This model takes into consideration the maximum value of external hazard which is caused by gamma-rays corresponds to a radium equivalent activity of 370 Bq/kg [17, 18].

### Excess lifetime cancer risk (ELCR)

Excess lifetime cancer risk measures the additional cancer risk induced by exposure to ionizing radiations. Based on the calculated values of AED, ELCR is calculated using the equation [22, 23]:

$$ELCR_{outdoor} = AED_{outdoor}(\mu Sv/y) \times DL \times RF \quad (7)$$

where AED is the annual effective dose, DL is the average duration of life which is 70 year, RF is the risk factor given as 0.05 by ICRP 1991 [24].

## Results and discussion

The activity concentrations of $^{226}Ra$, $^{232}Th$, $^{40}K$, and $^{137}Cs$ in different fertilizer samples were measured and listed in Table 2. The results showed that the mean value and a range of activity concentrations of $^{226}Ra$, $^{232}Th$, $^{40}K$ and $^{137}Cs$ in these fertilizers are $7 \pm 1$ (0.1–134), $4 \pm 1$ (0.1–74), $2500 \pm 100$ (1–12,000) and $0.1 \pm 0.01$ Bq/kg (0–1) Bq/kg respectively. The highest value of activity concentrations of $^{226}Ra$ and $^{232}Th$ are found in NPK fertilizer, while the lowest values were found in urea and calcium fertilizers. Except for fertilizer samples (NPK1, NPK2, NPK3, OR3, and OR4), the measured value of activity concentrations of $^{226}Ra$ and $^{232}Th$ are lower than the word wide average value of 35 and 30 Bq/kg for $^{226}Ra$ and $^{232}Th$ respectivily [21]. The variation of radionuclides activities in studying fertilizers may be due to the different origins of raw material and the chemical processes during manufacturing of the fertilizer.

As shown in Fig. 2, the activity concentration of $^{226}Ra$ in most studied fertilizer samples are higher than the activity concentration of $^{232}Th$ in these samples, which may be due to the accumulation of dissolved uranium and its products, in the form of urinal complex in the seawater during geological formation of the phosphate rocks [25].

Figures 3 and 4 shows the relationships between activity concentrations of $^{226}Ra$ and $^{232}Th$ with concentration of $P_2O_5$ in fertilizer samples. It is clear that there is no linear relationship between activity concentrations of $^{226}Ra$ and $^{232}Th$ and concentration of $P_2O_5$ in fertilizer samples, and the variation of radionuclides activities in studying fertilizers due to different origins of raw material and the chemical processes during the manufacturing of the fertilizer.

The highest value of activity concentration $^{40}K$ was found in potassium sulfate fertilizer, while the lowest value was found in urea and calcium fertilizers. This is due to high concentration of $K_2O$ in potassium sulfate fertilizer, because there is a good relationship between concentration of $K_2O$ and $^{40}K$ radioactive isotope content, and the presence of radioactive $^{40}K$, whose natural abundance in natural potassium K is 0.0118%, and this relation was found in Fig. 5. The activity concentration of $^{40}K$ in potassium sulfate and NPKs fertilizer samples was higher than the word wide average value for $^{40}K$ which is 400 Bq/kg [21]. While the activity concentration of $^{40}K$ in Urea, Calcium, NP, and organic samples, was lower than the word wide average value. The activity concentration of $^{137}Cs$ was found only in organic fertilizer samples, this may be due to the production process of the organic fertilizers. The results for the activity concentration of $^{226}Ra$, $^{232}Th$, $^{40}K$, and $^{137}Cs$, of the present work, compared with other studies are presented in Table 3.

In order to compare the activity concentrations of $^{226}Ra$, $^{232}Th$ and $^{40}K$ in fertilizer samples, the radium equivalent activity ($Ra_{eq}$) as a common index was used to obtain the sum of activities. To estimate the radiological risk to farmers and workers working in a fertilizer factory of the long-term exposure due to their application, the external absorbed dose rate ($D$), annual effective dose (AED), external hazard index ($H_{ex}$) and excess lifetime cancer risk (ELCR) for different types of fertilizer samples were calculated and listed in Table 4.

The value of radium equivalent activity ($Ra_{eq}$) in the fertilizer samples varied from (0.4 to 900) Bq/kg, the variation of radium equivalent activity ($Ra_{eq}$) in studying fertilizers, due to the different amount content of $^{226}Ra$, $^{232}Th$ and $^{40}K$ in fertilizers. The calculated value of radium equivalent ($Ra_{eq}$), except for fertilizer samples (K1, NPK1, NPK5, NPK6, NPK7 NPK12, NPK13, NPK15, NPK18 NPK20, NPK24 and L1), were lower than the recommended level of 370 Bq/kg [17, 18, 31].

The calculated value of the external absorbed dose rate ($D_R$) of the fertilizer samples varied from (0.2 to 480) nGy/h. The value of absorbed dose rate ($D_R$) in Potassium Sulfate, Potassium Nitrate and NPK complex fertilizers were higher than the worldwide average value, while the value of absorbed dose rate ($D_R$) in urea, diammonium phosphate, urea ammonium phosphate, calcium nitrate and organic were lower than the worldwide average value of 59 nGy/h [21].

**Table 2** The activity concentrations of $^{226}Ra$, $^{232}Th$, $^{40}K$ and $^{137}Cs$ in different types of fertilizer samples

| No. | Code of fertilizer samples | Activity concentrations of $^{226}Ra$ (Bq/kg) | Activity concentrations of $^{232}Th$ (Bq/kg) | Activity concentrations of $^{40}K$ (Bq/kg) | Activity concentrations of $^{137}Cs$ (Bq/kg) |
|---|---|---|---|---|---|
| 1 | K 1 | 1 ± 0.2 | 0.2 ± 0.02 | 12,000 ± 600 | BDA |
| 2 | K 2 | 16 ± 1 | 16 ± 1 | 1200 ± 60 | BDA |
| 3 | N 1 | 0.2 ± 0.04 | 0.2 ± 0.03 | 0.4 ± 0.03 | BDA |
| 4 | N 2 | 0.1 ± 0.03 | 0.1 ± 0.02 | 1 ± 0.3 | BDA |
| 5 | N + Ca (NCa 1) | 2 ± 0.1 | 0.2 ± 0.03 | 2 ± 0.3 | BDA |
| 6 | N + Ca (NCa 2) | 2 ± 0.2 | 0.2 ± 0.05 | 1 ± 0.2 | BDA |
| 7 | NP 1 | 0.6 ± 0.1 | 4 ± 0.2 | 53 ± 2 | BDA |
| 8 | NP 2 | 13 ± 1 | 3 ± 0.2 | 60 ± 3 | BDA |
| 9 | NP 3 | 8 ± 1 | 1.5 ± 0.1 | 14 ± 1 | BDA |
| 10 | NP 4 | 6 ± 1 | 12 ± 1 | 20 ± 1 | BDA |
| 11 | NP 5 | 0.2 ± 0.06 | 5 ± 0.2 | 2 ± 0.5 | BDA |
| 12 | NP 6 | 0.4 ± 0.1 | 10 ± 1 | 4 ± 0.5 | BDA |
| 13 | NP 7 | 0.2 ± 0.04 | 0.2 ± 0.03 | 1 ± 0.2 | BDA |
| 14 | NP 8 | 0.4 ± 0.1 | 0.2 ± 0.04 | 13 ± 2 | BDA |
| 15 | NPK 1 | 134 ± 6 | 1 ± 0.2 | 4000 ± 200 | BDA |
| 16 | NPK 2 | 13 ± 1 | 74 ± 3 | 1000 ± 50 | BDA |
| 17 | NPK 3 | 0.6 ± 0.2 | 0.2 ± 0.03 | 2200 ± 100 | BDA |
| 18 | NPK 4 | 0.3 ± 0.1 | 1 ± 0.3 | 1000 ± 50 | BDA |
| 19 | NPK 5 | 0.1 ± 0.01 | 0.8 ± 0.2 | 10,000 ± 500 | BDA |
| 20 | NPK 6 | 1 ± 0.2 | 1 ± 0.3 | 7000 ± 500 | BDA |
| 21 | NPK 7 | 1 ± 0.04 | 0.2 ± 0.04 | 5000 ± 250 | BDA |
| 22 | NPK 8 | 0.5 ± 0.2 | 1 ± 0.3 | 2400 ± 120 | BDA |
| 23 | NPK 9 | 0.3 ± 0.03 | 0.1 ± 0.02 | 3800 ± 200 | BDA |
| 24 | NPK 10 | 16 ± 1 | 15 ± 1 | 2400 ± 130 | BDA |
| 25 | NPK 11 | 0.6 ± 0.1 | 0.1 ± 0.02 | 3500 ± 180 | BDA |
| 26 | NPK 12 | 1 ± 0.3 | 0.2 ± 0.03 | 7000 ± 400 | BDA |
| 27 | NPK 13 | 1 ± 0.3 | 0.1 ± 0.02 | 6000 ± 300 | BDA |
| 28 | NPK 14 | 1 ± 0.01 | 0.3 ± 0.03 | 3400 ± 200 | BDA |
| 29 | NPK 15 | 1 ± 0.2 | 1 ± 0.4 | 5300 ± 300 | BDA |
| 30 | NPK 16 | 1 ± 0.2 | 0.2 ± 0.03 | 3200 ± 200 | BDA |
| 31 | NPK 17 | 0.6 ± 0.1 | 0.4 ± 0.1 | 2400 ± 100 | BDA |
| 32 | NPK 18 | 1 ± 0.2 | 0.3 ± 0.08 | 7000 ± 400 | BDA |
| 33 | NPK 19 | 1 ± 0.3 | 0.2 ± 0.04 | 4000 ± 200 | BDA |
| 34 | NPK 20 | 0.2 ± 0.03 | 0.1 ± 0.01 | 5000 ± 300 | BDA |
| 35 | NPK 21 | 0.3 ± 0.09 | 0.3 ± 0.08 | 2000 ± 100 | BDA |
| 36 | NPK 22 | 0.6 ± 0.1 | 0.1 ± 0.03 | 1800 ± 100 | BDA |
| 37 | NPK 23 | 1 ± 0.1 | 0.2 ± 0.05 | 2000 ± 100 | BDA |
| 38 | NPK 24 | 1 ± 0.2 | 0.3 ± 0.02 | 6200 ± 300 | BDA |
| 39 | NPK 25 | 1 ± 0.3 | 0.5 ± 0.1 | 2000 ± 30 | BDA |
| 40 | PK (L 1) | 0.6 ± 0.2 | 0.2 ± 0.03 | 6400 ± 300 | BDA |
| 41 | K (L 2) | 0.5 ± 0.1 | 0.1 ± 0.03 | 2100 ± 100 | BDA |
| 42 | N (L 3) | 1 ± 0.1 | 0.4 ± 0.06 | 21 ± 2 | BDA |
| 43 | P (L 4) | 1 ± 0.1 | 3 ± 0.1 | 8 ± 1 | BDA |
| 44 | Organic + NPK (L5) | 0.4 ± 0.1 | 0.2 ± 0.03 | 9 ± 1 | 0.2 ± 0.03 |
| 45 | Organic + NPK(OR1) | 7 ± 1 | 6 ± 0.2 | 1000 ± 100 | 0.3 ± 0.05 |
| 46 | Organic (OR 2) | 12 ± 1 | 6 ± 0.3 | 100 ± 5 | BDA |
| 47 | Organic (OR 3) | 42 ± 1 | 11 ± 1 | 600 ± 30 | 0.1 ± 0.04 |
| 48 | Organic (OR 4) | 38 ± 1 | 25 ± 1 | 1700 ± 100 | BDA |

**Table 2** (continued)

| No. | Code of fertilizer samples | Activity concentrations of $^{226}$Ra (Bq/kg) | Activity concentrations of $^{232}$Th (Bq/kg) | Activity concentrations of $^{40}$K (Bq/kg) | Activity concentrations of $^{137}$Cs (Bq/kg) |
|---|---|---|---|---|---|
| 49 | Organic (OR 5) | 2 ± 0.3 | 0.5 ± 0.1 | 300 ± 30 | BDA |
| 50 | Organic (OR 6) | 4 ± 0.4 | 2 ± 0.3 | 250 ± 30 | 1 ± 0.2 |
| Over all | Range | 0.1–134 | 0.1–74 | 1–12,000 | 0–1 |
| | Average | 7 | 4 | 2500 | 0.1 |

**BDA* below minimum detectable activity

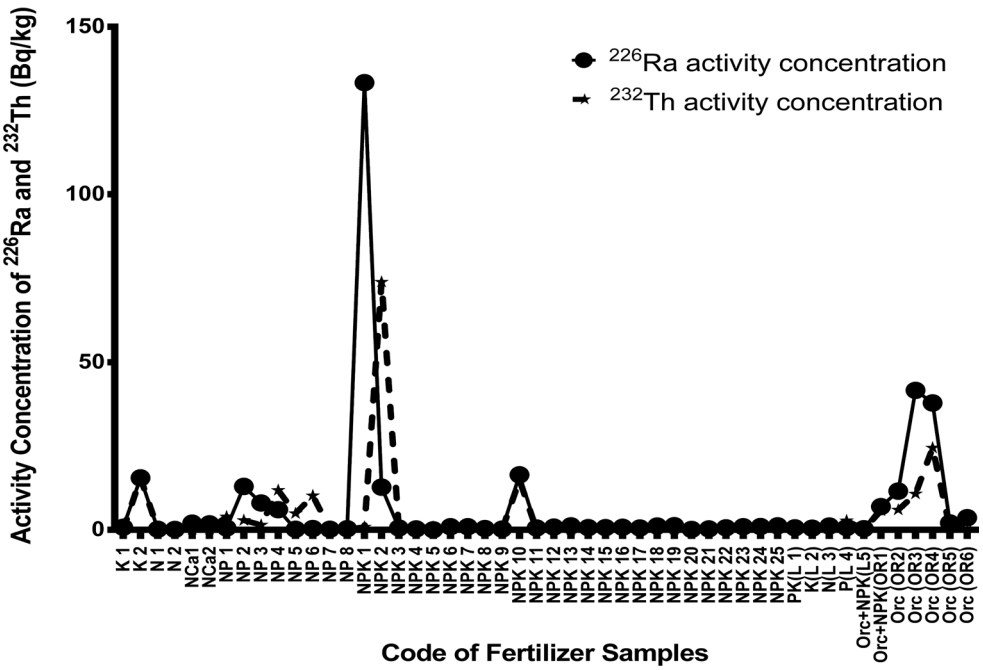


**Fig. 2** Activity concentrations of $^{226}$Ra and $^{232}$Th in fertilizer samples

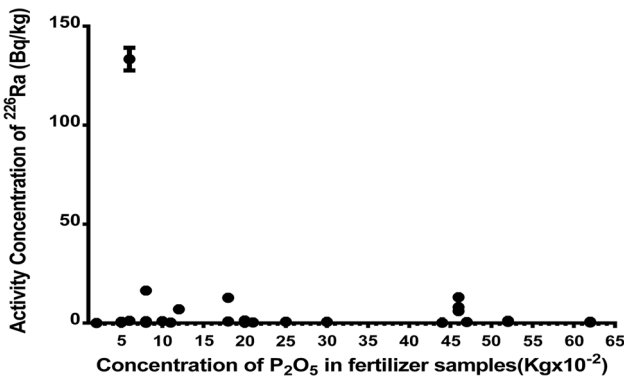


**Fig. 3** Relationship between activity concentration of $^{226}$Ra and concentration of $P_2O_5$ in fertilizer samples

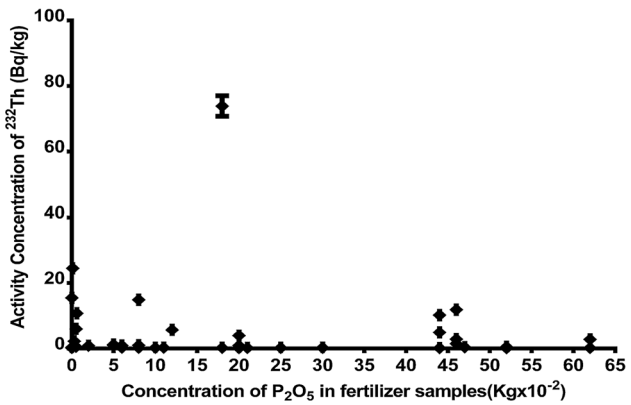


**Fig. 4** Relationship between activity concentration of $^{232}$Th and concentration of $P_2O_5$ in fertilizer samples

The calculated values of outdoor annual effective dose (AED) in the fertilizer samples varied from (0.2 to 590) μSv/y. The calculated value of annual effective dose in potassium sulfate, potassium nitrate and NPK complex fertilizers were higher than the worldwide average value, while the measured value of in urea, diammonium

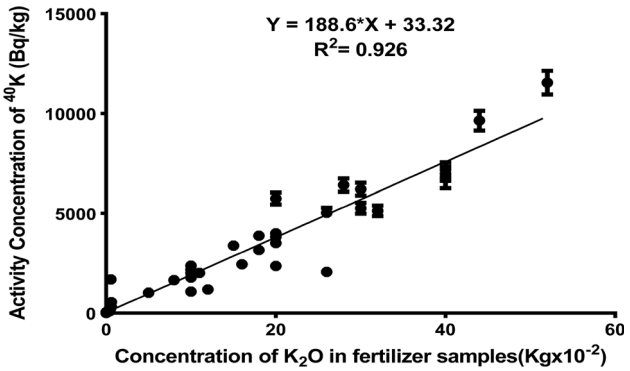


**Fig. 5** Relationship between activity concentration of $^{40}K$ and concentration of $K_2O$ in different fertilizer samples

phosphate, urea ammonium phosphate, calcium nitrate and organic were lower than the worldwide average value of 70 μSv/y [21].

The calculated value of external hazard index ($H_{ex}$) in the fertilizer samples varied from (0 to 2.4) mSv/y, the data indicated that the value of external hazard index in fertilizer samples (K1, NPK1, NPK5, NPK6, NPK12, NPK13, NPK18, NPK24, L1) were higher than the safety limits (1 mSv/y) [14, 15, 17]. While the value of external hazard index in other fertilizer samples within the safety limit. Thus, the use of chemical fertilizers (K1, NPK1, NPK5, NPK6, NPK12, NPK13, NPK18, NPK24, L1) in large extent can be harmful to the health of farmers and workers working in a fertilizer factory.

The calculated value of excess lifetime cancer risk (ELCR) in the fertilizer samples varied from (0 to $20 \times 10^{-4}$)/person. The results of the present study indicated that the use of plant fertilizers to enhance crop yield enhances the concentration of natural radioactive nuclides in soil and hence the exposure of farmers working in the fields.

No linear relationships have been found between external hazard index ($H_{ex}$) with $^{226}Ra$ and $^{232}Th$ activity concentrations, but present result in Fig. 6 shows a good correlation between external hazard index ($H_{ex}$) and $^{40}K$ activity concentration with a correlation coefficient of ($R^2 = 0.98$), this means that most of the hazard caused by nuclear radiation from fertilizer samples were coming from $^{40}K$ radioactive nuclide (or by the high concentration of $^{40}K$ radioactive nuclei in the fertilizer samples).

## Conclusions

The activity concentration of radioactive nuclide and related radiation hazards in plant fertilizer samples being used in Iraqi Kurdistan Region were determined by using a HPGe gamma spectrometer.

Except for fertilizer samples (NPK1, NPK2, NPK3, OR3, and OR4), the measured value of activity concentrations of $^{226}Ra$ and $^{232}Th$ are lower than the word wide average value. The activity concentration of $^{40}K$ in potassium sulphate and NPKs fertilizer samples was higher than the world average value, while its activity in urea, calcium, NP and organic samples was lower than the world average value. No relations have been found between activity concentrations of $^{226}Ra$ and $^{232}Th$ with the concentration of $P_2O_5$ in fertilizer samples. The use of potassium sulphate and NPKs fertilizer made the calculated value of external

**Table 3** Comparison of the obtained values of activity concentrations of $^{226}Ra$, $^{232}Th$, $^{40}K$ and $^{137}Cs$ in fertilizer samples with data from similar studies in the different country of the world

| Country | $^{226}Ra$ | | $^{232}Th$ | | $^{40}K$ | | $^{137}Cs$ | | References |
|---|---|---|---|---|---|---|---|---|---|
| Range | Min. | Max. | Min. | Max. | Min. | Max. | Min. | Max. | |
| Activity concentrations (Bq/kg) of $^{226}Ra$, $^{232}Th$, $^{40}K$ and $^{137}Cs$ in chemical fertilizer samples | | | | | | | | | |
| Algeria | 134 ± 24 | 190 ± 30 | 117 ± 10 | 131 ± 16 | 5312 ± 249 | 11,645 ± 550 | 7.5 ± 0.3 | 11.7 ± 0.6 | [26] |
| Nigeria | 10 ± 7 | 451 ± 14 | BDA | 15 ± 3 | 3972 ± 417 | 5089 + 111 | | | [27] |
| Egypt | 3 | 283 | 2 | 74 | 9 | 6501 | | | [25] |
| KSA | 29 | 121 | 3 | 57 | 227 | 4227 | | | [28] |
| Iraq | 13 ± 1 | 89 ± 3 | 1 ± 0.2 | 27 ± 1 | 12 ± 1 | 2276 ± 18 | | | [10] |
| Iraq | 0.1 ± 0.01 | 134 ± 6 | 0.1 ± 0.02 | 74 ± 3 | 1 ± 0.2 | 12,000 ± 600 | 0 | 0.3 ± 0.05 | Present study |
| Activity concentrations (Bq/kg) of $^{226}Ra$, $^{232}Th$, $^{40}K$ and $^{137}Cs$ in organic fertilizer samples | | | | | | | | | |
| Nigeria | 13 ± 0.01 | 66 ± 1 | 15 ± 0.01 | 60 ± 1 | 120 ± 1 | 908 ± 1 | | | [29] |
| KSA | 21 | 70 | 3 | 21 | 115 | 523 | | | [28] |
| Iraq | 12 ± 5 | 69 ± 7 | 11 ± 2 | 52 ± 5 | 165 ± 7 | 3593 ± 34 | | | [30] |
| Iraq | 2 ± 0.3 | 42 ± 1 | 0.5 ± 0.1 | 25 ± 1 | 100 ± 5 | 1700 ± 100 | 0 | 1 ± 0.2 | Present study |

*BDA* below minimum detectable activity

**Table 4** Radium equivalent ($Ra_{eq}$), External Hazard Index ($H_{ex}$), External Doses Rate ($D_R$), Annual Effective Dose (AED) and Excess lifetime cancer risk (ELCR) for different types of fertilizer samples

| No. | Code of samples | $Ra_{eq}$ (Bq/kg) | $D_R$ (nGy./h) | AED (μSv./y) | $H_{ex}$ (mSv/y) | ELCR × $10^{-4}$ (1/person) |
|---|---|---|---|---|---|---|
| 1 | K 1 | 900 ± 50 | 480 ± 25 | 590 ± 30 | 2.4 ± 0.1 | 20 ± 1 |
| 2 | K 2 | 130 ± 10 | 70 ± 5 | 80 ± 5 | 0.4 ± 0.02 | 3 ± 0.1 |
| 3 | N 1 | 0.4 ± 0.1 | 0.2 ± 0.04 | 0.2 ± 0.05 | 0 | 0 |
| 4 | N 2 | 0.4 ± 0.08 | 0.2 ± 0.04 | 0.2 ± 0.05 | 0 | 0 |
| 5 | N + Ca (NCa1) | 2.4 ± 0.2 | 1 ± 0.1 | 1 ± 0.1 | 0 | 0 |
| 6 | N + Ca (NCa2) | 2 ± 0.4 | 1 ± 0.2 | 1 ± 0.2 | 0 | 0 |
| 7 | NP 1 | 10 ± 1 | 5 ± 0.2 | 6 ± 0.3 | 0 | 0.2 ± 0.01 |
| 8 | NP 2 | 22 ± 1 | 10 ± 1 | 13 ± 1 | 0.06 ± 0.01 | 0.4 ± 0.02 |
| 9 | NP 3 | 11 ± 1 | 5 ± 0.3 | 1 ± 0.4 | 0 | 0.2 ± 0.01 |
| 10 | NP 4 | 25 ± 1 | 11 ± 1 | 13 ± 1 | 0.07 ± 0.01 | 0.4 ± 0.02 |
| 11 | NP 5 | 7 ± 1 | 3 ± 0.2 | 4 ± 0.2 | 0 | 0.1 ± 0.01 |
| 12 | NP 6 | 15 ± 1 | 7 ± 0.2 | 8 ± 0.2 | 0.04 ± 0.01 | 0.3 ± 0.08 |
| 13 | NP 7 | 0.4 ± 0.1 | 0.2 ± 0.05 | 0.3 ± 0.06 | 0 | 0 |
| 14 | NP 8 | 2 ± 0.3 | 1 ± 0.2 | 1 ± 0.2 | 0 | 0 |
| 15 | NPK 1 | 440 ± 20 | 220 ± 11 | 270 ± 10 | 1.2 ± 0.06 | 9 ± 0.4 |
| 16 | NPK 2 | 200 ± 10 | 95 ± 5 | 110 ± 5 | 0.5 ± 0.03 | 4 ± 0.2 |
| 17 | NPK 3 | 170 ± 10 | 90 ± 5 | 110 ± 5 | 0.5 ± 0.02 | 4 ± 0.2 |
| 18 | NPK 4 | 81 ± 5 | 44 ± 2 | 54 ± 5 | 0.2 ± 0.01 | 2 ± 0.1 |
| 19 | NPK 5 | 750 ± 40 | 400 ± 20 | 490 ± 30 | 2 ± 0.1 | 20 ± 1 |
| 20 | NPK 6 | 520 ± 40 | 280 ± 20 | 340 ± 30 | 1.4 ± 0.1 | 10 ± 1 |
| 21 | NPK 7 | 400 ± 20 | 210 ± 11 | 260 ± 10 | 1 ± 0.05 | 9 ± 1 |
| 22 | NPK 8 | 180 ± 10 | 100 ± 5 | 120 ± 20 | 0.5 ± 0.03 | 4 ± 0.2 |
| 23 | NPK 9 | 300 ± 15 | 160 ± 10 | 200 ± 10 | 0.8 ± 0.04 | 7 ± 0.4 |
| 24 | NPK 10 | 220 ± 12 | 120 ± 10 | 150 ± 10 | 0.6 ± 0.03 | 5 ± 0.2 |
| 25 | NPK 11 | 270 ± 14 | 150 ± 10 | 180 ± 10 | 0.7 ± 0.04 | 6 ± 0.3 |
| 26 | NPK 12 | 550 ± 30 | 300 ± 20 | 370 ± 20 | 1.5 ± 0.1 | 10 ± 1 |
| 27 | NPK 13 | 440 ± 25 | 240 ± 10 | 290 ± 20 | 1.2 ± 0.06 | 10 ± 1 |
| 28 | NPK 14 | 260 ± 15 | 140 ± 10 | 170 ± 10 | 0.7 ± 0.04 | 6 ± 0.3 |
| 29 | NPK 15 | 410 ± 20 | 220 ± 10 | 270 ± 10 | 1.1 ± 0.06 | 9 ± 1 |
| 30 | NPK 16 | 250 ± 13 | 130 ± 7 | 160 ± 10 | 0.7 ± 0.03 | 5 ± 0.3 |
| 31 | NPK 17 | 180 ± 10 | 100 ± 5 | 120 ± 10 | 0.5 ± 0.03 | 4 ± 0.2 |
| 32 | NPK 18 | 540 ± 30 | 290 ± 20 | 360 ± 20 | 1.5 ± 0.1 | 10 ± 1 |
| 33 | NPK 19 | 310 ± 20 | 170 ± 10 | 210 ± 10 | 0.8 ± 0.04 | 6 ± 0.4 |
| 34 | NPK 20 | 400 ± 20 | 210 ± 10 | 260 ± 10 | 1.1 ± 0.06 | 9 ± 0.5 |
| 35 | NPK 21 | 160 ± 10 | 80 ± 5 | 100 ± 5 | 0.4 ± 0.02 | 3 ± 0.2 |
| 36 | NPK 22 | 130 ± 10 | 70 ± 5 | 90 ± 5 | 0.4 ± 0.02 | 3 ± 0.2 |
| 37 | NPK 23 | 140 ± 10 | 80 ± 5 | 90 ± 5 | 0.4 ± 0.02 | 3 ± 0.2 |
| 38 | NPK 24 | 480 ± 25 | 260 ± 10 | 320 ± 20 | 1.3 ± 0.07 | 10 ± 1 |
| 39 | NPK 25 | 160 ± 3 | 85 ± 1 | 100 ± 2 | 0.4 ± 0.018 | 3 ± 0.6 |
| 40 | PK (L 1) | 500 ± 30 | 270 ± 15 | 330 ± 20 | 1.3 ± 0.07 | 10 ± 1 |
| 41 | K (L 2) | 160 ± 10 | 90 ± 5 | 110 ± 5 | 0.4 ± 0.02 | 4 ± 0.2 |
| 42 | N (L 3) | 3 ± 0.3 | 2 ± 0.1 | 2 ± 0.2 | 0 | 0 |
| 43 | P (L 4) | 5 ± 0.4 | 2 ± 0.2 | 3 ± 0.2 | 0 | 0 |
| 44 | Organic + NPK (L 5) | 1 ± 0.2 | 0.7 ± 0.1 | 1 ± 0.1 | 0 | 0 |
| 45 | Organic + NPK(OR 1) | 100 ± 5 | 50 ± 3 | 60 ± 5 | 0.3 ± 0.01 | 2 ± 0.1 |
| 46 | Organic (OR 2) | 30 ± 1 | 13 ± 1 | 16 ± 1 | 0.1 ± 0.01 | 0.5 ± 0.02 |
| 47 | Organic (OR 3) | 100 ± 4 | 50 ± 2 | 60 ± 5 | 0.3 ± 0.01 | 2 ± 0.1 |
| 48 | Organic (OR 4) | 200 ± 10 | 100 ± 5 | 130 ± 10 | 0.6 ± 0.03 | 4 ± 0.2 |

**Table 4** (continued)

| No. | Code of samples | $Ra_{eq}$ (Bq/kg) | $D_R$ (nGy./h) | AED (μSv./y) | $H_{ex}$ (mSv/y) | ELCR × $10^{-4}$ (1/person) |
|---|---|---|---|---|---|---|
| 49 | Organic (OR 5) | 30 ± 3 | 15 ± 1 | 20 ± 2 | 0.1 ± 0.01 | 0.6 ± 0.1 |
| 50 | Organic (OR 6) | 25 ± 3 | 13 ± 1 | 16 ± 2 | 0.1 ± 0.01 | 0.5 ± 0.06 |
| Over all | Range | 0.4–900 | 0.2–480 | 0.2–590 | 0–2.4 | 0–20 |
| | Average | 200 | 110 | 134 | 0.6 | 4 |

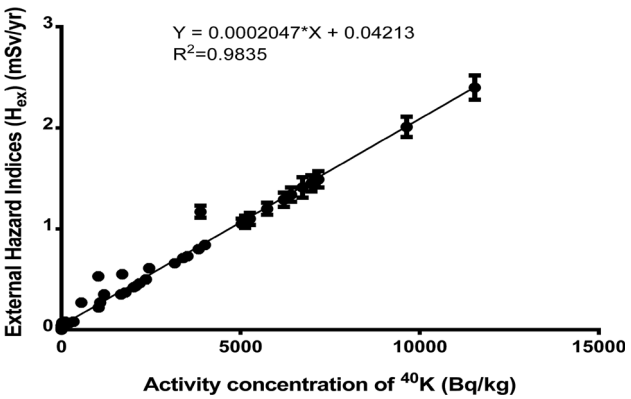


**Fig. 6** Relationship between activity concentration of $^{40}K$ and External Hazard Indices ($H_{ex}$) in fertilizer samples

hazard index ($H_{ex}$) higher than the recommended level and harmful for the health of farmers and workers working in a fertilizer factories and stores. On the other hand, fertilizer samples of urea, calcium nitrate, urea ammonium phosphate, di-ammonium phosphate and organic were safer for agriculture and they have a low effect on human health.

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