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# Measurement of Natural Radioactivity Level in Selected Phosphate Fertilizer Samples Collected from Iraqi Markets

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**Abstract**

The Gamma-ray spectrometry (spectra line Gp) and a high-purity germanium (HPGe) detector were used to determine the activity concentrations of $^{226}$Ra, $^{232}$Th, and $^{40}$K in fertilizer samples. $^{226}$Ra, $^{232}$Th, and $^{40}$K activity concentrations in fertilizers and phosphate raw. In this study the gamma index ($I_\gamma$), Radium equivalent dose ($Ra_{eq}$), dose rate ($D_R$), external hazard index ($H_{ex}$) internal hazard index ($H_{in}$), annual effective dose outdoor (AEDout) and annual effective dose indoor (AEDin) were found. This study could serve as a starting point for figuring out how fertilizer radiation affects human health.

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## 1. Introduction

The accumulation of primordial radionuclides like $^{238}$U and $^{232}$Th, as well as their decay products and singly occurring $^{40}$K, in soil and rocks of the environment, is directly influenced by terrestrial gamma radiation. Natural radioactivity and the resulting external gamma radiation exposure are primarily determined by local geological and geographical conditions, which vary in intensity in each location [1]. High-energy incident cosmic ray particles on the earth's atmosphere and radioactive nuclides that originate from the earth's crust and are present throughout the environment, including the human body, are the two main contributors to natural radiation exposures. Furthermore, industrial processes involving NORM may pose a radiological risk, so identifying and quantifying these risks is essential [2]. Fertilizers are important to ensure that crops obtain enough nutrients.to foster development and ensure a good harvest Fertilizers, on the other hand, may be harmful. Adulterated goods containing raw materials sourced from a variety of sources that are uncertain and/or suspect they can contain trace elements contaminants that are unintentionally added into soils, in addition to the certified nutritional ingredients for plants [3]. Phosphoric acid is the main raw material for phosphate fertilizers such as di ammonium phosphate (DAP), mono ammonium phosphate (MAP), potassium sulfate (K), Bitmous (B),urea ammonium phosphate (NP) single superphosphate (SSP), Nitrogen, Phosphorous and Potassium (NPK) which was made by reacting different amounts of $NH_3$ phosphoric acid directly During the phosphate rock reaction with sulphuric acid, the radioactive equilibrium between $^{238}$U, $^{232}$Th, and their decay products is broken, and the radionuclides migrate according to their solubility. Isotopes of uranium form highly soluble phosphate ion compounds, while $^{210}$Pb and $^{210}$Po isotopes accumulate in phosphorgypsum [4]. Furthermore, fertilizer materials used in plant nitration processes, such as phosphates (which contain $^{238}$U and $^{232}$Th) and $^{40}$K, are considered important sources of soil contamination and radioactivity. Due to external exposure during a resident's time in the farms and internal exposure through ingestion, this phenomenon may result in potential radiological risks [5].

## 2. Experimental Measurements

### 2.1 Sample collection and preparation

The fertilizers were obtained from different sources and fertilizer types, including 17 samples of commonly used fertilizers (potassium sulfate, NPK, MAP Urea, NP, DAP, and Bitmous) and one sample of phosphate raw as indicate in Table 1. taken for gamma spectrometric analysis, the samples were placed in small, airtight containers and held for a month in (hood) to reach the nuclear equilibrium state between radium and its daughter products. After a month, we put the samples in a one-kilogram marinelli beaker that have been washed with plain water and then distilled water. The samples were then dried in an oven at 80 $^{o}$C until they reached a constant weight [6].

**Table1.** Sample details.

| Sample code | Sample ID | Origin |
|---|---|---|
| Y1 | Potassium sulfate | Iraq |
| Y2 | Urea | Iraq |
| Y3 | NPK | Jordan |
| Y4 | MAP | Saudi |
| Y5 | NP | Iraq |
| Y6 | Potassium sulfate | Italia |
| Y7 | DAP | Saudi |
| Y8 | Bitmous | Romanian |
| Y9 | MAP | Jordan |
| Y10 | Urea | Iran |
| Y11 | Potassium sulfate | Holland |
| Y12 | NPK | United state |
| Y13 | Bitmous | Turkey |
| Y14 | Potassium sulfate | France |
| Y15 | NPK | Holland |
| Y16 | Urea | Iraq |
| Y17 | NPK | Russia |
| Y18 | Phosphate raw | Iraq |

## 2.2 Measurement setup

The study used a high pure germanium detector supplied by (BSI Baltic scientific instruments) composed of a germanium crystal (2×2) (HPGe) and computer software (spectral line Gp) shown in Figure 1. To minimize the contribution of X-ray fluorescence, the detector was placed inside a thick (4.5 cm) lead shield to reduce back ground radiation. The inner surfaces of the lead shield were graded lining with Copper sheet to reduce the contribution of X-ray fluorescence (0.8mm thick). The main amplifier, amplifier, and power supply are all connected to the device. The calculated background for counting (2 hrs.) [14]. Standard sources (CBSS 2) $^{241}$Am (59.3) Kev, $^{60}$Co (1173.1, 1332.3) Kev, and $^{137}$Cs (661.6) Kev were used to calibrate the detector Table 2. We counted each sample by placing it over the detector for (2 hrs.), we counted each sample by placing it over the detector for (2 hrs.), after which we measured the activity concentration of the samples by using software (spectral line Gp) as meantaited in Table 3. Some once spectrum of NP type of phosphate fertilizer (y5) shown in Figure 2.

**Table 2.** Calibration certificate radionuclides [12].

| Radionuclide | Half-life (days) | Activity (Bq/kg) | Combined standard uncertainty | Energy (Kev) |
|---|---|---|---|---|
| Am-241 | 157800 | 4.433 | 1.1 | 59.3 |
| Cd-109 | 4626 | 16.17 | 1.5 | 88.1 |
| Ce-139 | 137.5 | 0.740 | 1.1 | 165 |
| Co-57 | 271.26 | 0.855 | 1.1 | 122.1,136 |
| Co-60 | 1925.4 | 2.659 | 1.1 | 1173.1,1332.3 |
| Cs-137 | 11019 | 2.439 | 1.2 | 661.6 |
| Sn-113 | 115.1 | 3.087 | 2.2 | 392 |
| Sr-85 | 64.78 | 4.024 | 1.5 | 514 |
| Y-88 | 106.6 | 3.995 | 1.2 | 898 |
| Hg-203 | 46.72 | 2.064 | 2.4 | 898.02,1836.08 |

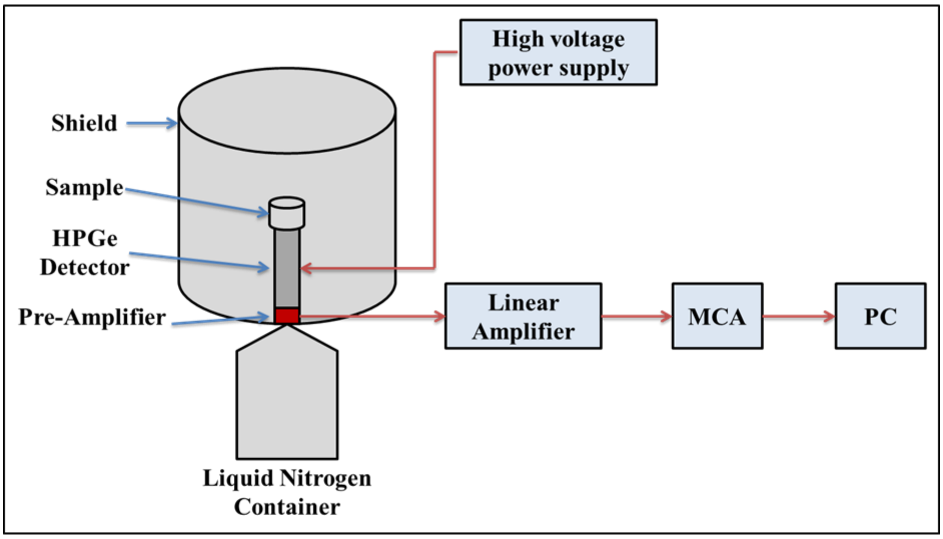


**Figure 1.** Experimental setup.

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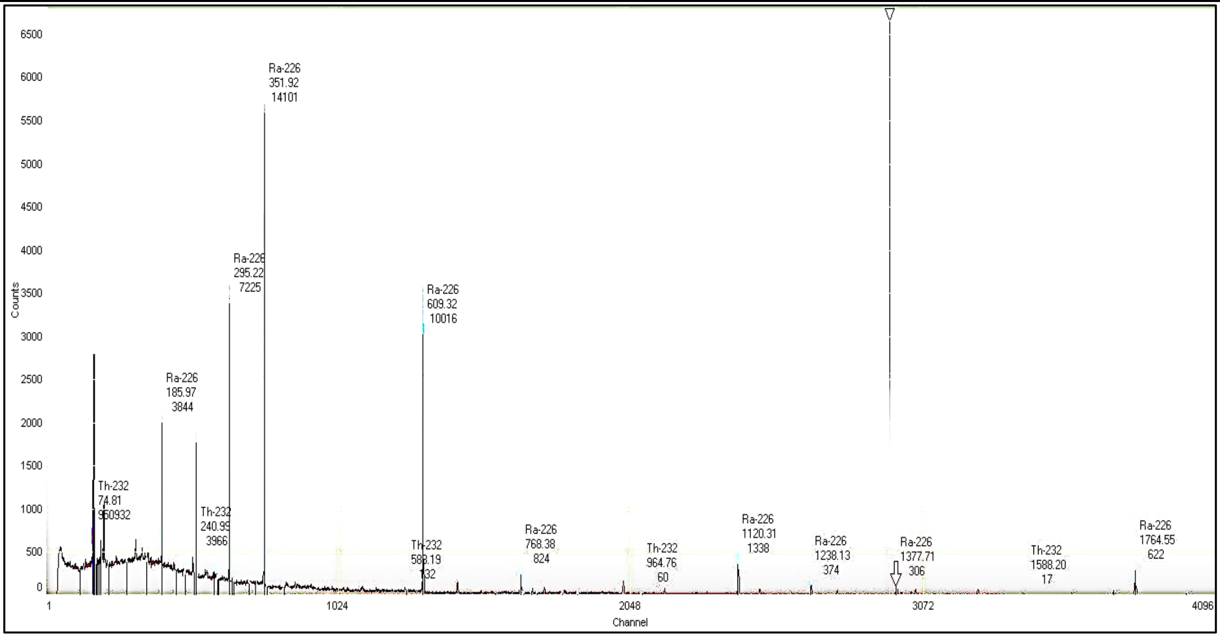


**Figure 2.** The spectrum of Np type phosphate fertilizer (y5).

## 2.3 Theoretical calculations

In this study, there are many parameters have been calculated, such as Activity concentration (A), Gamma index (Iγ), Gamma dose absorption (DR), Radium equivalent activity (Raeq), Annaual effective dose (AED), Hazard indices external (Hex) and internal (Hin).

***Activity concentration (A):*** The activity concentrations of natural radionuclides $^{226}$Ra, $^{232}$Th and $^{40}$K which is refer to number of nuclear decays per second can be calculated through γ-ray spectrometry by using the following formula [6]:

$$A = Net\ count/(\varepsilon \times I_\gamma \times T \times M) \qquad (1)$$

where ε the absolute gamma peak efficiency of the detector, $I_\gamma$ intensity for gamma-ray energy, T is the counting time for the measurement in seconds and M is the weight mass of the sample in kg. The results of activity concentration of fertilizers show in Table 3.

***Gamma index (Iγ):*** The decay intensity of the specific energy peak (Iγ) was calculated using the following equation [8]:

$$I_\gamma = (A_{Ra}/300) + (A_{Th}/200) + (A_K/3000) \qquad (2)$$

where $A_{Ra}$ is the specific activity of $^{226}$Ra, $A_{Th}$ is the specific activity of $^{232}$Th and $A_K$ is the specific activity of $^{40}$K, the results of gamma index of fertilizers shows in Table 4.

***Gamma dose absorption ($D_R$):*** ($D_R$) amounts in the air 1 m above the ground surface were determined on the basis of guidance given by UNSCEAR for the uniform distribution of naturally occurring radionuclides [8]:

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

The results of gamma dose absorption are shown in Table 4.

***Radium equivalent activity ($Ra_{eq}$):*** There is no uniform distribution of natural radioactive nuclides in the soil. The total exposure to radiation from $^{226}$Ra, $^{232}$Th and $^{40}$K nuclides was therefore expressed by radium equivalent activity ($Ra_{eq}$) in (Bq/kg). The radium equivalent activity of soil samples was calculated using the following formula [8]:

$$Ra_{eq} = (A_{Ra}) + (A_{Th} \times 1.43) + (A_K \times 0.077) \qquad (4)$$

The results of Radium equivalent activity show in Table 4.

***Annaual effective dose (AED):*** was estimated from the absorbed gamma dose concentrations ($D_R$) by using the dose conversion factor (F) of 0.7 Sv/Gy with an AED outdoor occupancy factor of 0.2 and 0.8 for AED indoor and determined using the following formula [9]:

$$AED(msv/year) = D_R(nGy/h) \times T \times F \qquad (5)$$

The results of annaual effective dose show in Table 4.

***Hazard indices external ($H_{ex}$) and internal ($H_{in}$):*** Described two indicators that describe internal and external radiation risks. The external hazard index is derived from the ($Ra_{eq}$) expression on the assumption that its permissible maximum value (equal to unity) corresponds to $Ra_{eq}$ (370 Bq/kg) upper limit. The external hazard index (Hex) is then defined
as follows [12]:

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

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

The results of hazal indices external ($H_{ex}$) and internal ($H_{in}$) show in Table 4.

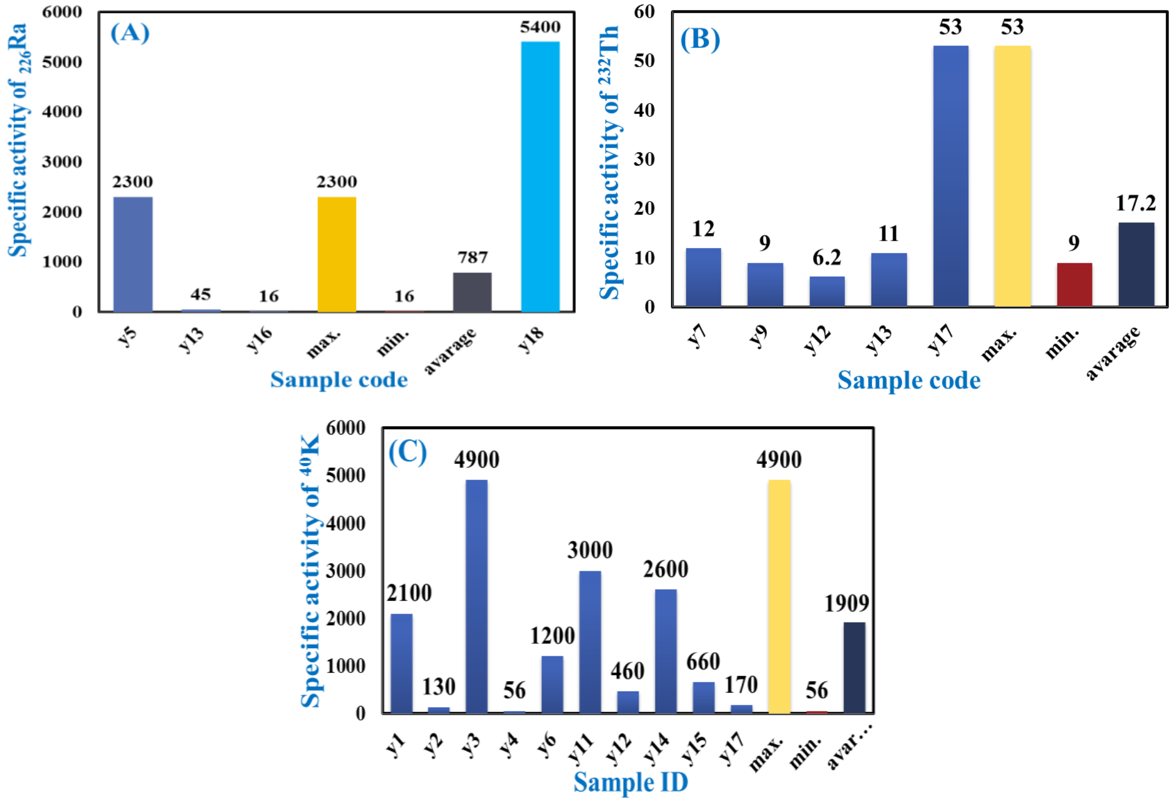


**Figure 3.** Specific activity for: (A) $^{226}Ra$, (B) $^{232}Th$, (C) $^{40}K$.

**Table 3.** The activity concentration of different types of fertilizers and phosphate raw.

| Sample code | Activity of $^{226}Ra$ (Bq/kg) | Activity of $^{232}Th$(Bq/kg) | Activity of $^{40}K$ (Bq/kg) |
|---|---|---|---|
| Y1 | ND | ND | 2100 |
| Y2 | ND | ND | 130 |
| Y3 | ND | ND | 4900 |
| Y4 | ND | 9 | 56 |
| Y5 | 2300 | ND | ND |
| Y6 | ND | ND | 1200 |
| Y7 | 12 | ND | ND |
| Y8 | ND | ND | ND |
| Y9 | ND | 12 | ND |
| Y11 | ND | ND | 3000 |
| Y12 | ND | 6.2 | 460 |
| Y13 | ND | 11 | ND |
| Y14 | ND | ND | 2600 |
| Y15 | ND | ND | 660 |
| Y17 | ND | 53 | 170 |
| Y18 | 5400 | ND | ND |

*ND: detection limit

**Table 4.** For different type of fertilizers The radiological hazards including gamma ray index (Iγ), absorbed dose rate (D) and radium equivalent activity (Raeq), internal (Hin) and external (Hex) hazard indices, internal (AEDEin) and external (AEDEex) annual effective dose.

| Sample Code | Iγ | DR(nGy/h) | $Ra_{eq}$ Bq/kg | $H_{in}$ | $H_{ex}$ | $AED_{indoor}$ µSv/y | AEDoutdoor µSv/y |
|---|---|---|---|---|---|---|---|
| Y1 | 0.7 | 87.57 | 161.7 | 0.43 | 0.43 | 0.42 | 0.10 |
| Y2 | 0.043 | 54.21 | 10.01 | 0.027 | 0.027 | 0.26 | 0.066 |
| Y3 | 1.63 | 204.33 | 343 | 1.018 | 1.018 | 1.002 | 0.250 |
| Y4 | 0.063 | 7.76 | 17.18 | 0.041 | 0.041 | 0.03 | 0.0095 |
| Y5 | 7.66 | 1062.6 | 2300 | 12.4 | 12.4 | 5.21 | 1.3 |
| Y6 | 0.4 | 50.04 | 92.4 | 0.42 | 0.24 | 0.24 | 0.06 |
| Y7 | 0.06 | 7.24 | 17.16 | 0.04 | 0.04 | 0.03 | 0.0088 |
| Y9 | 0.06 | 7.24 | 17.16 | 0.046 | 0.046 | 0.03 | 0.0088 |
| Y11 | 1 | 125.1 | 231 | 0.623 | 0.62 | 0.61 | 0.15 |
| Y12 | 0.18 | 22.92 | 44.28 | 0.119 | 0.119 | 0.11 | 0.028 |
| Y13 | 0.055 | 6.64 | 15.73 | 0.04 | 0.04 | 0.03 | 0.0081 |
| Y14 | 0.86 | 108.42 | 200.2 | 0.54 | 0.54 | 0.53 | 0.132 |
| Y15 | 0.22 | 25.02 | 46.2 | 0.13 | 0.12 | 0.13 | 0.03 |
| Y17 | 0.32 | 39.1 | 88.8 | 0.23 | 0.23 | 0.19 | 0.04 |
| Y18 | 18 | 2494.8 | 5400 | 29.18 | 29.18 | 12.23 | 3.05 |
| Min | 0.043 | 6.64 | 10.01 | 0.027 | 0.027 | 0.03 | 0.0081 |
| Max | 18 | 2494.8 | 5400 | 29.18 | 14.56 | 12.23 | 3.05 |

## 3. Results and Discussion

The spectra of 17 fertilizer samples different types from different markets in Iraq, as well as one phosphate raw sample from phosphate quarries in Al-Qaim, Iraq, were analyzed. Table 2. presents the activity concentrations of natural radioactive nuclides $^{226}Ra$, $^{232}Th$, and $^{40}K$. Phosphate row and Np type Iraqi phosphate fertilizer had the highest and lowest concentrations of $^{226}Ra$, with 5400Bq/kg and 2300 Bq/kg respectively as shown in Figure 3(A). With 53 Bq/kg, NPK Russian phosphate fertilizer had the highest concentration of $^{232}Th$. The lowest concentration of $^{232}Th$ was found in NPK American fertilizer, which had a concentration of 6.2 Bq/kg as shown in Figure 3(B). NPK Jordanian fertilizer had the highest activity concentration of $^{40}K$ at 4900 Bq/kg, while MAP Saudi fertilizer had the lowest activity concentration at 56 Bq/kg as shown in Figure 3(C). The other two Urea samples showed no activity because their ores lost naturally occurring radionuclides. In this study, activity concentrations in fertilizer samples from various countries were found to be within acceptable limits, which are 2300Bq/kg for $^{238}U$, 460Bq/kg Bq/kq for $^{232}Th$, and 10000Bq/kq for $^{40}K$, respectively (IAEA). We noticed that the activity concentrations of $^{226}Ra$ in raw phosphate were higher than the global average, while the activity concentrations of $^{232}Th$ were within the global average. All samples had activity concentrations of $^{40}K$ that were lower than the global average (IAEA). Table 4. shows the results of hazard indices,the highest value of gamma index Iγ was 18 in phosphate raw, while its lowest value of was 0.043 in Urea Iraqi fertilizer as shown in Figure 4(A) The study indicates that the gamma index values for the investigated were more than the recommended value of 1 for the gamma index provided by (UNSCEAR, 2000) [1]. The maximum value of absorbed gamma dose rate ($D_R$) was found to be 2494 nGy/h in phosphate raw, while the minimum value was found to be 6.64 nGy/h in Bitmous Turkish as shown in Figure 4(B). The present findings revealed that absorbed gamma dose rate values were higher than the recommended value of 55 nGy/h for the absorbed gamma dose rate provided by (UNSCEAR, 2000) [1]. The overall $Ra_{eq}$ is 5400 Bq/kg in phosphate raw, which is a high value as compared to the ICRP recommended value. The minimum value is 10.01Bq/kg in Urea Iraqi fertilizer as shown in Figure 4(C), which is acceptable. The external hazard $H_{ex}$ values ranging from 14.59 to 0.027 in Urea and phosphate raw respectively indicate that certain fertilizers have an external threat greater than unity and more than the action level as shown in Figure 4(E). The internal hazard $H_{in}$ values ranging from29.18 to 0.02 in Urea and phosphate raw respectively as shown in Figure 4(D) indicate that certain fertilizers have an external threat greater than unity and more than the action level. The highest value of outdoor annual effective dose equivalent $AED_{out}$ was 3.05 mSv/y in phosphate raw, while its lowest value of outdoor annual effective dose equivalent was 0.0081 mSv/y in Bitmous Turkish as shown in Figure 4(G),in the study indicate that the outdoor annual effective dose equivalent values for the investigated were more than the recommended value of 1mSv/y for the outdoor annual effective dose equivalent provided by (UNSCEAR, 2000) [1]. The highest value of outdoor annual effective dose equivalent $AED_{in}$ was 12.23mSv/y in phosphate raw, while its lowest value of outdoor annual effective dose equivalent was 0.03mSv/y in (Saudi DAP, Jordanian MAP and Turkish Bitmous) as shown in Figure 4(F), in the study indicate that the indoor annual effective dose equivalent values for the investigated were more than the recommended value of 1 mSv/y for the indoor annual effective dose equivalent provided by (UNSCEAR,2000) [1]. The results demonstrate that the fertilizers have a wide spectrum of gamma activity.

This could be as a result of the following factors:

1. The many sites where raw minerals for fertilizer production were sourced.
2. The distinctions between fertilizer factories that generate fertilizer.
3. The characteristics of radioactive elements in raw materials varied depending on where they were discovered.

The high values of the radioactivity in phosphate raw indicate to the raw minerals for fertilizer manufacturing were sourced from various locations, and radioactive elements in raw materials had various structures depending on where they were found [11].

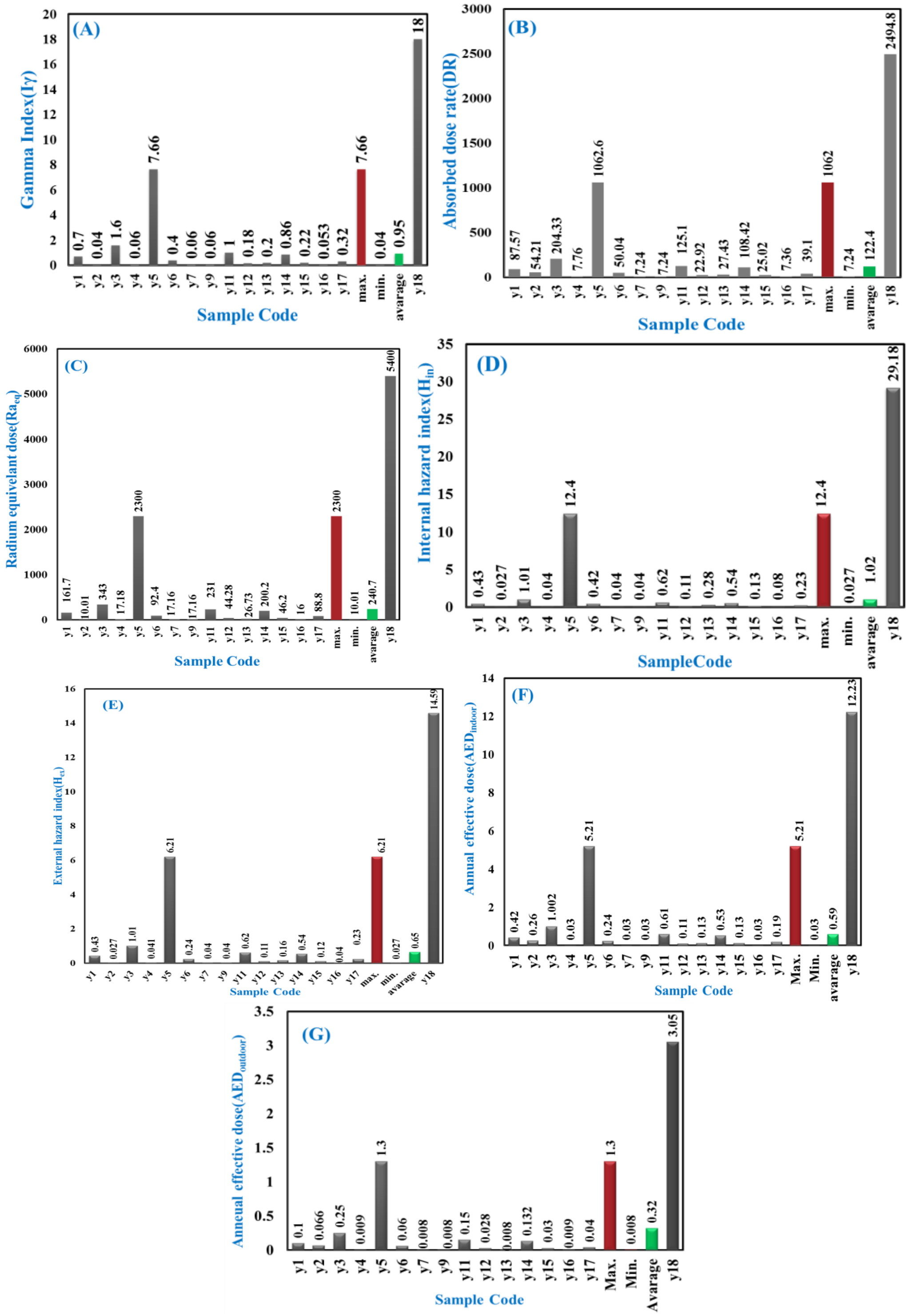


**Figure 4.** The Hazard indices of phosphate fertilizers.

## 4. Conclusions

1. The activity of fertilizer samples within the global word limit
2. Plant and animal fertilizers are preferred over artificial fertilizers because low level of radioactivity in plant and animal fertilizers.
3. High activity concentration in phosphate raw because it was random sample.

## 5. Aim of Study

The aim of this study is to measure the concentration of natural radioactivity in fertilizer samples and calculate the hazard indices of these radionuclides.

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