

Determination of Activity Concentration and Radiological Parameters of Some Radionuclides Found in Selected Fertilizers in Yola Adamawa State, Nigeria

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ABSTRACT

Agricultural fertilization increases efficiency and improves product quality recovery during agricultural activities. However, the major routes through which humans are exposed to ionizing radiation are via the food chain, which results in the absorption of radionuclides. This research work was carried out to determine the radioactivity concentration, hazard indices, and absorbed dose rate of some fertilizers used in Adamawa State, Nigeria. Six different fertilizer samples (Cow dung, Chicken dung, NPK 20:10:10, Urea, NPK 15:15:15, NARICT Minero-organic NPK 7:7:7) were used for the study, and a NaI (TI) gamma-ray spectrometer was used to determine the activity concentrations. The results showed that ²²⁶Ra concentration was lower than the recommended threshold of 35Bq/kg in all the samples. Except for urea and cow dung, the concentration of ⁴⁰K for all other fertilizer samples was above the UNSCEAR-recommended threshold of 420 Bq/kg. Also, the fertilizer samples were all above the recommended threshold of 45Bq/kg for ²³²Th. Internal and external hazard indices were lower than recommended limits of ≤ 1 , respectively, except for NPK 15:15:15, while the gamma index and dose rates were higher than the UNSCEAR recommended threshold of ≤ 1 , 60nGy/hr and 84nGy/hr, respectively, for all samples.

Keywords: Dose Rate, Radioactivity Concentration, Radionuclide, Fertilizers, Hazard Indices.

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Introduction

Fertilization increases efficiency and enhances the quality of product recovery in agricultural activities, and it is one of the most important ways of adding nutrients to the soil. Fertilizer is considered to be a source of natural radionuclides and a potential source of heavy metals. It contains heavy metals like Hg, Cd, As, Pb, Cu, and Ni; and natural radionuclides such as ²³⁸U, ²³²Th, ⁴⁰K, and ²¹⁰Po. However, in recent years, fertilizer usage has increased exponentially throughout the world, and this may result in serious health and environmental problems arising from the accumulation of radionuclides in soil and

plant systems. Plants absorb nutrients from fertilizers within the soil, and then pass them on to human beings through the food chain. Thus, fertilization leads to water, soil, and air pollution [1].

Estimating the levels of radiation in the environment is crucial in implementing appropriate controls for the sake of radiological protection since the constituents that make up the environment contains various number of radionuclides and their decay products. The major ways by which humans become exposed to these radiations are radiation from sources outside the body

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(external exposure), radionuclides that are ingested through consumption of food and water, or as inhaled radioactive gases (internal exposure) [2].

An investigation of gamma activity in the chemical fertilizers has been performed to determine the effect of the use of chemical fertilizers on human health. A possible negative effect of chemical fertilizers is contaminating the cultivated hands by trace metals and some naturally occurring radioactive materials (NORM) [3]. Radionuclides present in phosphate fertilizers affect the common people and farmers immensely. A large concentration of natural radionuclides in the fertilizers contaminates the environment and agricultural fields [4]. Direct inhalation of dust from phosphate fertilizers could affect farmers on agricultural land [5]. Exposure of workers and the public to radiation from fertilizers is, therefore, not unlikely [6]. Higher radiation levels are associated with the chemical fertilizers; therefore, the study of natural activity in artificial fertilizers is of great interest in the field of environmental pollution [5].

Based on the research conducted by Hatika, fertilizer samples (Urea, ZA, KCL, NPK, TSP, and Phosphate) contained ^{238}U , ^{232}Th , ^{226}Ra , and ^{40}K , elements in each sample. The elements found in the samples had different values. The highest activity of ^{238}U ($558.66 \pm 5.64\text{Bq/Kg}$) is in the TSP fertilizer sample. The highest activity of ^{232}Th ($2925.05 \pm 84.25\text{Bq/Kg}$) is found in the NPK fertilizer sample. The highest activity of ^{226}Ra ($69.77 \pm 0.94\text{Bq/Kg}$) is in the TSP fertilizer sample, and the highest activity of ^{40}K ($17.87 \pm 6.31\text{Bq/Kg}$) is found in the KCL fertilizer sample [7]. Radium equivalent activity in the sample was highest on the fertilizer samples KCL (835.31525 Bq/kg), followed by Phosphate fertilizers (495.331 Bq/kg); then NPK, TSP, ZA, and Urea, which are 325.886 , 82.003 , 1.978 , and 1.183 Bq/kg , respectively. Radium equivalent activity at KCL and Phosphate fertilizer samples exceeds 370 Bq/kg , which is the maximum permissible dose of radiation [8]. The minimum and maximum values of absorbed dose in the samples studied in the work were found to vary from 0.58 to 452.37 nGy/hr . The absorbed doses are 452.37 , 268.16 , 167.25 , 38.16 , 0.92 , and 0.58 nGy/hr for the KCL, phosphate, NPK, TSP, ZA, and Urea fertilizers, respectively. The index of gamma radiation risks is highest for the KCL at 7.27 , followed by Phosphate (4.31), NPK (2.64), TSP (0.56), ZA (0.014), and Urea (0.009) [7].

Ilori & Alausa studies showed also that, the activity concentration of ^{40}K , ^{238}U and ^{232}Th in the cow dung samples range from 115.21 to 225.44 Bq/kg with a mean value of $184.90 \pm 40.92\text{ Bq/kg}$ for ^{40}K ; 10.13 to 13.17 Bq/kg with a mean value of $11.50 \pm 0.78\text{ Bq/kg}$ for ^{238}U and 8.12 to 11.28 Bq/kg with a mean value of $10.20 \pm 0.71\text{ Bq/kg}$ for ^{232}Th [9]. The activity concentrations in cattle dung samples were higher than the corresponding values in the grass from the study area. This may be attributed to the ingestion of other sources of radionuclides, such as the drinking water available to the cattle grazing in the study area [10]. The ^{40}K value of $218.6 \pm 66.0\text{ Bq/kg}$ obtained in Abeokuta was higher than that of the present study. However, the ^{238}U and ^{232}Th activity concentration values of 10.5 ± 1.1 and $8.3 \pm 1.9\text{ Bq/kg}$ obtained in Abeokuta were lower when compared to those obtained in the present study.

This study aims to determine the activity concentrations, hazard risk indices, and dose rates of radionuclides found in some fertilizers used in Yola, Nigeria. This is done to ascertain the possible risk they may pose by the continuous application of these fertilizers to the same piece of land for cultivation over a long period.

Materials and Methods

Sample Area

The study area was Adamawa, Nigeria, a state sharing boundaries with Taraba, Borno, Gombe states, Cameroon, and the Republic of Niger.

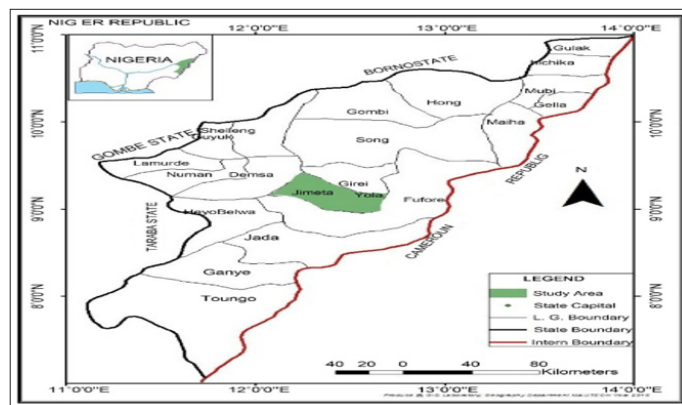


Figure 1: Map of Adamawa State, Nigeria (<https://www.googlemaps.com>).

Sample Collection

Six different fertilizer samples (NPK 20:10:10, NPK 15:15:15, NARIC Minero-organic NPK 7:7:7, Urea, Chicken dung, and Cow dung) were collected from agro-chemical stores in Jimeta modern market, Cattle range at Kasuwan Shanaye along bypass road Jimeta, NARICT Yola office, and Flova poultry farm along army barracks road Jimeta, Yola North, Nigeria.

Sample Preparation and Analysis

Sample preparation and analysis were carried out as described by CERT [12]. 1 Kg of each sample collected was crushed to a powdered form using a Vitamix A3500 ascent series smart blender, and sieved through a 1 mm mesh screen to remove the larger grain sizes. The ground samples were then air dried in a temperature-controlled room ($21\text{ }^{\circ}\text{C}$) for 24 hours so that moisture is removed from the different fertilizers. After moisture removal, the samples were ready and packaged in 1 Kg Ziploc bags for radionuclide analysis.

Evaluation of Radioactivity of Samples

The sealing process included smearing of the inner rim of each container lid with Vaseline petroleum jelly, filling the lid assembly gap with candle wax to block the gaps between lid and container, and tightly sealing the lid-container with masking adhesive tape. Radon and its short-lived progenies were allowed to reach secular radioactive equilibrium by storing the samples for 30 days before gamma spectroscopy measurements [12]. The data acquisition software used was Maestro by Canberra Nuclear Products. Each sample was measured for a period of 29000 seconds. The peak area of each energy in the spectrum

was used to compute the activity concentrations in each sample using equation (1):

$$C (Bq.kg^{-1}) = C_n / C_f \quad (1)$$

Where C is the activity concentration of the radionuclides in the sample measured in $Bq.kg^{-1}$, C_n is the count rate (counts per second). Count per second (cps) = $Net\ Count / Live\ Count$, C_{fc} is the calibration factor of the detecting system [12].

Calibration and Efficiency Determinations

Calibration of the system for energy and efficiency was done with two calibration point sources: Cs-137 and Co-60. These were done with the amplifier gain that gives 72% energy resolution for the 661.7KeV of Cs-137 and counted for 30minutes. The standards used to check for the calibration are the IAEA gamma Spectrometric reference materials RGK-1 for ^{40}K , RGU-1 for ^{226}Ra (Bi-214 peak), and RTG-1 for ^{232}Th (Ti-208). The background count rate was measured for 29000sec [12].

Table 1: Spectral Energy windows used in the Analysis.

Isotope	Gamma Energy (KeV)	Energy Window (KeV)
R-226	1764.0	1620 – 1820
Th-232	2614.5	2480 – 2820
K-40	1460.0	1380 – 1550

Table 2: Energy Calibration for Quantitative Spectral Analysis

Isotope	Calibration Factors		Activity Concentration Conversion Factors	Detection Limits	
	10^{-3} (cps/ppm)	10^{-4} (cps/ppm)	Bq/Kg	Ppm	Bq/Kg
^{40}K	0.0260	6.4310	0.0320	454.5400	14.5400
^{226}Ra	10.5000	8.6320	12.2000	0.3200	3.8400
^{232}Th	3.6120	8.7680	4.1200	2.2700	9.0800

Theoretical Framework

Risk Index Gamma Radiation

In determining the gamma radiation risk index ($I_{\gamma r}$), [13]. The equation used is:

$$I_{\gamma r} = 0.0067 A_{Ra} + 0.01 A_{Th} + 0.00067 A_K \leq 1 \quad (2)$$

where A_{Ra} , A_{Th} , and A_K are the activity of ^{226}Ra , ^{232}Th and ^{40}K .

External and Internal Hazard

The external and internal radiation exposure of a sample is defined in relation to an external and internal hazard called the external and Internal Hazard index, H_{ex} , H_{in} , and is expressed as

$$H_{in} = A_{^{238u}}/185 + A_{^{232Th}}/256 + A_{^{40K}}/4810 \leq 1 \quad (3)$$

where A is still the specific activity concentration, and H_{in} is the internal hazard index [13].

$$H_{ex} = A_{^{238u}}/370 + A_{^{232Th}}/259 + A_{^{40K}}/4810 \leq 1 \quad (4)$$

where A is the specific activity concentration, and H_{ex} is the external hazard index [13].

Absorbed Dose

Absorbed dose (D) is a measure of gamma radiation in the air at a height of 1 m from the surface to equalize the distribution of naturally radioactive materials, or commonly abbreviated as NORM, i.e., ^{226}Ra , ^{232}Th , and ^{40}K . The equation used to determine D is: [14]

$$D(out)Bq/kg = 0.427 A_{Ra} + 0.662 A_{Th} + 0.043 A_K \quad (5)$$

$$D(in)Bq/kg = 0.92 A_{Ra} + 1.1 A_{Th} + 0.081 A_K \quad (6)$$

where A_{Ra} , A_{Th} , and A_K are the activities of ^{226}Ra , ^{232}Th , and ^{40}K , respectively [13].

Results And Discussion

Activity Concentrations of ^{40}K , ^{226}Ra , and ^{232}Th in the Samples.

The results of the activity concentration of the radionuclides (^{40}K , ^{226}Ra , and ^{232}Th) in each of the different samples are presented in Table 3.

Table 3: Activity Concentration of ^{40}K , ^{226}Ra , and ^{232}Th in the Samples measured in Bq/Kg

	K-40 (Bq/Kg)	Ra-226 (Bq/Kg)	Th-232 (Bq/Kg)
NPK 20:10:10	2012.60±4.34	4.38±0.15	83.36±1.33
NPK 15:15:15	2116.26±3.53	5.99±0.14	152.52±1.08
NARICT NPK 7:7:7	1207.65±3.72	4.80±0.20	143.20±1.48
Urea	227.65±2.78	4.24±0.17	125.94±1.26
Chicken Dung	434.55±3.52	4.33±0.14	111.08±1.26
Cow Dung	306.16±3.49	2.71±0.13	108.76±3.93
Permissible limits [8]	420.00Bq/Kg	35.00Bq/Kg	45.00Bq/Kg

From Table 3, the concentration of ^{40}K in inorganic fertilizers ranges from 2116.27±3.53Bq/kg to 227.65±2.78 Bq/kg, with NPK 15:15:15 having the highest value and urea having the lowest value, respectively. These values of ^{40}K are contrary to what was obtained by Alam et. al; Alsaffar M.S. & Jaafar; Hussain & Hussain; & Alharbi A. & El. Taher A, where concentration of ^{40}K ranged from 199.20±41.03 - 212.32±32.90Bq/kg, 8-12628Bq/kg, 12-2276Bq/kg, and 80.85-5000Bq/kg [15,16]. Except for urea, other inorganic fertilizers are above the UNSCEAR world recommended limit value of 420Bq/kg. While the organic fertilizer ranges from 306.16±3.49Bq/kg to 434.55±3.52Bq/kg, cow dung has the lowest value, while chicken dung has the highest respectively. Cow dung is within the UNSCEAR world recommended value of 420 Bq kg⁻¹, while chicken dung, it is above the world recommended value of 420Bqkg⁻¹ [8].

The activity concentration of ^{226}Ra in inorganic fertilizers ranges from 5.99±0.14Bq/kg to 4.24±0.17Bq/kg, NPK 15:15:15 having the highest value, while urea has the lowest. These values of

^{226}Ra are contrary to what was obtained by Alam et. al; Alsaffar M.S. & Jaafar, Hussain & Hussain; & Alharbi A. & El. Taher A., where concentrations were higher and ranged from 5-329Bq/kg, 12-244Bq/kg, 10.97 ± 0.57 - 13.17 ± 0.44 Bq/kg, and 0.50 ± 0.20 - 134.00 ± 6.00 Bq/kg [15,16]. Therefore, this shows that most of the concentrations from the study were less than those from the reviewed study, possibly due to the differences in products from different manufacturers. That of organic fertilizers ranges from 4.33 ± 0.14 Bq/kg to 2.71 ± 0.13 Bq/kg, cow dung having the lowest value and chicken dung having the highest value. All the different fertilizer samples (inorganic, organic, and minero-organic fertilizers) had ^{226}Ra concentration lower than the recommended value of 35Bqkg⁻¹ set by UNSCEAR [8].

Concentration of ^{232}Th in inorganic fertilizers ranges from 83.36 ± 1.33 Bq/kg to 152.52 ± 1.08 Bq/kg, NPK 20:10:10 having the lowest value, while NPK 15:15:15 has the highest value, respectively. Values of ^{232}Th contrast that were obtained by Alam et. al; Alsaffar M.S. & Jaafar,; Hussain & Hussain; & Alharbi A. & El.Taher A. where concentration ranged from 3-22Bq/kg, 1-27Bq/kg, 0.20 ± 0.04 - 69.77 ± 1.23 Bq/kg, 5-39Bq/kg, 8.12 ± 1.41 - 10.36 ± 0.60 Bq/kg [15,16]. Results of the organic fertilizer range from 108.76 ± 3.93 Bq/kg to 111.08 ± 1.26 Bq/kg, cow dung having the lowest value, while chicken dung has the highest value. All the different fertilizer samples were all above the recommended value of 45Bqkg⁻¹ set by UNSCEAR [8].

Results from all samples showed consistently high concentrations of ^{40}K , ^{226}Ra , and ^{232}Th in NPK fertilizers and lower concentrations in the Nitrogen and organic fertilizers. NPK 15:15:15 had the highest concentrations of ^{40}K , ^{226}Ra , and ^{232}Th as compared to NPK 20:10:10 and NARICT NPK 7:7:7. This may be as a result of the percentage composition of phosphorus and potassium, since nitrogen posed less significant effect as seen in the pure nitrogen (urea) fertilizer. The study also showed a higher concentration of ^{40}K , ^{226}Ra , and ^{232}Th in chicken dung than in cow dung and urea, possibly due to the chemical composition of the chicken dung.

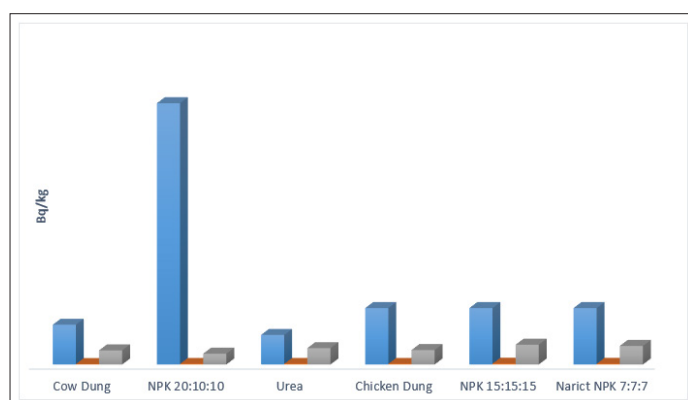


Figure 2: Activity Concentration of ^{40}K , ^{226}Ra , and ^{232}Th of all the samples.

Radiological Hazard Index in the Samples

Table 4: Values of the Radiological Hazard indices for the Samples

	$I_{\gamma T}$	H_{in}	H_{ex}
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NPK 20:10:10	2.21	4.38 ± 0.15	83.36 ± 1.33
NPK 15:15:15	2.98	5.99 ± 0.14	152.52 ± 1.08
NARICT NPK 7:7:7	2.27	0.84	0.82
Urea	2.81	0.56	0.55
Chicken Dung	1.43	0.55	0.53
Cow Dung	1.31	0.50	0.49
Permissible limits [8]	≤ 1	≤ 1	≤ 1

From Table 4, the gamma hazard risk index for inorganic fertilizers ranges from 2.21 to 2.98, with NPK 20:10:10 having the lowest value, while NPK15:15:15 has the highest value. These values are significantly contrary to those obtained by Gupta et al; Lambart R., *et. al.*, which showed that fertilizer samples in India, Indonesia, Egypt, Saudi Arabia, and Brazil ranged from 0.009 to 7.27 [17,3]. showing that the gamma risk index in other countries is significantly higher than that of the present study. That of organic fertilizers ranges from 1.31 to 1.43, cow dung having the lowest value, while chicken dung has the highest value. Implying that all the indices are greater than unity, thus the fertilizers require radiological attention for monitoring and protection [8].

All fertilizer samples had internal and external hazard indices less than unity, except for NPK 15:15:15, which had values of 1.07 and 1.05 for internal and external hazard indices, respectively. These values are also contrary with that obtained by Gupta et al; Lambart R., *et. al.*, which showed that fertilizer samples in India, Indonesia, Egypt, Saudi Arabia, and Brazil had internal and external indices ranging from 0.198 – 3.218 and 0.118 – 2.170, respectively [17,3]. Therefore, proper monitoring and protection are required in the application of NPK 15:15:15 fertilizers, as it is above the recommended value of ≤ 1 set by UNSCEAR [8].

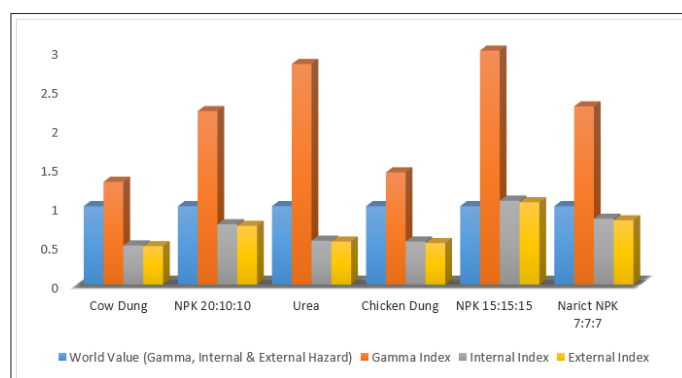


Figure 3: Comparison of gamma hazard index, internal and external hazard index with the world recommended value of ≤ 1 [8].

Result of Outdoor and Indoor Absorbed Dose

Table 5: Outdoor and Indoor Absorbed Dose Rates (nGy/hr) of the Samples

	D _{Outdoor} (nGy/hr)	D _{Indoor} (nGy/hr)
NPK 20:10:10	143.59	258.74
NPK 15:15:15	194.52	344.70
NARICT NPK 7:7:7	148.78	259.76
Urea	94.97	160.87
Chicken Dung	94.07	161.36
Cow Dung	86.32	146.93
Permissible limits [14]	60nGy/hr	84nGy/hr

The outdoor absorbed dose rate of the inorganic fertilizers ranged from 94.97nGy/hr to 194.52nGy/hr, NPK 15:15:15 having the highest value, while urea has the lowest value. The values were compared to the values obtained by Gupta *et al* and Lambart *et. al.*, which showed the values ranging from 32 – 241nGy/hr, 31 – 230nGy/hr, and 30 – 225nGy/hr [17,3]. Even though the results from present studies were all above the recommended value of 60nGy/hr, they still showed that outdoor absorbed dose rates were higher in the NPK fertilizers than in the pure nitrogen and organic fertilizers, with NPK 15:15:15 as the highest. The organic fertilizers range from 94.07nGy/hr to 86.32nGy/hr for chicken dung and cow dung, respectively. All the inorganic, organic, and mineroorganic fertilizers sampled were above 60nGy/hr, which is the world recommended value by UNSCEAR [14]. This means that individuals using all the sampled fertilizers could be exposed to hazard risks through the food chain and therefore need to carefully use protective measures during application.

The indoor absorbed dose rate for the inorganic fertilizers also ranges from 160.87nGy/hr to 344.70nGy/hr, NPK 15:15:15 having the highest value and urea having the lowest value. These values are also contrary to 40 – 340nGy/hr, and 41.074 – 58.481nGy/hr, obtained in separate studies conducted by Gupta *et al.* and Lambart *et. al.* [17,3]. Both indoor and outdoor absorbed dose rates were higher than the recommended value for all the different fertilizers. The indoor absorbed dose of organic fertilizer ranges from 146.93nGy/hr to 161.36nGy/hr for cow dung and chicken dung, respectively. The inorganic and organic fertilizers were above the recommended value of 84nGy/hr by UNSCEAR, which also showed that persistent use poses a risk to individuals through the food chain and application [14].

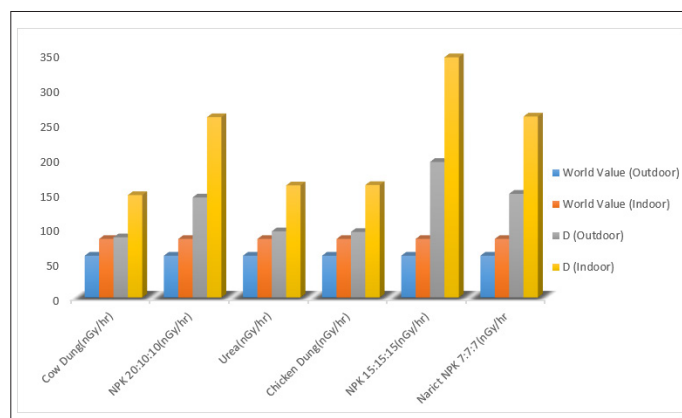


Figure 4: Comparison of Absorbed Dose with the world recommended values

Conclusion

Among the six different fertilizer samples discussed, the radionuclide activity concentration, maximum and minimum values varied across the three radionuclides examined (²²⁶Ra, ²³²Th, and ⁴⁰K), except for NPK 20:10:10, 15:15:15, and 7:7:7, all others were less than 420Bq/kg recommended value for ⁴⁰K all fertilizer samples had values for ²²⁶Ra less than the recommended value of 35Bq/k, while all the values for ²³²Th of the different fertilizer samples were above the recommended value of 45Bq/kg. The Internal and external indices were all below unity, which is the recommended maximum value by, except for the internal and external indices of NPK 15:15:15, which were all above unity [14]. This suggests adequate monitoring, protection, and safety measures to ensure the safety of individuals using such fertilizers. The gamma hazard index for all the different fertilizer samples was above unity, which shows and recommends attention to gamma hazards on all the fertilizers. Therefore, soil analysis should be performed carefully, before fertilizer application to soil, to minimize radiation hazard as low as reasonably achievable.

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