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Assessment of natural radionuclides in a fertilized farmland in Abeokuta, Nigeria: Implications for environmental radioactivity evaluation and monitoring

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Abstract. In this study, the concentrations of radionuclides were determined for soil samples obtained from a fertilized farm soils in Abeokuta, southwestern Nigeria. On the average, seven soil samples were obtained and kept in a sealed polythene bags. Then later dried and pulverized. Two hundred grams of each of the pulverized samples was held in a standard plastic container and left for about thirty days to allow for secular equilibrium between radium and its progenies. Radionuclides activity concentrations within the collected samples were measured using a calibrated NaI-based gamma-ray spectrometer. The detected natural radioactivity of soil samples are presented and radiological attributes for effective radiological hazards detection and monitoring for the study location are equally estimated. The results revealed that the specific activity for K-40 radionuclides at location L7 is higher than the world average concentration of 420 Bq/kg. This is not unconnected to the fertilizer within that portion of the farm. However, the estimated radiological attributes in the farmland are relatively lower that the threshold standards, which implies that the soils in the farmland are free of any radiation hazards and pose no threat to farmers and the residents of that area. It is however recommended that organic manure should be used in the farmland rather than fertilizer and routine check on environmental radioactivity of the farmland should be done regularly for adequate monitoring.

Keywords: Environmental radioactivity, Fertilized farmland, radiological parameters

1. Introduction
Everybody eats food for strength required for day to day activities. The food is majorly obtained from different farmlands and in order to get good yield of farm products, fertilizers may need to be applied to these farmlands. Humans are exposed to radiation everyday by consuming these plants. The fertilizer applications in soils may increase the concentrations of these radionuclides [1-3]. Hence, concentrations of radionuclides in fertilized-farm soil pose a major concern to human health after consuming the farm products, if their values exceed recommended limits. It is therefore necessary to devise the means to know the yearly ingestion dose and keep this dose at a minimum. Different researches have been carried out to evaluate activity concentrations of radionuclides in farmlands and it has been established that fertilizer
application may increase the concentrations of radionuclides in soil [2-4]. These natural radionuclides behave biologically and chemically the same way non-radioactive forms would behave [5]. Plants take up radionuclides in the soil. In the end man consumes the contaminated farm products which had absorbed radionuclides from the soil. Sooner or later, ingested radionuclides will emit ionizing radiation that can cause cell damage. This can lead to changes in the genetic make-up and DNA of the cells, or sometimes cause ultimate death of the cells [5]. Thus it is highly necessary to know the yearly radiation dose from fertilized farm soils. Outputs of such study would be compared with standard/recommended limits.

Many investigations have been carried out by researchers on the presence of radionuclides in fertilizers and on the implications of fertilizer usage on the environment [6-13]. [6]did a general paper review in South Africa on evaluation of the impacts of the ionizing radiation on livestock as important segment of food chain for humans and advocate for some precautionary measures to be taken. They highlighted grazing on radionuclide contaminated forages as one of the ways of exposing animals to ionizing radiation and pointed out that adequate systems for major food monitoring should be designed with the sole purpose of reducing radiation exposure due to consumptions of animals and their products. It is known from the works of [2] and [4]that application of fertilizer in soils used for growing forages is one of the ways of contaminating forages with radionuclides. [3]reported that foodstuffs in Tanzania possess radionuclides and heavy metals due to the enormous use of phosphates to boost the fertility of the farm soils. Their submission was in a paper review for different locations across the globe regarding the pollution and contamination of the environment by radioactive elements due to the use of phosphate rocks in farming and they came up with the mathematical modelling of the impacts of these contaminations on ecosystem. In this present study, assessment of the activity concentrations of $^{40}$K, $^{226}$Ra and $^{232}$Th has been carried out on a fertilized farm soil to estimate several radiological hazards attributes associated with environmental radioactivity in the farm.

2. Methodology

2.1 Study Area
The area of study is a farmland situated within the latitude $07^\circ 08.627^\prime$N - $07^\circ 08.949^\prime$N and longitude $003^\circ 10.214^\prime$E - $003^\circ 10.893^\prime$E in Abeokuta, southwestern Nigeria (Figure 1). The area is found in a sub-humid tropical region within Southwest Nigeria. The average annual precipitation of Abeokuta is 1,270 mm and its temperature is about 28 degree Celsius. The average yearly evaporation within this area is about 1,100 mm. The city is drained mainly by River Ogun in a general dendritic pattern. Abeokuta covers a geographical area of 1,256 square kilometre[14]. Figure 2 shows that the area lies geologically within the Eastern section of the Dahomey basin with east-westward trend sediments deposition and six lithostratigraphic units comprising Benin, Ilaro, Oshosun, Akinbo, Ewekoro and Abeokuta Formations from youngest to the oldest geological formation[14]. Abeokuta Formation denoted as Cretaceous has been classified as a Group divided into Araromi, Afowo and Ise Formations. Abeokuta Formation is of sequence of poorly sorted grits and pebbly sands with intercalations of siltstones, mudstones and shaly clay. Ewekoro Formation is known to be a Paleocene shallow marine deposit of non-crystalline and non-fossiliferous limestone strata. Akinbo shale units is of late Paleocene to Early Eocene overlaid by Eocene Shale of Oshosun Formation. Coarse sequence of estuarine, deltaic and continental sandy unit of Ilaro Formation overlie Oshosun Formation. Benin Formation is the youngest overlying the Ilaro Formation.

2.2 Samples Collection, Preparation and Radioactivity measurements
Seven soil samples were randomly obtained from the farmland. The sampling locations in the farmland and their corresponding geographical coordinates are presented in Table 1. The soils were scooped down to a depth of about 30 cm at each location point using hand trowel into a sealed polythene bags to prevent the samples from mixing up. In the laboratory, each soil sample was air dried, pulverized, homogenized and then sieved using a 2 mm mesh. A 0.2kg weight of sieved sample was poured into standard
container (plastic), tightened and sealed in order to prevent $^{220}\text{Rn}$ and $^{222}\text{Rn}$ gasses form escaping. The sealed samples were left for thirty days to allow for secular equilibrium between parent and daughter nuclei.

The radionuclides activity concentrations were measured using NaI (TI) detector-based gamma spectrometric system where the digiBASE system that combines a miniaturized preamplifier and detector with a powerful digital multichannel analyser and special features for fine time-resolution measurements. The digiBASE incorporates into the NaI (TI) detector provides a gain stabilizer to significantly reduce the sensitivity of the detector to changes in ambient temperature and magnetic fields. Three gamma ray lines of interest were 1460 keV, 1764 keV and 2615 keV which were resolved without much interference. The cylindrical plastic containers of radiation source were of diameters 7 cm. The seven soil samples each of mass 0.2 kg were dried, grinded and kept for more than thirty days in standard plastic containers to reach secular equilibrium were kept above the detector for counting process.

About 10800 seconds (3 hours) was set as the counting time, which is considered enough for the detector to be able to show clearly and be able to distinguish a desired peaks from a spectrum of signals. Multichannel analyser algorithm was used to compute the areas under each peak which represent the count number for a radionuclides in a particular sample. Uranium reference material termed RGU-1 from the International Atomic Energy Agency (IAEA) was used to calibrate the energy of the gamma spectrometer. The reference material was weighed into a standard cylindrical plastic sample container and placed on a NaI detector surface enclosed inside a lead shield of the spectrometer. This was counted for a lifetime of 10800 seconds. A spectrum was captured and specifically, three of the energy peaks identified on the spectrum were used in the energy calibration. Corresponding to the locations (channel numbers), the peaks of interest were: 295keV, 1120keV and 1765keV. In order to convert the count rate (cps) response of the spectrometer to desirable activity (Bq) for each of the three radio nuclides ($^{40}\text{K}$, $^{226}\text{Ra}$ and $^{232}\text{Th}$), the three reference materials RGK-1, RGU-1 and RGTh-1 from International Atomic Energy Agency (IAEA) were used. The $\gamma$ - ray lines of $^{214}\text{Bi}$ at 1764keV, $^{208}\text{TI}$ at 2014keV and 1460.8keV were used to determine the specific activity of $^{226}\text{Ra}$, $^{232}\text{Th}$ and $^{40}\text{K}$ respectively.

### 2.3 Estimation of Radiological Parameters

Radium equivalent activity is used in comparing the activity concentrations of the samples. It was used as a simple radiological index that considers radiation hazards connected to natural radioisotopes ($^{226}\text{Ra}$, $^{232}\text{Th}$ and $^{40}\text{K}$) in the fertilized farm soils. It also serves as a resourceful guide in providing the standard safety regulation regarding radiation protection for the farmers in particular and possibly the entire public near such farms. Hence, the radium equivalent activity ($\text{Ra}_{eq}$ in Bq·kg$^{-1}$) was taken as the sum of the weighted activities of $^{226}\text{Ra}$, $^{232}\text{Th}$ and $^{40}\text{K}$ assuming that 10 Bq·kg$^{-1}$ of $^{226}\text{Ra}$, 7 Bq·kg$^{-1}$ of $^{232}\text{Th}$ and 130 Bq·kg$^{-1}$ of $^{40}\text{K}$ give same $\gamma$ - ray dose rate. The mathematical relation that was used to obtain radium equivalent activity of each sample according to UNSCEAR [16] is given in Equation (1), where, $K_{Ra}$, $K_{Th}$ and $K_{K}$ are the specific activities or activity concentrations (in Bq·kg$^{-1}$ dry weight) of $^{226}\text{Ra}$, $^{232}\text{Th}$ and $^{40}\text{K}$, respectively.

$$\text{Ra}_{eq} = K_{Ra} + 1.43 K_{Th} + 0.077 K_{K}$$

The absorbed dose rate due to $\gamma$ - ray radiation in air (ADRA) at 1m (gonad height) away from the ground in order to ensure that the naturally occurring radionuclides are uniformly distributed was calculated by the relation in equation (2), where, ADRA is air absorbed dose rates; $K_{Ra}$, $K_{Th}$ and $K_{K}$ are the specific activities (Bq·kg$^{-1}$) of the radionuclides (radium, thorium and potassium) in the soils. Here,
0.462, 0.621 and 0.0417 conversion factors were used to convert the specific activities into the absorbed doses rate (nGy·h⁻¹·Bq⁻¹·kg⁻¹) based on guidelines given by United Nations Science Committee on the Effects of Atomic Radiation [16]. Determination of the external hazard index involves the use of equation (3) gives a value that quantifies the external hazard index, $H_{ex}$ [16]. To estimate annual effective dose equivalent (AEDE) rate otherwise called Outdoor annual dose equivalent, a mathematical relation as given by equation (4) is engaged [16].

$$ADRA \ (nGy/h^{-1}) = 0.462K_{Ra} + 0.621K_{Th} + 0.0417K_K$$

$$H_{ex} = \frac{K_{Ra}}{370} + \frac{K_{Th}}{259} + \frac{K_K}{4810} \leq 1$$

$$AEDE \ (mSv) = ADRA \times DCF \times OOF \times T$$

where,

- Dose Conversion Factor (DCF) is $0.7Sv \cdot Gy^{-1} \times 10^{-6}$;
- Outdoor Occupancy Factor (OOF) is 0.42; and
- $T$ is the time factor (24h $\times$ 365.25 = 8766 hours) [16].

The farmers spend more time on their farms; they spend about 10 hours of their day on the farm. This gave a factor of 10/24 = 0.42 for the outdoor occupancy factor [16].

3. Results and Discussion

The results presented (Table 2) shows the values of specific activities for the three primordial radionuclides detected. The mean activity concentrations obtained are: 261.29 ± 36.84 (K-40), 30.87 ± 6.81 (226Ra) and 47.10 ± 11.95 (232Th); the maximum values are 445.30 ± 33.46 (40K), 37.59 ± 9.07 (226Ra) and 70.64 ± 13.57 (232Th); the minimum values are 149.61 ± 11.89 (40K), 19.32 ± 9.93 (226Ra) and 34.19 ± 10.73 (232Th). Absorbed dose rate in air (ADRA), annual effective dose equivalent (AEDE), external radiological hazard index ($H_{ex}$), and radium equivalent activity ($Ra_{eq}$) were computed from the values of activity concentrations and also presented in Table 2. The mean laboratory ADRA was 36.69 nGy/h (0.026 µSv/h), maximum laboratory ADRA was 45.61 nGy/h (0.03 µSv/h) and minimum laboratory ADRA was 30.32 nGy/h (0.01 µSv/h). The mean AEDE is 94.56 µSv/y, maximum AEDE is 117.55 µSv/y and the minimum is 78.15 µSv/y. The mean external hazard index ($H_{ex}$) was 0.21. The maximum external hazard index was 0.25. The minimum external hazard index was 0.18. The mean radium equivalent activity ($Ra_{eq}$) was 78.14 Bq·kg⁻¹; the maximum ($Ra_{eq}$) value was 95.23 Bq·kg⁻¹ and the minimum ($Ra_{eq}$) value was 65.48 Bq·kg⁻¹. The specific activity of K-40 radionuclides at location L7 is more than the world average concentration of 420 Bq/kg. This is not unconnected to the fertilizer within that portion of the farm. The correlation plot in Figure 3 shows the correlation coefficient of 0.9975, implying that the results of the annual external dose equivalent are majorly cause by the natural radionuclides activity concentrations. The mean value of ADRA are less than the weighted mean dose rate of 59 nGy/h according to the United State Scientific Committee on the Effects of Atomic Radiations (UNSCEAR) [16]. Also, the AEDE values are observed to be lesser than the International Commission on Radiological Protection Publication (ICRP) [17] standard dose limit for members of public which is 1 mSv/y. The values of the external hazard index in the farmland are also well below unity.

4. Conclusions

The specific activity of natural radionuclides within the soil samples from a farmland in Abeokuta, Nigeria were obtained using a calibrated NaI-based gamma-spectrometer. Dose rate, annual effective dose, external hazard index and radium equivalent activity were estimated from the values of the activity concentrations. The ranges and means of measured activity concentrations were 149.61±11.89-445.30±33.46, 216.29±36.84; 19.32±9.93–37.59±9.07, 30.87±6.81 and 34.19±10.73-70.64±13.57,
47.10±11.95 Bq/Kg for $^{40}$K, $^{226}$Ra and $^{232}$Th respectively. The ranges and means of calculated dose rates, annual effective doses, external radiological hazard index and radium equivalent activities were 0.01-0.03, 0.026 µSv/h; 78.15-117.55, 94.56 µSv/y; 0.18-0.25, 0.21 and 65.48-95.23, 78.14 Bq/Kg respectively. The implications of the low values of these estimated radiological attributes within the farm is that the farmland is free of any radiation hazards and pose no threat to the farmers and the residents of that area. It is however recommended that organic manure should be used on the farmland rather than fertilizer and routine check on the farmland for environmental radioactivity should be done regularly for adequate monitoring.
Table 1: The Location points and their geographical coordinates.

| LOCATIONS | Latitude     | Longitude     |
|-----------|--------------|---------------|
| L1        | N 070 08.627' | E 003010.433' |
| L2        | N 070 08.708' | E 003010.454' |
| L3        | N 070 08.733' | E 003010.414' |
| L4        | N 070 08.723' | E 003010.416' |
| L5        | N 070 08.711' | E 003010.422' |
| L6        | N 070 08.736' | E 003010.400' |
| L7        | N 070 08.732' | E 003010.390' |

Table 2: Specific activity of $^{226}$Ra ($^{238}$U), $^{232}$Th and $^{40}$K, Absorbed Dose rate, Annual Effective Dose, External Hazard Index and Radium Equivalent Activity from the farmland.

| LOCATIONS | $^{40}$K | $^{226}$Ra | $^{232}$Th | ADRA (nGy/h) | ADRA (µSv/h) | AEDE (µSv/y) | Radium Eq. Activity (Bq/Kg) | H$_{EX}$ |
|-----------|---------|-----------|-----------|--------------|--------------|--------------|-------------------------------|---------|
| L1        | 175.34 ± 29.32 | 19.32 ± 9.93 | 35.89 ± 11.56 | 30.37 | 0.02 | 78.28 | 65.88 | 0.18 |
| L2        | 161.97 ± 33.17 | 19.72 ± 3.67 | 38.81 ± 14.63 | 30.32 | 0.02 | 78.15 | 65.48 | 0.18 |
| L3        | 149.61 ± 33.27 | 22.11 ± 8.45 | 34.19 ± 10.73 | 33.44 | 0.02 | 86.18 | 72.75 | 0.2 |
| L4        | 278.53 ± 43.98 | 38.49 ± 2.98 | 43.52 ± 16.19 | 37.25 | 0.03 | 96.01 | 78.43 | 0.21 |
| L5        | 319.91 ± 42.37 | 44.08 ± 11.01 | 70.64 ± 13.57 | 39.03 | 0.03 | 100.6 | 82.67 | 0.22 |
| L6        | 288.37 ± 42.37 | 37.59 ± 9.07 | 59.90 ± 6.02 | 40.8 | 0.03 | 105.1 | 86.54 | 0.23 |
| L7        | 445.30 ± 33.46 | 34.76 ± 2.58 | 46.72 ± 10.98 | 45.61 | 0.03 | 117.5 | 95.23 | 0.25 |
| MEAN      | 261.29 ± 36.84 | 30.87 ± 6.81 | 47.10 ± 11.95 | 36.69 | 0.026 | 94.56 | 78.14 | 0.21 |
| MAX       | 445.30 ± 33.46 | 37.59 ± 9.07 | 70.64 ± 13.57 | 45.61 | 0.03 | 117.5 | 95.23 | 0.18 |
| MIN       | 149.61 ± 33.27 | 19.32 ± 9.93 | 34.19 ± 10.73 | 30.32 | 0.02 | 78.15 | 65.48 | 0.25 |

ADRA = Absorbed Dose Rate in Air, AEDE = Annual effective Dose Equivalent, H$_{EX}$ = External Hazard Index
Figure 3: Correlation plot of $R_{aeq}(\text{Bq} \cdot \text{kg}^{-1})$ and AEDE ($\mu\text{Sv}/\text{y}$).

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