BERKELEY PUBLICATION
AND RESEARCH INTERNATIONAL

12.31.2022

Journal of Health, Metabolism & Nutrition Studies Vol. 20 No. 3

DETERMINATION OF RADIOACTIVITY LEVELS AND ITS POTENTIAL RISKS IN CHEMICAL FERTILIZERS USED IN NORTH-EASTERN NIGERIA

[1]BALA ABUBAKAR YUNUSA; [2]KABIRU DAFUWA; AND [3]SANI IDRIS

[1&3]Physics Department, School of Secondary Education (Science), Aminu Saleh College of Education, Azare Bauchi State. [2]School of Educational Services, Aminu Saleh College of Education, Azare Bauchi State.

ABSTRACT

This work aims to determine radioactivity levels and its potential risks in chemical fertilizers used in North-eastern Nigeria. The specific activities of ^{238}U, ^{232}Th and ^{40}K will be measured by using a gamma ray spectrometer based on a NaI(Tl) detector. The radium equivalent activity (Bq/kg) of different samples of NPK and urea fertilizers used in the study area will also be calculated. Both specific activities and radium equivalent activity af the fertilizer samples will be compared in order to determine the type of fertilizer with higher radioactivity. The specific activity and absorbed dose rate at 1m above the ground surface (nGy/h) after NPK fertilizers agricultural applications will be calculated and the maximum value of dose rate will be obtained and compared with the world average outdoor exposure due to terrestrial

Introduction:

Application of varieties of chemical fertilisers in agricultural sector for maximising crop yield has become common practice nowadays. In Nigeria, farmers make use of fertilisers to replenish the natural nutrients depleted from soil due to over cultivation of crops, weathering and erosion of land (Jibiri & Fasae, 2011). Fertilizer increases efficiency and obtains better quality of product recovery in agricultural activities. More than 30 million tons of phosphate fertilizers are annually consumed worldwide, which increase crop production and land

ISSN: 1213-4406

gamma radiation. All the aforementioned parameters will be calculated and analysed using the appropriate equations and data obtained will be presented in tables. Finally recommendation will be given.

Keywords: Radioactivity, chemical fertilizer, Risk, Radionuclide, Radiation

reclamation. Chemical fertilizers are chemical compounds that provide necessary chemical elements and nutrients to the plants and mainly contain phosphate, nitrate, ammonium and potassium salts. Phosphate rocks together with potassium ores and nitrogenous compounds are the main raw materials used for fertilizers in industrial production (Jankovic, et al., 2013). A possible negative effect of chemical fertilizers is contaminating the cultivated lands by trace metals and some naturally occurring radioactive materials (NORM). Radionuclides present in phosphate fertilizers affect the common people and farmers immensely. The large concentration of natural radionuclides in the fertilizers contaminates the environment and agricultural field. Direct inhalation of dust of phosphate fertilizers could affect the farmers on agricultural land (Hussain & Hussain, 2011).

Radioactivity is the spontaneous emission of energy in the form of particles or waves (electromagnetic radiation), or both, from the atomic nucleus of certain elements. It was discovered in 1896 by Henri Becquerel (a French Physicist) who observed that uranium emitted penetrating rays continuously and without initiation. This pioneering work was followed by Pierre and Marie Curie shortly afterwards. They proved that the radioactivity of uranium was an atomic property and not a chemical one (Abiye, 2005). Long-live radioactive elements such as uranium (^{238}U), thorium (^{232}Th) and potassium (^{40}K) and any of their decay products, such as radium and radon are examples of naturally occurring radioactive materials (NORM) (Zaim. & Atlas, 2016). Natural radioactivity is wide spread in the Earth's environment and it exists in various geological formations like soils, rocks, plants, sand, water, air and building materials. Hence, humans should beware of their natural environment with regard to the radiation health effects (chronic lung diseases, cancers etc.) (Mir & Rather, 2015).

While, radiation can be defined as the emission and propagation of energy in the form of rays or waves from the atoms and molecules of a radioactive substance as a result of nuclear decay. It can be classified into two main types namely ionizing and non-ionizing. Ionizing radiation is the type of radiation capable of ionizing an atom, and ionization usually occurs when one of the orbital electrons of an atom has been completely removed from it. The resultant effect of this is that the residual atom becomes positively charged (positive ion or cation), and the freed electron becomes negatively charged (negative ion or anion). The effects of radiation may be divided generally into four types. They include acute somatic effects, developmental effects, genetic effects, and late somatic effects (Noz and Maguire, 1979). Acute somatic effects occur in individuals within days or weeks of their exposure. It include injuries to the brain which causes delirium and convulsions; damages to the eye lens resulting in cataracts; gastrointestinal tract injury resulting into nausea and vomiting; sterility if ovaries or testes are damaged; and damages to the bone marrow which affects the body's ability to fight infection. Depending upon the degree of irradiation however, certain tissues such as the bone marrow, the intestinal lining, or the skin, may be able to replace cells killed by radiation within a few days. Some damages are however permanent above a certain dose level (UNSCEAR, 1982).

Based on the issues discussed, this study is set to investigate radioactivity levels and Potential risks of Chemical Fertilizers Used in North-eastern Nigeria.

Problem Statement/ Justification

Radioactivity of phosphate rocks leading to health problems from radiation at the level of the industrial processes which involves mining and transportation of phosphate ores and production of fertilizers. Relatively large concentrations of natural radionuclides present in phosphate fertilizers contaminate the environment and agricultural lands during cultivation. At the usage level, when fertilizers dispersed into the geo and biospheres, have a potential to transfer to living beings. Leaching of the minerals and wastes is another potential source of radioactivity dissemination which may contribute to enhanced exposure of workers, public and the environment to these radionuclides (Jankovic, et al., 2013; Sahu, Ajmal, Bhangare, Tiwari, & Pandit, 2014). The uptake and distribution of radionuclides in soil depends on several factors such as soil pH, type and amount of clay, exchangeable Ca and K and organic matter contents,

ISSN: 1213-4406

physiochemical properties of the radionuclide, type of crop (crop species and variety, and cultural practices), fertilizer application, irrigation, plowing, liming and climate conditions (Bramki, Ramdhane, & Benrachi, 2018).

A possible negative effect of chemical fertilizers is contaminating the cultivated lands by trace metals and some naturally occurring radioactive materials. The large concentration of natural radionuclides in the fertilizers contaminates the environment and agricultural field. Direct inhalation of dust of phosphate fertilizers could affect the farmers on agricultural land. The main reason for the high radioactivity in the leafy fertilizers was ^{40}K. Chemical fertilizers caused increase in annual exposure dose by no more than 0.15% which should be taken into account at the continuing use of chemical fertilizers (Hussain & Hussain, 2011). The fertilizers which are all water soluble substances dissolve instantly in water. These dissolved radionuclides, taken up by aquatic foods and fishes, enter into the human body through vegetables, food grains, fruits, aquatic food and fishes, and through the meat and milk of pastured animals (Alam, ve diğerleri, 1997; Uosif, Mostafa, Elsaman, & Moustafa, 2014). Extended use of phosphate fertilizer raises the dose rate in air above the ground by about 0.8 nGy/h. This may give rise to an additional exposure of man at continuous residence on such previously contaminated farm land of 0.007 mSv/y.

Soil not only acts as a source of continuous radiation exposure to humans but also as a medium of migration for transfer of radionuclides to biological systems, which can cause harmful biological effects such as DNA damage and cancer. Despite a well-known effect of cancer, scientists have long known that ionizing radiations with high doses may also cause mental retardations in the children of mothers exposed to radiations during the pregnancy period. All types of food including wheat and apples contain a detectable amount of radioactivity which successively relocates into the human body via the ingestion pathway. The activity of food is strictly linked to the activity of the soil where the food was grown. Knowledge of the concentration and the distribution of the radionuclides in these materials is very important because they provide useful information for the monitoring of environmental contamination by natural radioactivity (Bramki, Ramdhane, & Benrachi, 2018). Radionuclides enter the human body in ways of direct inhalation of airborne particulates, ingestion through the mouth,

Vol. 20, No. 3

International Journal of Health, Metabolism and Nutrition Studies

and entry through the skin. The ultimate state of the radionuclides depends upon their chemical and physical form. After uniform distribution, some radionuclides irradiate the entire body at the same rate (Ghosh, Deb, Bera, Sengupta, & Patra, 2008). While much is known to have been published on the geology, structures and mineralization in Northeast, Nigeria, only very little is known about the extent of radioactivity and its Potential risks of Chemical Fertilizers Used in the area. Therefore, there is need to determine the radioactivity levels and Potential risks of Chemical Fertilizers Used in North-eastern Nigeria.

Objectives Of The Study

The aim and objectives of this study is to determine the radioactivity levels and Potential risks of Chemical Fertilizers Used in North-eastern Nigeria

The specific objectives of the study are to:

1. determine Specific activities of ^{238}U, ^{232}Th and ^{40}K in different samples of granule and leafy NPK fertilizers used in the area using a Gamma ray spectrometer.
2. determine Specific activities of ^{238}U, ^{232}Th and ^{40}K in different samples of the Urea fertilizer used in the area using a Gamma ray spectrometer.
3. Calculate the radium equivalent activity (Bq/kg) of different samples of NPK and urea fertilizer used in the area.
4. obtain the absorbed dose rate at 1m above the ground surface (nGy/h) after NPK agricultural application.

Methods and Procedure

Study Area

The area is located between latitude 6° 26′–13° 45′N and longitude 8° 42′–14°39′E. It comprises of Borno, Yobe, Bauchi, Gombe, Adamawa and Taraba states (figure 1). It covers an area of 262,578 Km^2 and has a population of 18,984,299 persons according to 2006 Nigerian population census. It is characterized by relatively high temperatures throughout the year with the annual average varying from 28.32 °C in Yola to 25.92 °C in Bauchi while rainfall ranges between 467 mm at Nguru to 1091 mm at Ibi (Soltan, Attia, & Usman, 2017).

Figure 1: location of the study area

Sample Collection and Preparation

In this study, a total of eight (8) chemical fertilizer samples (four (4) NPK granular type, two (2) NPK leafy type and two (2) urea type) were collected from the local retail markets in the commercial city centres in six states from the north-eastern part of Nigeria.

The fertiliser samples were oven dried at 100°C until there were no detectable change in the mass of the samples. The dried samples were thoroughly crushed and passed through a 2-mm sieve. One kilogram from each sample was packed and sealed in a beaker of one liter. The measurements were performed five weeks later to ensure the secular equilibrium (Hamby and Tynybekov, 2002; Vassas et al, 2006; El-Zakla et al, 2007). The specific activities were performed by gamma rays spectrometry, employing a NaI(Tl) (BI CRON) of 2"×2" crystal dimensions detector. The resolution of the system was noted. The counting efficiency of the system was measured by using standard sources. Uranium-238 (^{238}U) was detected by the 1.76 MeV energy that correspond to the absorption of ^{214}Bi of ^{238}U series and was used to identify and quantify the natural uranium. Natural thorium-232 (^{232}Th) was identified by the ^{208}Tl peak which correspond to the 2.62 MeV absorption energy (Becegato, 2008). Potassium-40 (^{40}K) was identified and quantified by means of the absorption of the 1.46 MeV energy which correspond to a sole natural isotope ^{40}K. In order to subtract the background from each measurement, an empty beaker (with the same geometry) was measured. The accumulation time for each sample was recorded just to obtain gamma spectrum with appropriate to the detector statistics.

Data Analysis

The row data obtained under the method were presented in the tables, processed and analyzed using the established mathematical equations described below:

The specific activity of the radionuclides (^{238}U, ^{232}Th, and ^{40}K)

The specific activity of ^{238}U, ^{232}Th, and ^{40}K were calculated using the following equation :

$$Specific\ activity = \frac{\text{CPS x 100 x 100}}{\text{Eff x B. I. x m}} \pm \frac{\text{SD}_{\text{CPS}}\text{ x 100 x 100}}{\text{Eff x B. I. x m}}$$

Where, CPS = Net count rate per second, B.I. = Branching Intensity, Eff = Efficiency of the detector, m = Sample mass in kg and SDCPS = Standard Deviation of net count rate per second.

Radium equivalent activity

From the radiation point of view, the high content of fertilizers was significant in the accumulation of large quantities of chemical fertilizers in the warehouse, which increased the Radon concentration in the surrounding air. The widely used radiation hazard index Ra_{eq} is called the radium equivalent activity; the radiation equivalent activity is a weight sum of activities of the three natural radionuclides ^{238}U, ^{232}Th and ^{40}K based on the estimation that 10 Bq/kg of 226Ra, 7 Bq/kg of

^{232}Th and 130 Bq/kg of ^{40}K produce the same gamma ray dose rate (Beretka and Matthew, 1985). The radium equivalent activity was calculated by using the following relation.

$$Ra_{eq} = C_U + 1.43C_{Th} + 0.077C_K$$

Where C_U, C_{Th} and C_K are the specific activities of ^{238}U, ^{232}Th and ^{40}K in (Bq/kg) unit respectively.

Results and Discussion

The spectra of 8 chemical fertilizers samples were analyzed. The results of specific activities of ^{238}U, ^{232}Th and ^{40}K and radium equivalent activity (Raeq) in different samples (four NPK granular types, two NPK leafy types and two urea types) are presented in Table 1.

Table 1 - Specific activities of 238U, 232Th and 40K and radium equivalent activity (Bq/kg) in NPK and Urea fertilizers.

Type	Sample	U-238	Th-232	K-40	Raeq
Granule Fertilizers	NPK1	11.816±1.249	1.204±0.184	759.649±9.246	63.127±2.158
	NPK2	< dL	< dL	< dL	< dL
	NPK3	72.435±2.017	3.482±0.251	102.683±3.766	4.341±0.180
	NPK4	< dL	2.614±0.141	854.648±10.861	84.163±2.234
	UREA1	< dL	< dL	< dL	< dL
	UREA2	< dL	< dL	< dL	< dL
Leafy Fertilizers	NPK5	< dL	< dL	2154.293±15.181	145.010±1.131
	NPK6	< Dl	< dL	1690.823±12.218	123.271±1.182

dL: detection limit, 2.8, 1.16 and 6.0 Bq/kg for ^{238}U, ^{232}Th and ^{40}K respectively.

For ^{238}U radionuclide, only two samples (NPK1 and NPK3) had specific activities values of (11.816 and 72.435 Bq/kg, respectively). The value for NPK3 was higher than the world average of 41 Bq/kg (UNSCEAR, 1983). Such result could be explained in a high concentration of ^{238}U in Phosphate rocks which was used

Vol. 20, No. 3

International Journal of Health, Metabolism and Nutrition Studies

widely as raw material for manufacturing the NPK chemical fertilizers. The leafy NPK fertilizer samples (NPK5 and NPK6) had no ^{238}U specific activity. Of all 6 NPK samples, only three samples (NPK1, NPK3 and NPK4) showed thorium with specific activities of 1.204, 3.482 and 2.614 Bq/kg respectively, which were less than the world average of 52.2 Bq/kg (UNSCEAR,1983).

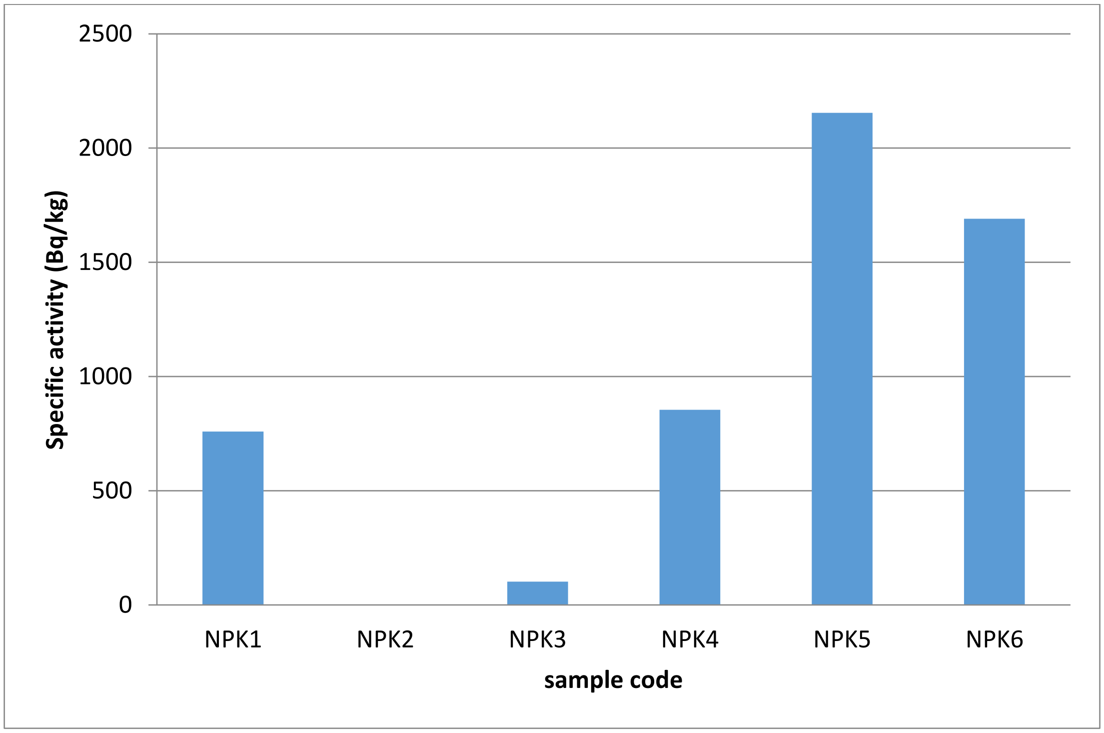

Figure 1 - Specific activities of ^{40}K (Bq/kg) in NPK granular and leafy fertilizers.

The ^{40}K radionuclide was present in all the NPK samples, except NPK2. The highest value of 2154.293Bq/kg was in NPK5 sample, while the lowest value of 102.683Bq/kg was in NPK3 sample. Table 1 indicated that the specific activities of ^{40}K radionuclides in leafy NPK fertilizer types were higher than that in the granular NPK fertilizer (as shown in Fig. 1). These values, except two, were also higher than the world average which was 230 Bq/kg for (UNSCEAR, 1983). The NPK2 sample had no activity detected due to the lack of concentrations of radionuclides in its raw materials. For urea samples, no activity was found in urea fertilizers because their ores did not contain natural radionuclides.

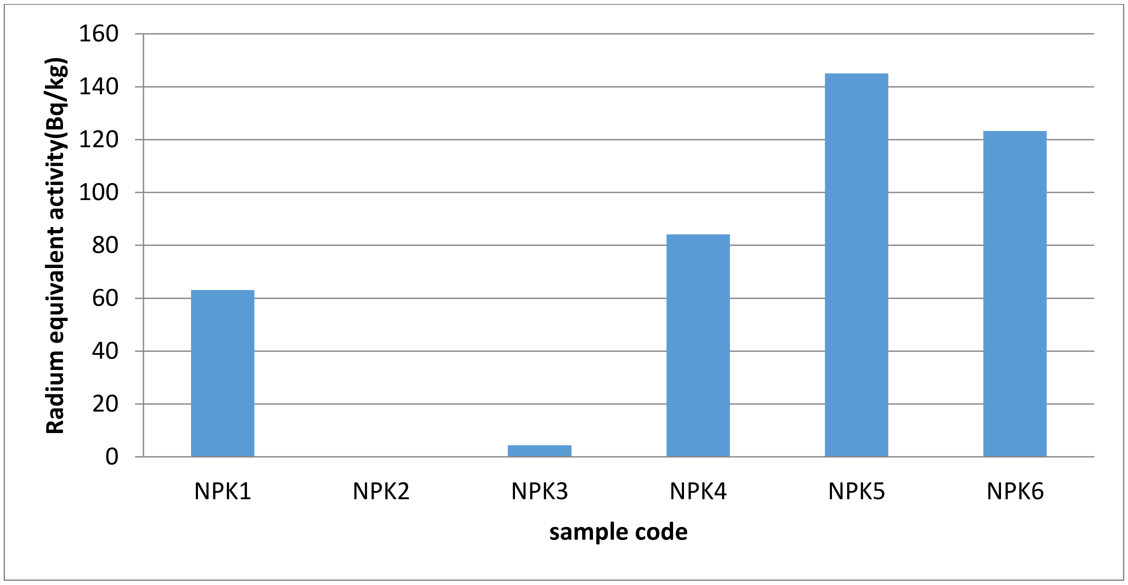

Figure 2 - Radium equivalent activity (Bq/kg) for NPK granular and leafy fertilizers.

Table 1 showed that the radium equivalent activity in the investigated samples ranged from 4.341 Bq/kg in NPK3 to 145.010 Bq/kg in NPK5 sample. The highest radium equivalent activity value was less than the world average (UNSCEAR, 2000). In general, radium equivalent activity in the leafy NPK fertilizers was higher than that in the granular NPK fertilizer (Fig. 2). This difference in radium equivalent was due to the high content of ^{40}K.

Table 2 - Annual additional increment (Bq/kg) of ^{238}U, ^{232}Th and ^{40}K, and calculated absorbed dose rate 1m above the ground surface (nGy/h) originated from NPK fertilizers agricultural applications.

Sample Code	Surface activity(Bq/kg)^a			Dose rate (nGy/h)^b
	U-238	**Th-232**	**K-40**	
NPK1	0.009	0.001	0.514	0.02108
NPK2	0.112	0.011	0.064	0.05267
NPK3	0.051	0.003	0.483	0.04672
NPK4	0.010	0.002	0.612	0.02512
UREA1	-	-	-	-
UREA2	-	-	-	-

NPK5	-	-	1.635	0.06164
NPK6	-	-	1.273	0.04564

[a]In the top 5 cm of the soil that has average density value of 1.5 (75 kg soil weight per 0.05 m3 volume).
[b]Estimated using dose rate conversion factors (DRCF) = 0.462, 0.604 and 0.0417 nGy/h per Bq/kg to ^{238}U, ^{232}Th
and ^{40}K, respectively (UNSCEAR, 2000).

The external exposure due to radionuclides in the chemical fertilizers used in the agricultural area was proportional to the intensity of fertilizers application. Based on this application rate, the annual addition of natural specific activities distributed per unit mass (Bq/kg) corresponded to 0.009-0.112, 0.001 - 0.011 and 0.064 - 1.635 Bq/kg for ^{238}U, ^{232}Th and ^{40}K, respectively (Table 2).

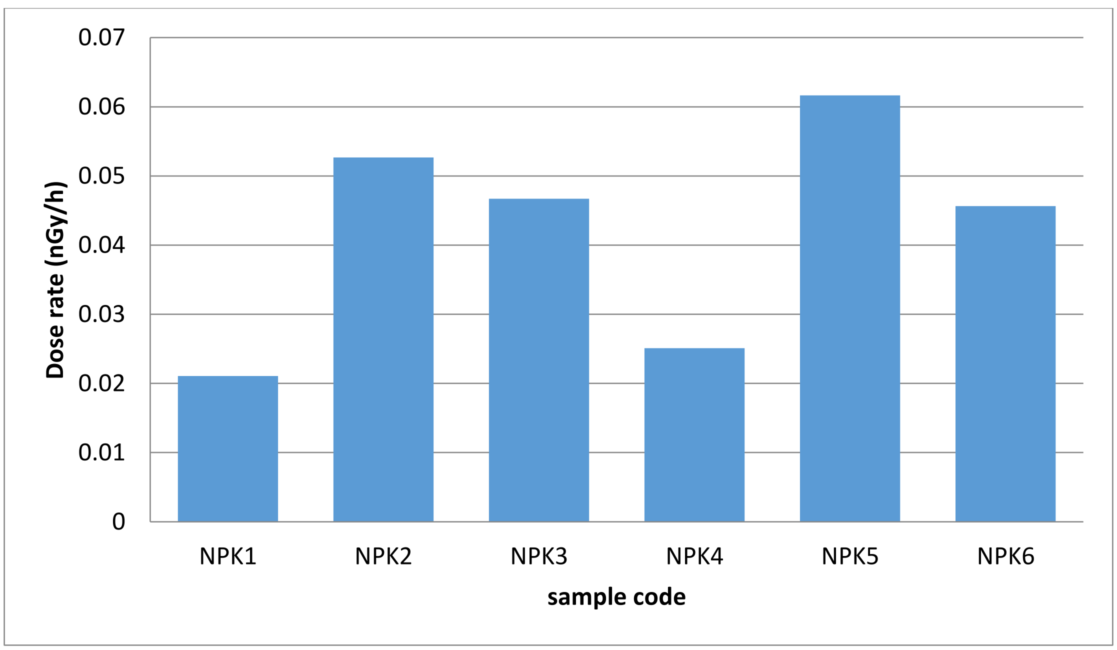

Figure 3 - Absorbed dose rate 1 m above the ground surface (nGy/h) originated from NPK fertilizers agricultural applications.

The increase in the external radiation exposure due to the application of granule and phosphate fertilizers ranged from 0.02108 to 0.06164 nGy/h (Fig. 3). The maximum value represented 0.12% of the world average outdoor exposure due to terrestrial gamma radiation (51 nGy/h) (UNSCEAR, 2000). The results showed a wide variation in gamma activity in NPK fertilizers. This could be attributed due to: the difference in the factories of manufactured fertilizers; the difference in the places from which the raw minerals for manufacturing the fertilizers were taken and the raw materials contained radioactive elements different properties depending on local geology.

CONCLUSION

From the results, it could be concluded that.

1. The high content of uranium radionuclide in the granular NPK fertilizer commonly used in the north-east Nigeria must be taken into account.
2. The main reason for the high radioactivity in the leafy fertilizers was ^{40}K.
3. The urea fertilizers did not cause any effect on human health from the viewpoint of biological effect of radiation.
4. Chemical fertilizers caused increase in annual exposure dose by no more than 0.12% which should be taken into account at the continuing use of chemical fertilizers.
5. The radium equivalent activity values were within the worldwide average, in spite of the high specific activity of some nuclei in chemical fertilizer,

References

Abiye, O. S. (2005). A Study of Natural Radiation Levels and Distribution of Dose Rates Within The Younger Granite Province of Nigeria. Unpublished Phd Thesis. University of Jos.

Alam, M. N., Chowdhury, M. I., Kamal, M., Chose, S., Banu, H., & Chakraborty, D. (1997). Radioactivity in Chemical Fertilizers Used in Bangladesh. *Appl. Radiat. Isot., 48*(8), 1165-1168.

Beretka, J., & Matthew, P.J. (1985), Natural radioactivity of Australian building materials, industrial wastes and by-products. *Health Phys.*, 48 (1), 87-95.

Bramki, A., Ramdhane, M., & Benrachi, F. (2018). Natural radioelement concentrations in fertilizers and the soil of the Mila region of Algeria. *Journal of Radiation Research and Applied Sciences*(11), 49-55.

El-Zakla, T., Abdel-Ghny, H.A., Hassan, A.M. (2007), Natural radioactivity of some local fertilizers. *Rom. J. Phys.*, 52 (5-7), 731-739.

Ghosh, D., Deb, A., Bera, S., Sengupta, R., & Patra, K. (2008). Measurement of natural radioactivity in chemical fertilizer and agricultural soil: evidence of high alpha activity. *Environ Geochem Health*(30), 79–86.

Hamby, D.M., Tynybekov, A.K. (2002), Uranium, Thorium, and Potassium in Soils Along the Shore of Lake Issyk-Kyol in the Kyrghyz Republic. *Environ. Monit. Assess.*, 73 (2), 101-108.

Hussain, R. O. & Hussain, H. H. (2011).Investigation the Natural Radioactivity in Local and Imported Chemical Fertilizers. *An international journal,(54), 4*: pp. 777-782,

Hussain, R., & Hussain, H. (2011). Investigation the Natural Radioactivity in Local and Imported Chemical Fertilizers. *Brazilian Archives of Biology and Technology, 54*(4), 777-782.

Jankovic, M., Nikolic, J., Pantelic, G., Rajacic, M., Sarap, N., & Todorovic, D. (2013). Radioactivity in Chemical Fertilizers. *Proceedings of 9th Symposium of the Croatian Radiation Protection Association*, 578

Jibiri, N. N. & Fasae, K. P. (2011). Activity Concentrations of ^{226}ra, ^{232}th And ^{40}k In Brands of Fertilisers Used In Nigeria. *Radiation Protection Dosimetry*, *Vol. 148, No. 1*, pp. 132–137 doi:10.1093/rpd/ncq589

Mir, F., & Rather, S. (2015). Measurement of radioactive nuclides present in soil samples of district Ganderbal of Kashmir Province for radiation safety purposes. *Journal of Radiation Research and Applied Sciences* (8), 155-159.

Noz, M. E. and Maguire, G. Q. (1979). Radiation Protetion in the Radiologic and Health Sciences. Henry Kimpton Publishers. London 211p

Sahu, S., Ajmal, P., Bhangare, R., Tiwari, M., & Pandit, G. (2014). Natural radioactivity assessment of a phosphate fertilizer plant area. *Journal of Radiation Research and Applied Sciences*(7), 123-128.

Soltan, F. H., Attia, M. E. & Usman, S. H. (2017).Trends of Rainfall and Temperature over North-Eastern Nigeria (1949-2014). *Journal of Environmental Science, Toxicology and Food Technology, (11) 5. PP 01-09*

UNSCEAR, (1983), *Exposure from Natural Sources.*United Nations Scientific Committee on the Effects of Atomic Radiation. Report to General Assembly, United Nations, New York.

UNSCEAR, (1993), *Sources and Effects of Ionizing Radiation*. United Nations Scientific Committee on the Effects of Atomic Radiation. Report to General Assembly, United Nations, New York.

UNSCEAR, (2000), *Exposure from Natural Sources*. United Nations Scientific Committee on the Effects of Atomic Radiation. Report to General Assembly, United Nations, New York.

Uosif, M., Mostafa, A., Elsaman, R., & Moustafa, E. (2014). Natural radioactivity levels and radiological hazards indices of chemical fertilizers commonly used in Upper Egypt. *Journal of Radiation Research and Applied Sciences*(7), 430-437.

Vassas, C., Pourcelot, L., Vella, C., Carpéna, J., Pupin, J.P., Bouisset, P., & Guillot, L. (2006). Mechanisms ofenrichment of natural radioactivity along the beaches of the Camargue, France. *J. Environ. Radioactiv.*, 91 (3), 146-159.

Zaim., N., & Atlas, H. (2016). Assessment of radioactivity levels and radiation hazards using gamma spectrometry in soil samples of Edirne. *Turkey. J Radioanal Nucl Chem*. doi:DOI10.1007/s10967-016-4908-0