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Natural radioactivity and environmental risk assessment of Sokoto phosphate rock, Northwest Nigeria

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Phosphate rock samples collected from the phosphate bearing formations of Sokoto State, Northwest Nigeria were assessed for their radiological contents by gamma spectrometric technique (sodium iodide (NaI (TI) detector). The mean activity concentrations of ^{226}Ra , ^{232}Th and ^{40}K in all the studied samples were $720.1 \pm 4.2 \text{ Bqkg}^{-1}$, $33.5 \pm 1.4 \text{ Bqkg}^{-1}$, and $315.3 \pm 6.7 \text{ Bqkg}^{-1}$ respectively. Contributions from radium contents to the overall radionuclide activity concentrations were quite significant. The calculated mean value of the absorbed dose rate was found to be 3.6 times the maximum value for the world background level, which shows the severity of radiation risk the public is exposed to in the study areas. The computed values for radiation hazard indices were above the world's proposed safety limits by UNSCEAR, which place the study areas among high background radiation zones. Some restrictive measures and precautionary actions are therefore obligatory for the local farmers and the general public from the view point of radiation protection.

Key words: Natural radioactivity, Sokoto, phosphate rock, radiation hazard, NaI (TI) detector, radiation dose.

INTRODUCTION

Phosphate rocks, which can be of sedimentary, volcanic or biological origin (Sam and Holm, 1995), generally refer to rocks with high concentration of phosphate minerals (Cevik et al., 2010). Phosphate rocks are the starting raw material for all phosphate products and fertilizer production (Khater et al., 2001; Ogunleye et al., 2002). They are also used in the preparation of food and animal feeds, and as a source of phosphorus for agricultural purposes as well as for the manufacturing of certain industrial phosphorus-based chemicals and detergents

(Abbady et al., 2005; Khan et al., 1998). Phosphate rocks contain relatively high amounts of naturally occurring radioactive materials (NORMs) from ^{238}U and ^{232}Th and their respective descendants, and ^{40}K in concentrations that depend on geographical and geological origin. Though the concentrations of NORMs observed in phosphate rocks of all types are similar to those observed normally in soils, uranium and its decay products tends to occur at elevated concentrations in sedimentary phosphates (Harb et al., 2008; Khater et al., 2001). This

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uranium enrichment makes phosphates one of the major sources of technologically enhanced natural radiation (TENORM), (Abbady et al., 2005), which contributes to human exposure. It is widely believed that the radioactivity associated with phosphate rocks of sedimentary origin is formed by the adsorption and co-precipitation of uranium with calcium (Khater et al., 2001). Phosphate ores are found to occur as phosphorites, $[\text{Ca}_3(\text{PO}_4)_2]$, which are very old marine deposits associated with fossils; and apatite, $\text{Ca}_5[(\text{PO}_4)_3(\text{F})]$, which is of igneous origin (Schmidt, 1993; OSPARCOM, 1997; Abbady et al., 2005). van Straaten (2002), reported that sedimentary marine phosphate rock deposits accounts for about 75% of the world's phosphate source, which according to Okosun (1989), makes it a great economic base that provides most of the raw materials for the manufacture of phosphate fertilizers and phosphorus based chemicals

Mining and processing of phosphate rock and its use in the manufacturing industry, redistributes the radioactive trace elements and ^{238}U and its decay products throughout the environment and also in the final products, thereby enhancing the concentrations of natural radionuclides in the end products and wastes of the phosphate industry. This constitutes ways in which the workers, the public and the environment are exposed to enhanced radioactivity (IAEA, 2004; UNSCEAR, 1993). Significant radiological investigations have been conducted on phosphate rocks and their products in different countries of the world, which has led to establishment of standards and regulations for the protection of man and his environment from the radiation effects that will arise due to the usage of the rock. Sam et al. (1999) assessed the radiological and chemical constituents of Uro and Kurun rock phosphates from Sudan. Khater et al. (2001), investigated the radiological impacts of natural radioactivity in Abu-Tartor phosphate deposits, Egypt. Saueia et al. (2005) undertook similar investigation for phosphate rock, phosphogypsum and phosphate fertilizers in Brazil. Abbady et al. (2005), studied the natural radioactivity and dose assessment for phosphate rocks from Wadi El-Mashash and El-Mahamid Mines in Egypt. Cevik et al. (2010), reported the radiological, structural and chemical characteristics of Mardin-Mazidagi phosphate rock in Turkey.

As a result of increasing interest by the Nigerian local farmers in direct application of phosphate rocks to agricultural soils as a substitute for and ready alternative to chemical fertilizer, which is becoming increasingly expensive and out of reach, it becomes important to assess the potential radioactivity exposures of phosphate rocks.

In the present work, gamma spectrometric technique is used to quantify the activity concentrations of ^{226}Ra , ^{232}Th and ^{40}K of Sokoto phosphate rocks. The human radiation exposure from the radioactive content of phosphate rock

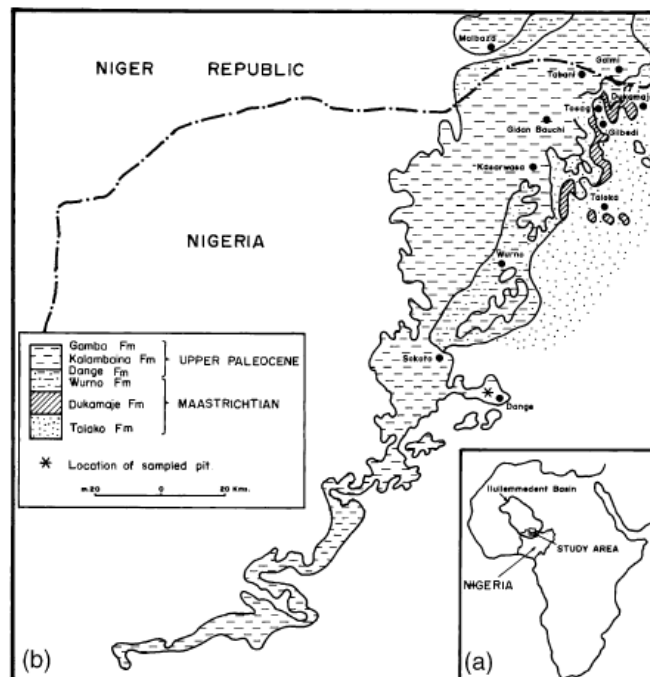


Figure 1. (a) Map of Africa, showing the Sokoto sub-basin, Northwest Nigeria in relation to the Illelmeden basin. Inset: Location of (b). (b) Simplified geological map of the southeastern sector marine and marginal facies of Illelmeden basin, Sokoto sub-basin (Okosun, 1997).

samples was estimated, along with possible environmental implications of direct application of Sokoto phosphate rocks for agricultural purposes.

Sokoto phosphate deposit

The Sokoto basin, Northwest Nigeria is Nigeria's sector of extensive Illelmeden Basin (Figure 1). A phosphate deposit of great significance is located in Sokoto state, northwestern Nigeria, in a paleocene sedimentary sequence. Marine sediments were deposited in Sokoto basin during the maastrichtian-paleocene trans-Saharan transgressions.

The phosphates, whose basic components are from vertebrate remains, are present in the Dange formation which from Paleocene age occur as nodular beds and nodular disseminations in the shales and siltstone of the formation. The Dange formation is phosphate bearing in the entire basin though the concentration differs from place to place. Phosphate rocks are found in commercial deposits in Dukamaje, Dange, Gilbadi, Dankilo-Dange, Wurno, Kindiru, Dillingu and Gada in Sokoto State. Ogunleye et al. (2002) also reported occurrence of phosphate nodules and pellets in Gidan Bauchi, Illela and Kalambiana. Etu-Efeotor (1998), Okosun (1997) and Ogunleye et al. (2002), reported that phosphate deposit thickness in Dukamaje formation range between 1 to 5 m,

Table 1. Mean activity concentrations of ^{226}Ra , ^{232}Th and ^{40}K (Bqkg $^{-1}$) in Sokoto phosphate rock samples.

Sample ID	Number of samples	Activity concentrations (Bqkg $^{-1}$)		
		^{226}Ra	^{232}Th	^{40}K
DNG	3	650.7±3.8 (602.5 - 732.0)	35.7±1.2 (17.0 - 58.6)	264.3±6.4 (111.2 - 392.3)
KDR	3	1073.2±5.9 (1055.7 - 1087.3)	55.9±1.7 (39.2 - 85.3)	380.3±8.3 (310.3 - 459.0)
DLG	3	804.5±3.7 (854.7 - 883.6)	28.9±1.0 (17.7 - 40.8)	394.6±5.6 (375.8 - 428.4)
WRN	3	510.1±4.1 (443.7 - 617.8)	19.5±1.8 (13.2 - 27.7)	272.2±7.7 (202.7 - 362.8)
DNK	3	562.0±3.3 (491.0 - 606.3)	27.4±1.3 (24.7 - 32.2)	265.2±5.3 (110.7 - 404.7)
Av. range	15	720.1±4.2 (443.7 - 1087.3)	33.5±1.4 (13.2 - 85.3)	315.3±6.7 (111.2 - 459.0)

while the phosphate nodules/pellets occur in sizes of 0.1-1cm. Amapu (1998), investigated the agronomic potential of direct application of Sokoto phosphate rock as an alternative phosphate fertilizer for Nigeria's sub-humid Savannah region. Phosphate nodules from Sokoto basin are presently mined superficially by the natives especially women and children during the non-farming season and transported, as a major raw material to feed the Crystal Talc Nigeria, Kagara, Niger State and Federal Superphosphate Fertilizer company plant at Kaduna, Kaduna State, for the production of phosphate fertilizer. They are also blended by the locals and applied directly in the agricultural farms as substitute for fertilizer.

MATERIALS AND METHODS

The Sokoto phosphate deposits occur generally as nodules on the surface, with nodular beds measuring to between 0.2-0.3 m. The phosphate nodules are superficially mined on daily basis mostly by women and children. To measure the natural radioactivity, a total of 15 phosphate rock samples, three each, were collected from five villages which represents the Sokoto phosphate bearing formations. The villages are Dange (DNG), Dankilo (DNK), Wurno (WRN), Kindiru (KDR) and Dillingu (DLG). Phosphate nodules were picked at random from four different points and mixed together thoroughly to give a true representation of each sampling point throughout all the sample locations. The samples, each about 1 kg in weight, were packed in well labelled sampling polyethylene bags, properly tied and sealed to avoid cross contamination of samples, and transported to the laboratory for analysis at the Center for Energy Research and Training (CERT), Ahmadu Bello University, Zaria, Kaduna State.

In the laboratory, the samples were opened and all weeds, stones and other organic and non-phosphate materials, were picked out. The samples were sundried under ambient temperature to almost constant weight for 72 h. The dried samples were crushed into fine powder and sieved through a 2-mm mesh. The dried samples were homogenized, weighed and packed into identical 7.2 cm diameter and 6.0 cm height radon impermeable marinelli beakers which holds between 300-350g of sample. The beakers were then subjected to a triple stage sealing process to ensure the containment of radon-222 (Jamok, 2014), and stored at room temperature for a period of 5 weeks to allow for radioactive equilibrium between the parents and their daughter nuclides (Amin et al., 2013; Ghose et al., 2012; Ibeanu, 2002; Kumar et al., 2003).

The radiometric analysis of the samples was carried out using a gamma spectrometric technique comprising a 7.62 x 7.62 cm NaI

(TI) detector coupled to a computer based multichannel analyzer (MCA) for data acquisition. To effectively reduce the gamma-ray background radiation from the acquired spectrum, the detector is housed in a 6 cm thick lead shield lined with cadmium and copper sheets. The system was set at operating energy range of 0-3000 keV and energy resolution of 661.6 keV peak of ^{137}Cs . For accurate quantitative determination of ^{226}Ra , ^{232}Th and ^{40}K in the samples, the detector calibration is necessary to ensure a good relationship between the respective peak positions of the spectrum and their corresponding gamma-ray energies. RGU-1, RGTh-1 and RGK-1 gamma spectrometric reference materials (gamma sources of known energies) supplied by the International Atomic Energy Agency (IAEA), whose activity is distributed homogeneously within the same volume and container geometry as that of the samples, was used for the energy calibration and absolute photopeak efficiency evaluation of the detector. Each sample was counted for 29,000 s. Knowing that through the period of sample storage, uranium attained circular equilibrium with its daughters, the gamma energy line of 1764 keV (15.6%) for ^{214}Bi used to represent the activity concentration of ^{226}Ra , while the activity concentration of ^{232}Th was evaluated from the average activity of photopeaks of 911.2 keV (25.8%) for ^{228}Ac and 238.6 keV (43.6%) for ^{212}Pb . The ^{40}K activity was determined from its 1460.8 keV (10.66%) single characteristic gamma line. An empty sealed beaker in the same geometry as the samples was first counted in the same manner for 29,000 s to determine the radiation background around the detector environment. The obtained background was used to correct the net peak area of gamma rays of measured radionuclides (Uosif and El-Taher, 2008). The activity concentrations were computed using the equation (Cevik et al., 2010; Khandaker et al., 2012):

$$A(\text{Bq/kg}) = \frac{N}{\varepsilon_{\gamma} \times I_{\gamma} \times M \times t} \quad (1)$$

Where A is the activity concentration of the nuclide, N is the total net count of a specific gamma emission, ε_{γ} is the detection efficiency for the specific gamma emission, I_{γ} is the emission probability, M is the mass of the sample (kg) and t is the counting time.

RESULTS AND DISCUSSION

Phosphate samples collected from the villages visited were analysed for their natural radioactivity concentrations using gamma ray spectrometry. The measured specific activity values for ^{226}Ra , ^{232}Th and ^{40}K along with their statistical uncertainties are summarized in Table 1. These values are actually taken as representing

Table 3. Activity concentration of ^{226}Ra , ^{232}Th and ^{40}K (Bqkg^{-1}) and Ra_{eq} (Bqkg^{-1}) in phosphate samples from different countries.

Country	Activity concentration (Bqkg^{-1})			Ra_{eq} (Bqkg^{-1})	Reference
	^{226}Ra	^{232}Th	^{40}K		
Pakistan (Hazara)	440	50	207	527.4	Khan et al. (1998)
Egypt (El-Mahamid)	567	217.3	217.3	921	Abbady et al. (2005)
Egypt (El-Mashash)	666	329.4	329.4	1182	Abbady et al. (2005)
Tanzania	5022	717	286	6069	Makweba and Holm (1993)
Algeria	619	64	22	712	Olszewska-Wasiolek (1995)
Sudan (Kurun)	393	6.9	141.3	414	Sam et al. (1999)
Tunisia	250	28	167	303	Cevik et al. (2010)
Nigeria (Sokoto)	720.1	33.5	315.3	792.3	Present work

the NORMs level in Sokoto phosphate rocks. The activity concentration levels for NORMs in phosphate rock samples from the five villages range from 44.37 to 1087.3 Bqkg^{-1} , with a mean value of $720.1 \pm 4.2 \text{ Bqkg}^{-1}$ for ^{226}Ra , from 13.2 to 85.3 Bqkg^{-1} with an average value of $33.5 \pm 1.4 \text{ Bqkg}^{-1}$ for ^{232}Th and from 111.2 to 459.0 Bqkg^{-1} with a mean value of $315.3 \pm 6.7 \text{ Bqkg}^{-1}$ for ^{40}K . The results show that the activity levels are higher for phosphate rocks from Kindiru (KDR) village. Generally however, the ^{226}Ra activity concentration values are higher than those of ^{232}Th and ^{40}K indicating that Sokoto phosphates are of sedimentary origin. It can be clearly seen that ^{238}U and its daughters plays a prominent role in contributing to the overall radioactivity content of phosphate rocks (Abbady et al., 2005; Khater et al., 2001). Although the values of radium activity concentration varies within the phosphate rock samples from the five villages, which according to Guimond (1990) and Ivanovich and Harmon (1992), may be due to uranium solubility under oxidation conditions and the percentage of P_2O_5 present, they are generally high. This can pose external radiation risk especially to women and children who mined this commodity on daily basis.

Table 3 give a comparison of activity concentration of natural radioactivity of phosphate rocks from different parts of the world. It can be seen from this table that the activity concentration of ^{226}Ra in this present study is relatively higher than those reported for many countries, except for Tanzania, while the average activity concentration value for ^{40}K falls within the same range as that reported in literature, except for Algeria that is relatively lower. The activity concentration value of ^{232}Th reported for Egypt, Tanzania and Algeria are however higher than that reported in this present study.

Numerous propositional criteria abound in literature for estimating environmental and human hazard due to radiation exposure (Bashir et al., 2013; Sabiha-Javied et al., 2010; Tufail, 2012). ^{226}Ra , ^{232}Th , and ^{40}K are not distributed uniformly within the soils and other environmental samples. A common radiological index, the radium

equivalent activity (Ra_{eq}), which therefore, compare the activity concentrations of the above three radionuclides and their associated radiation hazards, has been defined. This definition is consequent upon an assumption that 370 Bqkg^{-1} of ^{226}Ra , 259 Bqkg^{-1} of ^{232}Th and 4810 Bqkg^{-1} of ^{40}K produce the same gamma dose rate (Abbady, 2005; Cevik et al., 2010; Saueia et al., 2005). Ra_{eq} is calculated using the equation (Beretka and Mathew, 1985; Yu et al., 1992):

$$\text{Ra}_{\text{eq}} (\text{Bqkg}^{-1}) = A_{\text{Ra}} + 1.43A_{\text{Th}} + 0.077A_{\text{K}} \quad (2)$$

Where, A_{Ra} , A_{Th} and A_{K} are the respective activities of ^{226}Ra , ^{232}Th and ^{40}K in Bqkg^{-1} .

The total air absorbed dose rate, D (nGyh^{-1}), 1.0 m above the ground, from outdoor external exposure to natural radioactivity for the population living in the phosphate bearing areas is calculated using the formula (UNSCEAR, 2000):

$$D = 0.462A_{\text{Ra}} + 0.604A_{\text{Th}} + 0.0417A_{\text{K}} \quad (4)$$

Where, A_{Ra} , A_{Th} and A_{K} are the respective average activity concentrations (Bqkg^{-1}) of Ra, Th, and K in the phosphate rock samples.

Jibiri et al. (2007), reported that absorbed dose rates does not give a direct representation of exposure risk of an individual. To estimate the annual effective dose equivalent to an individual from outdoor terrestrial gamma radiation, UNSCEAR (2000), proposed two conversion factors: 0.7 Sv.Gy^{-1} as conversion coefficient from absorbed dose in air to effective dose received by adults, and 0.2 as outdoor occupancy factor, signifying that 80% of time is spent indoors across the world. The annual effective dose equivalent (AE) is estimated using the equation (UNSCEAR, 2000):

$$\text{AE} (\text{mSvyr}^{-1}) = D \times T \times \xi \quad (5)$$

Where, D (nGyh^{-1}) is the absorbed dose rate, T ($= 0.2 \times 24\text{hr} \times 365$) is the outdoor occupancy time, and ξ ($= 0.7 \times 106 \text{ SvGy}^{-1}$) is the conversion factor.

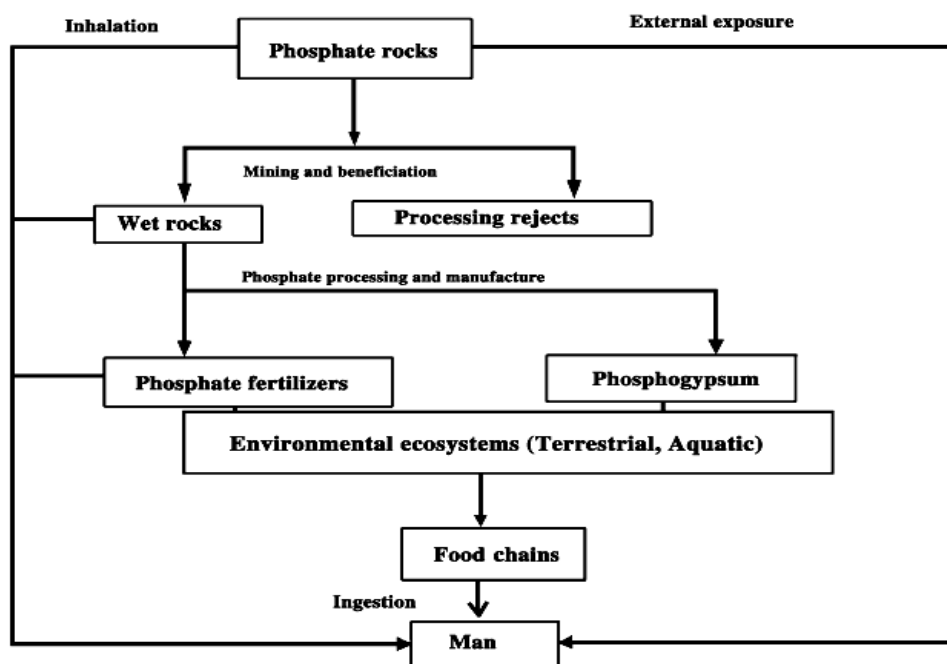


Figure 2. Environmental pathways of natural radionuclides from phosphate rocks.

Table 2. Mean values of total absorbed dose rate (D) and annual effective dose equivalent (AE) of Sokoto phosphate rock sample.

Sample ID	Ra _{eq} (Bq/kg)	D (nGy/h)	AE (mSv/yr)
DNG	722.2 (656.3-778.9)	334.5 (303.2-361.5)	0.4 (0.4-0.4)
KDR	1182.4 (1135.6-1234.0)	547.5 (526.1-570.6)	0.7 (0.6-0.7)
DLG	876.1 (748.3-942.3)	406.9 (347.9-436.9)	0.5 (0.4-0.5)
WRN	558.9 (502.6-664.6)	259.6 (233.1-309.4)	0.3 (0.3-0.4)
DNK	621.7 (545.6-672.8)	288.3 (252.0-312.9)	0.4 (0.3-0.4)
Av. Range	792.3 (502.6-1234.0)	367.3 (233.1-570.6)	0.4 (0.3-0.7)

The two basic environmental pathways by which radiation dose from phosphate rocks are imparted to the general public and phosphate workers in particular are the external and internal exposures represented in Figure 2 (Khater et al., 2001).

The calculated results for radium equivalent activity (Ra_{eq}), total absorbed dose rate in air (D) and the annual effective dose equivalent (AE) are presented in Table 2. Phosphate rock samples from Kindiru (KDR) recorded the maximum value of Ra_{eq} activity (1182.4 Bqkg^{-1}), while phosphate samples from Wuruno (WRN) had the minimum Ra_{eq} activity value of 558.9 Bqkg^{-1} . The mean value of Ra_{eq} across the entire study areas is 792.3 Bqkg^{-1} which is higher than the safety limit of 370 Bqkg^{-1} recommended by the Organization for Economic Cooperation and Development (NEA-OECD, 1979).

The total absorbed doses calculated from the measured activities of phosphate samples varies from 259.6 nGy^{-1}

for WRN samples to 547.5 nGy^{-1} for KDR samples, with a mean value of 367.3 nGy^{-1} . These high dose rate values witnessed across the entire study areas may not be unconnected with the high radium contents of the phosphate rocks. The mean value of the total absorbed dose rate as seen in Table 2 is almost 3.6 times the maximum value of the world's soil background level of 102 nGy^{-1} proposed by UNSCEAR (2000), which shows the severity of radiation risk the inhabitants of the study areas are exposed to. The computed values for the annual effective dose equivalent in the studied phosphate rock samples as presented in Table 2 varies from 0.3 mSvyr^{-1} for WRN sample to 0.7 mSvyr^{-1} for KDR samples, with a general mean value of 0.4 mSvyr^{-1} . These values are found to be lower than the safety limit of 1 mSvyr^{-1} recommended by the International Commission on Radiological Protection (ICRP-60, 1990). A plot of radium equivalent activity and average dose rates with their res-

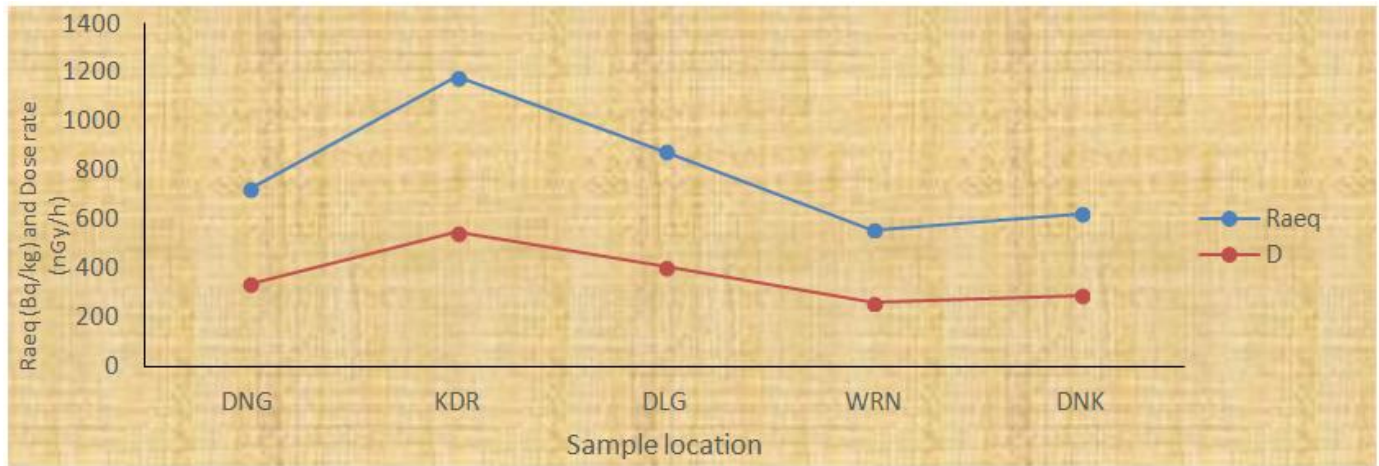


Figure 3. Radium equivalent and average dose rates for phosphate rock samples from different locations.

pective sample locations are shown in Figure 3. These values are higher than the respective recommended safety limits, which presents a serious radiation risk to the inhabitants of the study areas, especially when the untreated phosphate rocks are used generally by the local farmers as substitute for fertilizer. Women and children within the study areas are at greater radiation risk from internal dose due to inhalation of radon and dust during mining and blending of phosphate rocks for direct application to plants. Detail study of this exposure pathway is therefore recommended so as to estimate the occupational radiation dose in these phosphate bearing areas of Sokoto state and to enforce restrictions and precautionary measures where necessary, from the view point of radiation protection.

Conclusion

The results of this preliminary radiological investigation of Sokoto phosphate rocks clearly demonstrates the radiological burden an average member of phosphate bearing communities are exposed to. The obtained results show that ^{238}U and its decay daughters are the primary contributors to the radioactivity level of phosphate bearing formations of Sokoto State. The computed values of external radiation exposure from natural radioactivity of phosphate rocks show that the radioactivity of Sokoto phosphate rock samples is above the proposed safety limit of 1 (370 Bqkg^{-1}), which demonstrates the severity of radiation risk the local farmers are exposed to especially when using the untreated ground phosphates as fertilizer. It is also obvious that phosphate industry, among other geological materials' processing industries, play a significant role in the radiation dose received by the public. The data obtained from this investigation under-

scores the need for in-depth radiological assessment of all the phosphate bearing communities in Sokoto State. Underground water supplies and local building materials from these communities should be investigated for their contribution to the radiation dose incurred by the general public.

Conflict of Interests

The author(s) have not declared any conflict of interests.

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