

## Geogenic Radiological Dose and Activity Concentrations in Phosphate Rocks and Industrial Tailings from Kaduna, Northwestern Nigeria

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### ARTICLE INFORMATION

### ABSTRACT

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This study assesses the activity concentrations of Naturally Occurring Radioactive Materials (NORMs) in phosphate rocks, tailings, and related materials from a superphosphate fertilizer industry in Kaduna, Nigeria. Samples of sludge, soil, sediment, phosphate rock, and phosphogypsum were analyzed using Instrumental Neutron Activation Analysis (INAA) with high-purity germanium detectors. Radiological parameters, including Annual Effective Dose Equivalent (AEDE), External and Internal Hazard Indices, Annual Gonadal Dose Equivalent (AGDE), Radium Equivalent Activity (Raeq), Absorbed Gamma Dose Rate (D), Activity Utilization Index (AUI), and Gamma Index ( $I_\gamma$ ) were computed following UNSCEAR (2000) formulations. The mean AEDE, AGDE, and Raeq values were 87.0  $\mu\text{Sv/y}$ , 517.7  $\mu\text{Sv/y}$ , and 137.3 Bq/g, respectively. Most samples exceeded global averages, indicating potential radiological risks to the surrounding environment. These findings highlight the need for continuous monitoring and improved waste management practices in the fertilizer industry.

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### INTRODUCTION

Phosphate rocks may contain high levels of radioactivity due to their composition of naturally occurring uranium, potassium, and thorium, and their daughter isotopes radium, radon, and polonium. Phosphate is mainly mined from apatite and phosphorite, where the enrichment of phosphate has been increased by geological and biological processes. Uranium is concentrated during the enrichment of phosphate such that a high phosphate concentration generally corresponds to a high uranium content (50 - 300ppm). Marine-deposited phosphate typically has a

higher level of radioactivity than igneous phosphate deposits because of the presence of uranium in seawater (Abou El-Anwar et al., 2017). However, thorium is more expected to occur in igneous phosphorite. Long-lived radioactive elements, such as uranium (238U), thorium (232Th), and potassium (40K), and their decay products, including radium and radon, are examples of naturally occurring radioactive materials (NORMs) (Arabi et al., 2016).

Phosphates are used extensively as a source of phosphorus for fertilizers and for the manufacturing of phosphoric acid

and gypsum. Phosphate ores typically contain about 1500 Bq/g of uranium and radium (Anselmo S P and J. M. Godoy, 2002). In most cases, during the production process of fertilizer, phosphate ore is treated with sulfuric acid, thus enriching the fertilizers in uranium by up to 150% relative to the ore, while 80% of  $^{226}\text{Ra}$ , 30% of  $^{232}\text{Th}$ , and 5% of uranium are left in phosphogypsum (UNSCEAR, 2008). Moreover, the beneficiation of the ore during the production phase does not result in a reduction of NORM. Also, the gaseous and particulate emissions generated during the processing of phosphates contain  $^{238}\text{U}$  and  $^{226}\text{Ra}$ . The discharge of these emissions to the environment leads to exposure of the population to radiation hazards and metal element contamination of soils, water bodies, and air (UNSCEAR, 2000). The radioactivity of these phosphate ores (due to uranium, thorium, and radium) can reach 10,000 Bq/g

In addition, toxic metal elements are relatively dense metals or metalloids that are associated with potential toxicity, especially in environmental contexts (Srivastava and Goyal, 2010). These metals include cadmium, mercury, lead, arsenic, manganese, chromium, cobalt, nickel, copper, zinc, selenium, silver, antimony, and thallium. They are found naturally in the earth and can become concentrated as a result of human activities.

Processing phosphate occasionally leads to assessable doses of radiation to people. For instance, phosphate rocks holding approximately 120 ppm uranium have been utilized as a source of uranium as a byproduct in the USA (an estimated 17,000 TU). During the production of fertilizers, the naturally occurring radionuclides (NORM) are redistributed at trace levels throughout the

environment, thus transforming into a source of radioactivity (Uosif et al., 2014). Since most radionuclides come into the fertilizers, the focus of related studies has been on investigating the concentration of NORM in chemical fertilizers produced from phosphate (Uosif et al., 2014; Shahul Hameed et al., 2014). This is imperative as radioactive elements can migrate from agricultural fertilizers to soil and plants, and eventually to humans through ingestion of food (Rehman et al, 2006). However, not much emphasis has been given to external exposure and environmental contamination effects during handling, packing, and transporting of the fertilizers, as well as in the production phases of beneficiation, acid leaching, and separation. Over time, there has been mounting concern about the radiological impact of the inordinate and haphazard disposal of tailings from the processing and extraction activities of both large- and small-scale operators (Funtua and Elegba, 2007). The radiation levels have long been considered to exceed the annual dose limit for members of the public (Funtua et al., 2004).

Therefore, this study sets out to investigate environmental risk and establish a radiological baseline by determining the concentration levels and risks associated with the use and processing of phosphate rock in the production of super phosphate fertilizer. This is achieved by assessing the concentration levels of radioelements and metal elements in soils, sludge, dust, phosphate rock, and phosphogypsum samples collected from around the super phosphate fertilizer company located within the Kaduna metropolis in northwest Nigeria (Fig.1).



Fig. 1: Aerial view of the study area (super phosphate fertilizer company and environs) Kaduna (inset: map of Nigeria showing location of Kaduna (google map 21.08.2025))

The risks and hazards associated with the deposition of these contaminated industrial wastes and their use by inhabitants of the area, sometimes as soil fertility supplements on farmland, were evaluated. The area is located in a densely populated area of Kaduna metropolis (Fig.1); therefore, the results obtained will go a long way in coming up with proactive mitigation measures by authorities e.g., the National Environmental Standards and Regulations Enforcement Agency (NESREA) whose responsibility it is, to regulate the activities of corporate organizations with regards to environmental implications of their activities.

## **MATERIALS AND METHODS**

### ***Sampling and Sample Preparation***

Two samples, each of sludge, soil sediment, dust, phosphate rock, and phosphogypsum, were randomly collected within and around the super phosphate fertilizer company using the EPA standard sampling procedure. Sludge samples were collected from only two tanks in the industry premises. Soil samples were collected, one from the premises of the industry and another outside the premises. Dust samples were collected from roofs around the premises. Phosphate rock samples were collected from the rock pile freshly brought from the phosphate quarry, while phosphogypsum samples were collected from another pile approved for collection by the authorities of the fertilizer company. The industrial area is a restricted area; hence, authorization had to be sought before samples were collected. Areas approved for the team to collect samples were only accessible at the time of collection, as major work was already going on around the company. All the samples collected were carefully packed into a polythene bag and transported to the laboratory at the Center for Energy Research and Training (CERT), Ahmadu Bello University, Zaria, for analysis.

The sampling techniques administered in this study were consistent with those proffered in ISO 18400: 101 -205 for soil quality, and ISO 18589: 2, 7 for measurement of radioactivity in the environment. IAEA soil-7 was used as a standard reference material relevant to the sample matrix and included as an additional sample for irradiation. Before sending the sample into the reactor through the pneumatic system, 0.2000g of each sample was carefully placed in a high-density polyethylene bag, heat sealed, and stored in the decorator. For short-lived irradiation, one sample was packaged per capsule, while for long-lived irradiation, five to seven samples were packed in one

capsule as required by the Nigeria Research Reactor 1 (NIRR-1) protocol for geological samples. The sealed capsules were made air-tight, ready for irradiation. The spatula and forceps were cleaned with acetone before handling samples.

### ***Instrumentation***

The samples were analyzed at the Reactor Engineering Section of the Center for Energy Research and Training (CERT), Ahmadu Bello University (ABU), Zaria, Nigeria, using the Instrumental Neutron Activation Analysis (INAA) technique using a Miniature Neutron Source Reactor (MNSR). INAA can analyze NORMs, trace, minor, and major elements in different sample types (Arabi et al., 2016).

Although MNSR has more flux stability, longer life span, higher negative temperature coefficient of reactivity, and low under-moderation, the inherent limitations include intermittent operation, low flux, and its eventual decrease over the operational period, and the necessity of using cyclic INAA rather than single rabbit irradiations, which affect the precision and accuracy of results. Nonetheless, the maximum relative error of the results calculated with MNSR compared with the measurement data is approximately 3.5% (Arabi et al., 2016).

The reactor uses high-enriched uranium as fuel, and light water as a moderator and coolant. The natural radioactivity in these samples was determined using High-resolution gamma-ray spectrometers. The High Purity Germanium (HPGe) detector (model GEM 30P4 - 76) with a resolution of 1.74keV FWHM, operated at 1332.5keV of <sup>60</sup>Co, H.V. biased supply model 659 Ortec, 5kV, spectroscopy amplifier model 672 Ortec, acquisition interface card with computer and basic spectroscopy software (WINSPAN) was employed in the analysis. The samples were pulverized together with the standard and then sealed and irradiated at a flux of  $5 \times 10^{11} \text{ ncm}^{-2}\text{s}^{-1}$  for 6 hours. Afterward, the samples were counted for 1800s and 3600s after 4 and 14 days of cooling, respectively. Identification of gamma rays of product (uranium, thorium, and potassium) radionuclide through their energies and quantitative analysis of their concentrations was achieved using the gamma-ray spectrum analysis software. The activation product's half-life and energy of the photopeak are detailed in a previous study (Ahmed, 2005).

### Calculation of risks and hazard indices

The risks and hazard indices were calculated from values obtained and analyzed from the results of INAA. These indices are discussed below.

#### Annual Effective Dose Equivalent ( $\mu\text{Sv/y}$ )

The annual effective dose (AEDE) is the effective dose that can be received by the public with fewer risks from the effects of ionizing radiation, though the cumulative value could be higher when added to the dose received from cosmic radiation. To estimate the annual effective dose rate, it is necessary to use the conversion coefficient from the absorbed dose in air to the effective dose ( $0.7 \text{ SvGy}^{-1}$ ) and the outdoor occupancy factor ( $0.2 \text{ mSvy}^{-1}$ ) proposed by UNSCEAR (2000). Therefore, the effective dose rate is determined as follows (Eq. 1):

$$H_R (\text{mSvy}^{-1}) = D_R (\text{nGyh}^{-1}) \times 24\text{h} \times 365.25\text{d} \times 0.2 \quad \text{Eq 1}$$

(Out-door occupancy factor)  $\times 0.7 \text{ SvGy}^{-1}$ (conversion factor)  $\times 10^{-6}$

$$H_R = D_R \times 8766 \times 0.2 \times 0.7 \times 10^{-6} = D_R \times 0.00123$$

Where:  $D_R$  = estimated dose rate

$D(\text{nGyh}^{-1})$  is given by Eq. 6

#### External and Internal Hazard Index ( $H_{ex}$ , $H_{in}$ )

The external hazard index is an evaluation of the hazard of the natural gamma radiation (Arabi et al., 2016; Mujahid, S. A. et al., 2008), while the internal exposure to radon and its daughter products is quantified by the internal hazard index ( $H_{in}$ ) (Mujahid et al., 2008; Xinwei, 2004). In addition to the external radiation hazard they pose, radon and its short-lived daughters are also hazardous to the respiratory organs. The internal exposure caused by radon and its daughter products can be quantified using the internal hazard index ( $H_{in}$ ) (UNSCEAR, 2000), which is defined by Eq. 2 below:

$$H_{in} = A_{Ra}/185\text{Bqg}^{-1} + A_{Th}/259\text{Bqg}^{-1} + A_K/4810\text{Bqg}^{-1} \quad \text{Eq 2}$$

The internal hazard index is defined to reduce the acceptable maximum concentration of  $^{226}\text{Ra}$  to half the value appropriate for external exposure alone. This criterion was proposed by Krieger, R. (1981), to assess the safety of use of materials in the construction of dwellings.

$$H_{in} \leq 1$$

The external hazard index is a criterion used for assessment of the radiological suitability of a material using the relation given in UNSCEAR, 2000, as expressed in equation 3 below (Beretka, J. and P.J. Mathew, 1985):

$$H_{ex} = A_{Ra}/370\text{Bqg}^{-1} + A_{Th}/259\text{Bqg}^{-1} + A_K/4810\text{Bqg}^{-1} \leq 1 \quad \text{Eq 3}$$

Where:

$A_{Ra}$ ,  $A_{Th}$ , and  $A_K$  are the activities of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$ , and  $^{40}\text{K}$  in  $\text{BqG}^{-1}$ .

#### Annual Gonad Dose Equivalent ( $\mu\text{Sv/y}$ )

The annual gonad dose equivalent (AGDE) covers the dose received by certain organs resulting from the effect of ionizing radiation. These organs include gonads, female breast, active bone marrow, and bone surface cells (UNSCEAR, 1988). In UNSCEAR (1988), the activities of bone marrow and bone surface cells are organs of interest; therefore, the annual gonad dose equivalent (AGDE) was introduced to take care of the specific activities arising from Ra, Th, and K. The AGDE was calculated using Eq. 4 below:

$$\text{AGDE} (\text{mSvy}^{-1}) = 3.09A_{Ra} + 4.18A_{Th} + 0.31A_K \quad \text{Eq 4}$$

#### Radium Equivalent Activity ( $Ra_{eq}$ ) ( $\text{Bq/g}$ )

A common radiological index referred to as radium equivalent was used in this study to evaluate the actual activity level of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$ , and  $^{40}\text{K}$  in the samples and the radiation hazards associated with these radionuclides. This is because the distribution of natural radionuclides in the samples under investigation is not uniform, and it is assumed that  $10 \text{ Bqg}^{-1}$  of Ra,  $7 \text{ Bqg}^{-1}$  of Th, and  $130 \text{ Bqg}^{-1}$  of K produce an equal gamma-ray dose (Krisiuk, 1971; Strandén, 1976).

This index is usually known as radium equivalent activity (UNSCEAR, 2008), and is calculated using Eq. 5 below:

$$Ra_{eq} = A_{Ra} + 1.43A_{Th} + 0.077A_K \quad \text{Eq 5}$$

#### 2.3.5 Absorbed gamma ray dose rate ( $\text{nGy/h}$ )

The absorbed gamma ray dose quantifies the mean gamma dose rate in air at 1 m from the ground surface when samples are used as building material. The global average

value is about 84 nGyh<sup>-1</sup>. The estimation of the Absorbed Gamma Dose Rate (D<sub>R</sub>) was calculated to take care of the mean values of gamma dose rate in air at 1m from the ground surface for different kinds of building materials. These values were calculated using Eq. 6, presented as follows:

$$D_R (\text{nGyh}^{-1}) = 0.92A_{\text{Ra}} + 1.1A_{\text{Th}} + 0.0807A_{\text{K}} \quad \text{Eq 6}$$

### **Activity Utilization Index (AUI)**

Because building materials are mostly of geologic origin, they act as sources of radiation and also as shields against outdoor radiation (UNSCEAR, 1993). In houses constructed from different building materials, the activity concentrations of natural radionuclides in those materials strongly affect the indoor absorbed dose, while the walls absorb the radiation emitted by outdoor sources. Consequently, dose rates in indoor air will be elevated according to the concentrations of naturally occurring radionuclides in the construction materials used. To calculate dose rates in air from <sup>226</sup>Ra, <sup>232</sup>Th, and <sup>40</sup>K in building materials, and by applying the appropriate conversion factors, another parameter referred to as activity utilization index (AUI) was obtained using Eq. 7 below:

$$AU = (A_{\text{Ra}}/50\text{Bqg}^{-1})f_U + (A_{\text{Th}}/50\text{Bqg}^{-1})f_{\text{Th}} + (A_{\text{K}}/500\text{Bqg}^{-1})f_{\text{K}} \quad \text{Eq 7}$$

Where  $f_{\text{Th}}$ ,  $f_{\text{Ra}}$ , and  $f_{\text{K}}$  are the fractional contributions to the total dose rate in air, attributable to gamma radiation from the actual concentrations of these radionuclides. In the NEA-OECD report (1979), the typical activities per unit mass of <sup>232</sup>Th, <sup>226</sup>Ra, and <sup>40</sup>K in building materials are reported as 50, 50, and 500 Bq/g, respectively.

### **Gamma representation index (I<sub>γ</sub>)**

The gamma activity concentration index (I<sub>γ</sub>), defined by Righi, S. and Bruzzi (2006) and the European Commission (1990) as a screening tool for identifying hazardous construction materials. Materials with a gamma index > 6 correspond to a dose rate higher than 1 mSv-1 and therefore must be avoided. It is given as Eq. 8 below:

$$I_\gamma = (C_{\text{Ra}}/300\text{Bqg}^{-1})f_U + (C_{\text{Th}}/200\text{Bqg}^{-1})f_{\text{Th}} + (C_{\text{K}}/3000\text{Bqg}^{-1})f_{\text{K}} \quad \text{Eq 8}$$

Values of  $I_\gamma \leq 2$  correspond to a dose rate criterion of 0.3mSvy<sup>-1</sup>, whereas  $2 < I_\gamma < 6$  corresponds to a criterion of 1

mSvy<sup>-1</sup> (European Commission, 1990; Anjos, R.M., 2005). Materials with  $I_\gamma > 6$  should be avoided, as these values correspond to dose rates higher than the 1 mSv y<sup>-1</sup> value recommended for the population (European Commission, 1999).

### **Excess lifetime cancer risk (ELCR)**

Excess lifetime cancer risk (ELCR) is defined as the fatal cancer risk per Sievert. For the stochastic effect, the ICRP 60 document (Xinwei, L. and China. J. (2004) recommends a value of 0.05 from the public. This risk factor was calculated using the equation (Eq. 9) below:

$$\text{ELCR} = \text{AEDE} \times \text{DL} \times \text{RF} \quad (5) \quad \text{Eq 9}$$

Where:

AEDE, DL, and RF are the annual effective dose equivalent, duration of life (70 years), and risk factor (Sv<sup>-1</sup>), fatal cancer risk per Sievert. For stochastic effects, ICRP60 uses values of 0.05 for the public (Stranden, 1976).

### **Potential Heavy and Toxic Elements**

Potential heavy and toxic elements in the samples were evaluated, plotted, and analyzed for compliance with world averages and standards as presented in the appropriate tables and corresponding figures that allow for pictorial variability of the element in each of the samples as provided in the discussion section.

## **RESULTS AND DISCUSSION**

### **Radionuclide Concentrations**

The activity concentrations of <sup>238</sup>U, <sup>232</sup>Th, and <sup>40</sup>K in all samples were measured in Bq/g using Instrumental Neutron Activation Analysis (INAA), and the results were presented and plotted (Table 1). It can be seen that the activity concentration of <sup>238</sup>U, <sup>232</sup>Th, and <sup>40</sup>K was recorded in all samples except for one of the phosphate samples (PRK3), where <sup>40</sup>K is below detection level (BDL). The activity concentration of <sup>40</sup>K fluctuated from BDL to 6388±40 Bq/g in the sediment sample, with a mean concentration value of 1241.9 Bq/g for all the samples. The <sup>238</sup>U and <sup>232</sup>Th values (Bq/g) range from 1.6±0.3 - 35.5±0.4 and 2.3±0.1- 8.1±0.3, with mean concentration values of 35.38 and 4.4 Bq/g, respectively.

The activity levels of <sup>238</sup>U in the phosphogypsum samples are considerably lower than those of the other samples,

while  $^{232}\text{Th}$  activity was slightly lower. These depleted activity levels of  $^{238}\text{U}$  and  $^{232}\text{Th}$  in phosphogypsum are indicative of its inherently low primordial concentrations of radionuclides. The relatively higher radionuclide activity concentration values recorded in the sediment sample can be attributed to the fact that soil around the factory receives most of the waste, i.e., settling dust,

dumped waste (sludge), or even some fragments of the phosphate rocks used inside the factory. The movement and long queues of transporting trucks within the vicinity of the factory also contribute to this higher radionuclide concentration in soils.

Table 1 Activity concentration (Bq/g) of  $^{238}\text{U}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  in the samples

NORM	SAMPLE ID									
	SLG1	SLG2	PGS1	PGS2	SED1	PRK1	PRK2	PRK3	DUST 1	DUST 2
$^{40}\text{K}$	1556±34	1003±29	BDL	294±90	6388±4	867±14	979±16	BDL	575±13	757±15
$^{238}\text{U}$	35.5±0.4	35.4±0.5	7.2±0.2	6.8±0.2	1.6±0.3	34.5±0.4	44.3±5	120±1	33.7±0.4	34.8±0.4
$^{32}\text{Th}$	3.8±0.2	2.9±0.2	2.3±1	2.3±0.1	8.1±0.3	5.7±0.2	5.3±0.3	7.9±0.3	3.1±0.2	2.6±0.2

Table 2 Comparison of activity concentration of  $^{238}\text{U}$ ,  $^{232}\text{Th}$ , and  $^{40}\text{K}$  with other values around the world

Location	Ref	Phosphate Rock Activity (Bq/g)			
		$^{226}\text{Ra}$	$^{238}\text{U}$	$^{232}\text{Th}$	$^{40}\text{K}$
India	Shahul, et al., 2014	1.29	1.34	0.09	0.01
Brazil	Mazzilli <i>et al.</i> , 2000	0.6	0.04	0.1	0.02
Egypt	Ahmed, N. K. (2005)	0.1	-	0.04	0.5
This work (Nigeria)		0.21	0.082	0.0066	0.923

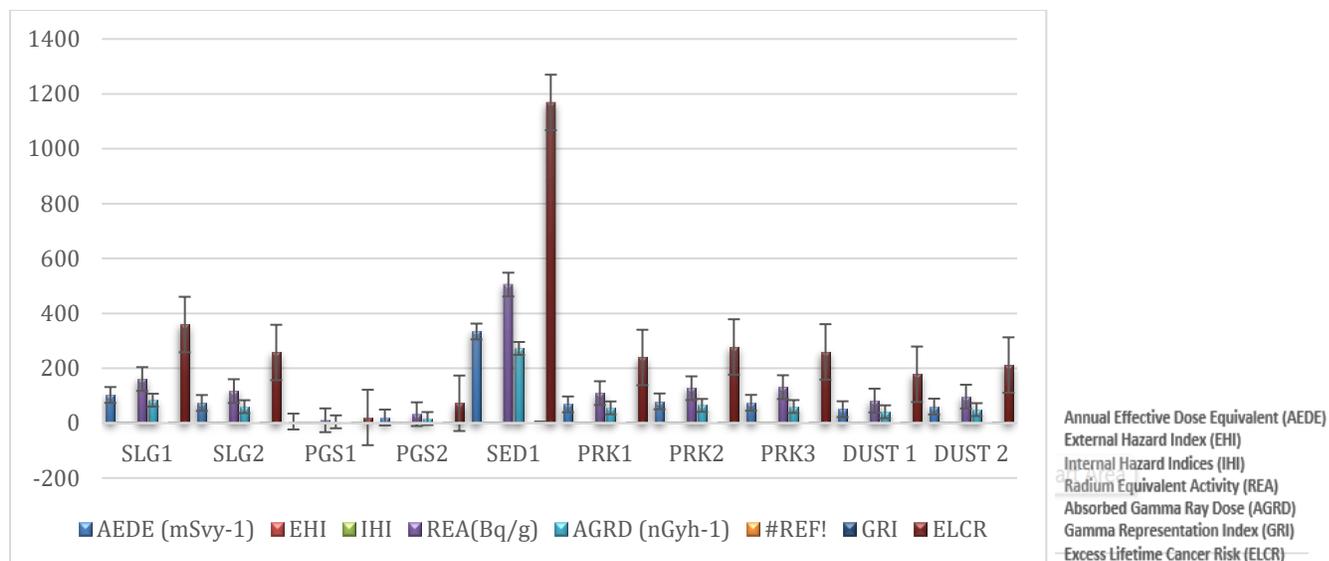


Figure 2: A plot of the radiological parameters calculated for all the samples

The activity concentration values of the radionuclides (Table 2) were compared with values obtained in different locations around the world. According to the UNSCEAR (2000) report, the world average values of activity concentration for  $^{238}\text{U}$ ,  $^{232}\text{Th}$ , and  $^{40}\text{K}$  are 35 Bq/g, 35 Bq/g, and 370 Bq/g, respectively. The measured mean value of activity concentration of  $^{238}\text{U}$  for the study area is within the world average range, whereas the phosphogypsum and sediment samples are much lower. Although the phosphate rock sample (PRK3) significantly exceeds this average (Table 1). However, the mean activity concentrations of  $^{40}\text{K}$  exceeded the world average, while  $^{232}\text{Th}$  is comparatively depleted.

The activity concentration values for some selected countries are presented in Table 2. In general, the phosphate rocks are enriched with  $^{238}\text{U}$  (0.082–1.7 Bq/g) as compared to  $^{232}\text{Th}$  (0.0066–0.09 Bq/g). However,  $^{40}\text{K}$  concentration fluctuated from BDL to 0.02 Bq/g, with this study recording higher values. The  $^{238}\text{U}$  and  $^{232}\text{Th}$  concentrations of the samples are observed to be slightly less than the earlier reported values, while  $^{40}\text{K}$  is considerably higher. For phosphogypsum, the activity concentration of  $^{238}\text{U}$  and  $^{232}\text{Th}$  recorded in this study is relatively lower than values reported elsewhere across the world. In contrast, the  $^{40}\text{K}$  concentration values are higher. This variation in the activity concentration level of the radionuclides can be attributed to the geological variation in mining of the ores and chemical processing involved in the production phase (Shahul Hameed et al., 2014).

### **Radiological Parameters**

Radiological parameters such as annual effective dose equivalent ( $\text{mSv}^{-1}$ ), internal hazard index, external hazard index, annual gonad dose equivalent ( $\text{mSv}^{-1}$ ), radium equivalent activity ( $\text{Bq}^{-1}$ ), absorbed gamma ray dose rate ( $\text{nGyh}^{-1}$ ), activity utilization index and gamma representation index were also calculated from the activity concentration values of the different radionuclide and the result plotted in Fig.2.

The annual effective dose (AEDE) for the public is 1  $\text{mSv}^{-1}$  as set by UNSCEAR (Arabi et al., 2016). The AEDE values recorded for the entire sample are greater than the permissible limit. For all the samples studied, the range of values recorded is 5.88 - 333.95  $\text{mSv}^{-1}$  with a mean value of 87.0  $\text{mSv}^{-1}$ . A plot of internal and external hazard index ( $H_{\text{in}}$  and  $H_{\text{ex}}$ ) parameters indicates that values are relatively higher in the sediment sample (range of 0.028 - 1.36, and a mean value of 0.4) than in any other sample, and

lowest in the phosphogypsum samples (range of 0.048 - 1.37 and a mean value of 0.5).

Furthermore, the results indicate that the sediment sample has the highest annual gonad dose equivalent (AGDE) ( $2044.6 \text{ mSv}^{-1}$ ) with a range of 31.86 - 2044.63  $\text{mSv}^{-1}$  obtained for all the samples and a mean value of 517.7  $\text{mSv}^{-1}$ , as shown in Table 4.

Radium equivalent activity is a common radiological index employed in this study to evaluate the actual activity level of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$ , and  $^{40}\text{K}$ , since the distribution of NORM in the samples is not uniform. The variation observed for radium equivalent activity followed the same pattern as the other parameters, where the sediment sample and phosphogypsum have the highest and lowest values, respectively. The radium equivalent activity values range from 10.49 to 505.06 Bq/g with a mean value of 137.3 Bq/g.

The absorbed gamma ray dose value obtained is lower than the global average ( $84 \text{ nGyh}^{-1}$ ) for all the samples, except the sediment sample ( $272.3 \text{ nGyh}^{-1}$ ). The mean value for all the samples is  $70.9 \text{ nGyh}^{-1}$ . The phosphogypsum sample recorded the lowest value of  $31.86 \text{ nGyh}^{-1}$ .

The activity utilization index (AUI) values for most of the samples were found to be greater than <2 except for phosphogypsum 1 (PGS1) and phosphate rock 3, with AUI values of 0.1 and 1.3, respectively. AUI for the entire sample ranged from 0.11 - 81631.22 with a mean value of 9388.9, while the sediment sample had the highest value of 81631.22. The results are vital as materials with an activity utilization index (AUI) of <2 have been recommended for construction.

The gamma activity concentration index ( $I_{\gamma}$ ) for all the samples ranged from 0.071 - 4.35, with the sediment sample recording the highest value of 4.35. The average gamma representative index is 1.1. Based on this parameter, the samples can be used for construction since their values are <6. The European Commission (1999) suggests that building materials with a gamma activity concentration index with <6 should be exempted from all restrictions concerning their radioactivity, provided that the excess gamma radiation originating from them does not increase the annual effective dose to members of the public by more than 0.3  $\text{mSv}$  (Anjos, 2005).

The excess lifetime cancer risk (ELCR) values obtained are presented in Fig. 2. For this parameter (ELCR), all the samples studied had values significantly greater than the 0.05 recommended by the ICRP (1994). The values range from 20.6 to 1169, with a mean of 304.57.

The radiological parameters of the studied materials exceed the world averages. Although these studied materials are not used in the construction of dwellings, except for the soil sediment sample and the phosphate rock to some extent, the materials contribute a lot to environmental radioactivity.

### Metal elements

The result of the analysis of all the heavy metals in the soil sample (Table 3) indicates that all the element concentrations fall within common ranges in soils around the world, except manganese (Mn), whose concentration is 6610 mg/g, almost twice the highest value (3,000 mg/g) for the world average (Table 3). Also, the sediment sample (SED1) has the highest values recorded for Al, Ta, Cs, Hf, Ta, Rb, and Br. Cr and V, while samples of phosphate rocks (PRK1 and PRK2) have the highest level of As, Mn, Sr, Hf, Sb, Zn, Ba, with almost all the samples having Ta, Cs, Rb, Ti, and Br levels lower than the detection limit, except SED1. Sample of phosphate rock (PRK1) had the highest value of 21.9 mg/g for arsenic (As), which

is twice the value obtained in the soil sample (SED1) as obtained from the study area. Also, the chromate (Cr) value obtained in the SED1 sample ( $149 \pm 4$  mg/g) has the highest value, as can be observed from the Table, followed by all samples of phosphate rocks (PRK1, PRK2, and PRK3), with values 62, 63, and 75 mg/g, respectively. This might be the source of the high value recorded in SED1 around the study area. Comparing these values with the FAO/WHO guidelines for maximum allowable limit for concentration of heavy metals in soil (mg/g) and other values worldwide (Tables 3), results obtained from heavy metals (As) (21.9 mg/g), Cr (149 mg/g) and Mn (6610 mg/g) are greater than the FAO/WHO guidelines (Table 6) of 20 mg/g, 100 mg/g and 5000 mg/g for the As, Cr, and Mn, respectively, while values for Zn is the same with maximum allowable limit. Mean values for heavy metals (Fig. 3) indicate that  $Al > Ta > Mn$  (9840.1, 8884, and 2086.8 Bq/g), while Cs has the lowest value (3.0 Bq/g) among all the samples within the Cs detectable limit.

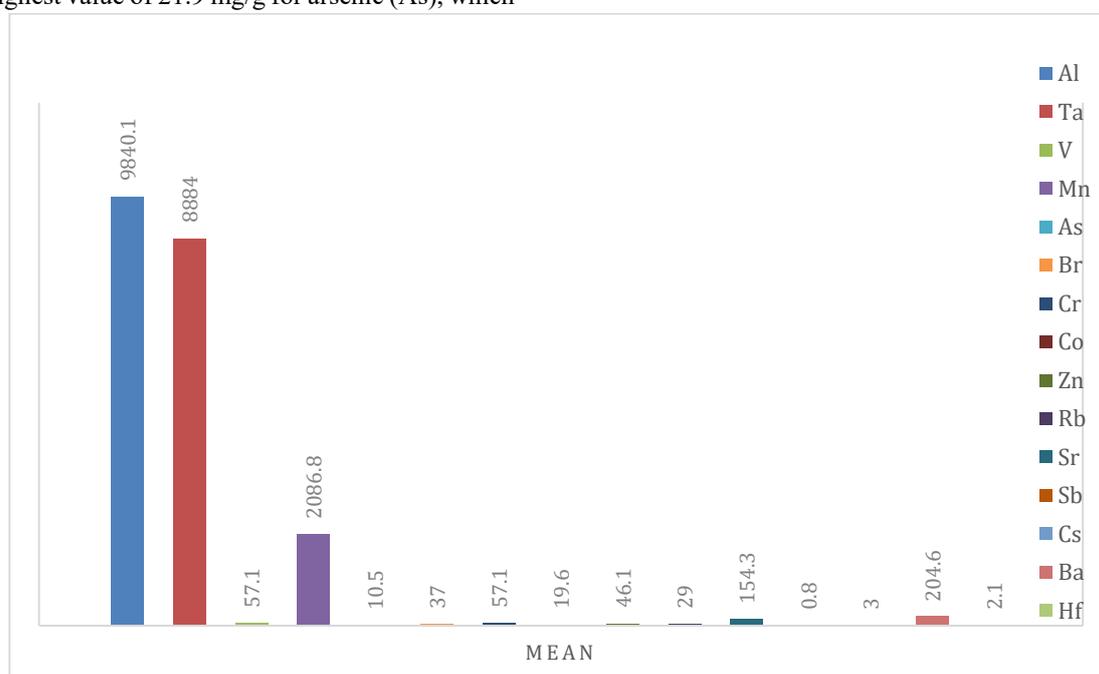


Figure 3: Mean (Bq/g) levels of metal elements in all samples

Table 3. Comparison of the Concentrations of Metal Elements in the Sample and the FAO/WHO Guidelines

METAL	This work (mg/g)	FAO/WHO Guidelines (mg/g)	Soil (SED 1) (This work) (mg/g)	Common Range in Soils (mg/g)
Aluminium (Al)	$57440 \pm 12 - 4587 \pm 21$		$57440 \pm 12$	10,000-300,000
Arsenic (As)	$2.3 \pm 0.1 - 21.9 \pm 0.3$	20	$10.9 \pm 0.4$	1-50; 1-4010
Barium (Ba)	BDL - $388 \pm 39$		$134 \pm 36$	100-3,000
Chromium (Cr)	$20 \pm 2 - 149 \pm 4$	100	$149 \pm 4$	1-1,000; 5-3,000
Manganese (Mn)	$12.1 \pm 0.4 - 6610 \pm 7$		$847 \pm 3$	20-3,000
Zinc (Zn)	BDL - $106 \pm 9$	300	$82 \pm 8$	10-300

## CONCLUSION

This study aimed to evaluate the levels of NORM and metal elements in various materials located within and around a phosphate fertilizer factory to assess the radiological risks and metal element levels in comparison to those found elsewhere globally. The levels of all the evaluated parameters varied from one sample to another, with the sediment sample recording the highest values for all parameters, indicating that the environment around the factory is highly vulnerable to all the contaminants studied. The radiological parameters of the studied materials exceed the world averages. Hence, the samples, particularly the soil sediment and phosphate rock, to some extent might contribute to both indoor and environmental radioactivity due to their potential uses as construction materials. Moreover, the major river around the factory, River Kaduna, may not be spared by these contaminants, as runoff and drains from and around the factory certainly flow into the river and partly percolate/infiltrate down to the groundwater level, which may end up in wells and ultimately in homes as drinking water.

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### *Declaration of Interest*

The authors wish to declare that there is no conflict of interest concerning this work.

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