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Research article

Evaluation of radiological risk associated with local building materials commonly used in Northwestern Nigeria

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ABSTRACT

In this study, potential radiological risk due to the activity concentrations of primordial radionuclides (^{226}Ra, ^{232}Th, and ^{40}K) in commonly used local building materials (sand, clay, kaolin and gypsum) in Northwestern Nigeria were assessed using NaI (Tl) detector. The measured activity concentrations ranged from 47 to 63 Bq kg^{-1} for ^{226}Ra, 24–32 Bq kg^{-1} for ^{232}Th, and 219–257 Bq kg^{-1} for ^{40}K respectively. The mean values of ^{232}Th, and ^{40}K for all samples were below the respective world averages of 45 and 420 Bq kg^{-1} with that of ^{226}Ra for all the samples higher than the world average value of 32 Bq kg^{-1}. The potential radiological risks were assessed by determining radium equivalent activity (Ra_{eq}), internal and external hazard indices (H_{in} and H_{ex}), absorbed gamma dose rates (D_R), internal annual effective dose rates (IAED), and annual gonadal dose equivalent (AGDE) and activity utilization index (AUI). The assessed parameters were found to range between 104 and 125 Bq kg^{-1}, 0.99 to 1.15, 0.28 to 0.34, 48 to 58 nGyh^{-1}, 0.76 to 0.86 mSvy^{-1}, and 0.78 to 0.96 respectively. The Ra_{eq} and D_R for all the analyzed samples were found to be within International recommended limits of 370 Bq kg^{-1} and 59 nGy h^{-1} as recommended by UNSCEAR.

1. Introduction

Radiation exposure to humans in everyday lives usually comes from the ground, building materials, air, water, the universe and even radionuclides within their bodies [1]. Natural radiation sources are the major source of radiation exposure to the general public, which have been in existence since the creation on of the earth [2].

Terrestrial radionuclides found on the earth usually came into existence with the creation of the planet [3]. Some of these radionuclides take a very long time to decay and become non-radioactive (about hundreds of millions of years), and are still part of the human and non-human biota system [3].

These radionuclides are known to have originated from the earth crust and then find their way into the environment via different media such as soil, air, water, and building materials [4]. It is evident that these raw materials (e.g., sand, soil, cement, tiles, marble etc.) that are used for building human dwellings, recreational centres, schools, hospitals as well as used for radiation shielding

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purposes contains natural radionuclides such as, ^{238}U, ^{232}Th and ^{40}K in different proportions based on their geological origins [5,6]. These materials have been evidently found to be associated with radiation emission, which was as a results of the radionuclides transmitted to them by the main parent material used in producing them [7].

Humans may likely be exposed to radiation from these building materials through external as well as internal pathways [8]. This is because some of them were sourced from different areas serving as potential sources of indoor radioactivity [6], which may likely have a very serious negative impact on the exposed individuals [6].

Due to the detrimental health effects associated with gamma radiation emitted from building materials and the growing concern among the scientific community, several researchers were engaged in assessing the activity concentrations of the natural radionuclides in such materials across the globe, with few reported for Nigeria [4–6,8–27].

In this study, the radiological risk associated with local building materials commonly used in Northern Nigeria was assessed in order to have information on the possible radiological risks to human due to the usage of such materials for building construction and other related purposes. The result obtained will serve as a beneficial addition to the databases, especially studies that addressed local building materials, that are commonly used by majority of the populace dominant in rural arears and in some urban settlement, but were never assessed before. The study can further stress the Nigerian Government effort towards local content promotion as well as further compliment the United Nation Sustainable Development Goals (SDGs), especially SDGs 8, 11 and 15. The data can also be used in the establishment of national standards for the use and management of building materials in light of global recommendations. In addition, the results regarding the measured activity concentrations can be used in recommending the samples to be properly utilized for possible radiation shielding purposes.

2. Methodology

2.1. Sample collection and preparation

Four different commonly used local building materials in Northwestern Nigeria were collected with the aid of local suppliers, the areas (Zamfara, Sokoto and Kebbi) fall within Cretaceous geological formations (which is of Sedimentary rock origin), while Katsina, Kano, Jigawa and Kaduna fall within Precambrian basement (which originates from Metamorphic and Igneous rocks) [28,29]. About 2 kg of the selected materials namely; clay, sand, kaolin, and gypsum were collected, weighed and packed in a pre-cleaned plastic polyethylene bags and transported to the laboratories at the Physics Department and Centre for Energy Research and Training, Ahmadu Bello University for preparation and analysis. The collected samples were oven dried at 105 °C for 48 h in order to remove moisture contents and then allow to cool down to room temperature and later crushed, grounded and then sieved for homogeneity by passing through a 500 and 250 μm mesh.

2.2. Measurements of ^{226}Ra, ^{232}Th, and ^{40}K concentrations and gamma radiation dose rate

The prepared samples were analyzed for the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K using NaI (Tl) gamma ray detector situated at the Health Physics section, Centre for Energy Research and Training, Ahmadu Bello University, Zaria. The detector consist of a 7.62 × 7.62 cm NaI (Tl) detector housed in a 10 cm thick lead-shielded, cadmium-lined assembly with copper sheets for the reduction of background radiation, which was equipped with a MAESTRO computer system program for data acquisition and spectra analysis. IAEA standard reference materials (RGK-1, IAEA-448 and RGTh-1) were used for the quantitative analysis of the ^{226}Ra, ^{232}Th, and ^{40}K respectively [30]. Each of the prepared sample was counted for approximately 8 h with the activity concentration of ^{226}Ra determined using the gamma energy line of ^{214}Bi (1760 keV), ^{232}Th using ^{208}Tl (2614 keV), and while the activity concentration of ^{40}K was determined directly from its 1460 keV gamma line. Energy and efficiency calibration was carried out using a point source a 500 mL of ^{60}Co (1173 and1332 keV), ^{241}Am (59.54 keV), ^{137}Cs (661.62 keV) multi-nuclides standard solution respectively. The net number of counts under each photo peak of interest was then background subtracted using the time correct spectrum taken using the blank container. The specific activity concentration was calculated using Equation (1) [31].

$$A\,(\mathrm{Bq}/\mathrm{kg}) = \frac{C}{\varepsilon t p M} \tag{1}$$

where C is the count rate, ε is efficiency, p is transition probability, M is the mass of each sample in kg and t is measurement time.

2.3. Radiological hazards assessment

2.3.1. Radium equivalent (Ra_{eq})

Radium equivalent Ra_{eq} (Bq kg^{-1}) is a radiological risk index that presents the activity levels of ^{226}Ra, ^{232}Th, and ^{40}K by a single quantity taken into account the associated radiation hazard, was calculated using Equation (2) [1,14].

$$Ra_{eq}\left(\mathrm{Bq\ kg^{-1}}\right) = A_{Ra} + 1.43A_{Th} + 0.077A_K \tag{2}$$

2.3.2. Internal and external hazard index (H_{in}, H_{ex})

The main aim of hazard index is to assess the risk of natural gamma radiation and keep it below unity [32]. The internal and external hazard indices gives the internal exposure by the population to carcinogenic radon and were computed using Equations (3) and (4) [33,34].

$$H_{in} = \left(\frac{A_{Ra}}{185}\right) + \left(\frac{A_{Th}}{259}\right) + \left(\frac{A_K}{4810}\right) \leq 1 \tag{3}$$

$$H_{ex} = \left(\frac{A_{Ra}}{370}\right) + \left(\frac{A_{Th}}{259}\right) + \left(\frac{A_K}{4810}\right) \leq 1 \tag{4}$$

where A_U, A_{Th} and A_K are activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K, respectively in Bq kg^{-1}. The value of this index should be below unity in order for the radiation risk to be acceptable for the public.

2.3.3. Gamma radiation dose rate (D_R)

The main source of gamma radiation in the environment was terrestrial sources, which means that, there exist a strong link between the terrestrial gamma radiation and the radionuclide contents. The absorbed dose rates in air 1 m above the ground was calculated using Equation (5) from the measured values of ^{226}Ra, ^{232}Th, and ^{40}K activities using the factors 0.462, 0.604, and 0.0417 (nGy h^{-1} per Bq kg^{-1}) [35].

$$D_R\ (\text{nGy h}^{-1}) = 0.462A_{Ra} + 0.604A_{Th} + 0.0417A_K \tag{5}$$

where A_U, A_{Th}, and A_K are the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K respectively in Bq kg^{-1}.

2.3.4. Internal annual effective dose (AED)

The annual effective dose due to the activity concentrations of the natural radionuclides in the building materials was calculated using Equation (6) with the dose coefficient (0.7 Sv Gy^{-1}) and occupancy factor 0.8 for indoors [35]. An indoor annual effective dose was considered here because people spend more than 80% of their time indoors.

$$IAE\ \left(\text{mSvy}^{-1}\right) = D_R\left(\text{nGyh}^{-1}\right) \times 24 \times 365 \times 0.8 \times 0.7 \times 10^{-6} \tag{6}$$

2.3.5. Annual gonadal dose equivalent

UNSCEAR considers some organs with rapidly dividing cells such as gonads, bone marrow and bone surface cells as organs of interest [35,36]. The annual gonadal equivalent dose (AGDE) is a measure of the genetic significance of the yearly dose received by the population's reproductive organs. The annual gonadal dose equivalent (AGDE) due to the activities of ^{226}Ra, ^{232}Th and ^{40}K was

Table 1
Mean specific activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in the analyzed samples.

Sample Code	State	Activity of natural radionuclides (Bq kg^{-1})			^{232}Th:^{226}Ra
		^{226}Ra	^{232}Th	^{40}K	
KN CLAY	Kano	51 ± 4	24 ± 2	238 ± 19	0.5
KN SAND		55 ± 4	27 ± 2	238 ± 19	0.5
KN KAOLIN		59 ± 5	27 ± 2	238 ± 19	0.4
KN GYPSUM		55 ± 4	24 ± 2	223 ± 17	0.4
KD CLAY	Kaduna	55 ± 4	27 ± 2	242 ± 19	0.5
KD SAND		59 ± 5	24 ± 2	242 ± 19	0.4
KD KAOLIN		55 ± 4	27 ± 2	238 ± 19	0.5
KD GYPSUM		55 ± 4	27 ± 2	219 ± 18	0.5
KT CLAY	Katsina	59 ± 5	32 ± 3	247 ± 20	0.5
KT SAND		63 ± 6	24 ± 2	257 ± 21	0.4
KT KAOLIN		55 ± 4	27 ± 2	242 ± 19	0.5
KT GYPSUM		51 ± 5	29 ± 2	242 ± 19	0.6
KB CLAY	Kebbi	55 ± 5	27 ± 2	238 ± 19	0.5
KB SAND		63 ± 6	29 ± 2	257 ± 21	0.5
KB KAOLIN		59 ± 5	29 ± 2	252 ± 20	0.5
KB GYPSUM		55 ± 4	27 ± 2	238 ± 19	0.5
JG CLAY	Jigawa	47 ± 4	29 ± 2	247 ± 20	0.6
JG SAND		59 ± 5	29 ± 2	247 ± 20	0.5
JG KAOLIN		55 ± 4	29 ± 2	238 ± 19	0.5
JG GYPSUM		55 ± 4	29 ± 2	238 ± 19	0.5
SK CLAY	Sokoto	59 ± 5	27 ± 2	247 ± 20	0.4
SK SAND		55 ± 4	29 ± 2	252 ± 20	0.5
SK KAOLIN		51 ± 3	27 ± 2	242 ± 19	0.5
SK GYPSUM		51 ± 3	27 ± 2	238 ± 19	0.5
ZM CLAY	Zamfara	55 ± 4	29 ± 2	238 ± 19	0.5
ZM SAND		55 ± 4	27 ± 2	242 ± 19	0.5
ZM KAOLIN		55 ± 4	24 ± 2	238 ± 19	0.4
ZM GYPSUM		59 ± 5	29 ± 2	247 ± 20	0.5

estimated using Equation (7) [35].

$$\mathrm{AGDE}\left(\mu\mathrm{Sv\ y}^{-1}\right) = 3.09\mathrm{A_{Ra}} + 4.18\mathrm{A_{Th}} + 0.314\mathrm{A_K} \tag{7}$$

2.3.6. Activity utilization index (AUI)

This estimate the dose rate in air from different combinations of the various radionuclides and was determined by Equation (8) [36, 37].

$$AUI = \left(\frac{A_{Ra}}{50Bqkg^{-1}}\right) \times f_{Ra} + \left(\frac{A_{Th}}{50Bqkg^{-1}}\right) \times f_{Th} + \left(\frac{A_K}{500Bqkg^{-1}}\right) \times f_K \tag{8}$$

where A_{Ra}, A_{Th} and A_K are the respective activity concentrations of ^{238}U, ^{232}Th and ^{40}K while f_{Ra}, f_{Th}, and f_K are the fractional contributions of the radionuclides to the total dose rate in air due to gamma radiation and with respective values of 0.462, 0.604 and 0.041.

3. Results and discussion

3.1. Activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in the analyzed samples

The mean measured activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in the analyzed building materials commonly used in the Northwestern part of Nigeria are presented Table 1.

It can be observed from the table that, the measured activity concentrations ranges from 47 to 63 Bq kg^{-1} with a cumulative mean values of 56 ± 4 Bq kg^{-1} for ^{226}Ra, 24 to 32 Bq kg^{-1} with a mean value of 27 ± 2 Bq kg^{-1} for ^{232}Th, and 219 to 257 Bq kg^{-1} with a mean value of 242 ± 19 Bq kg^{-1} for ^{40}K respectively. The mean values of ^{232}Th, and ^{40}K for each of the 28 analyzed samples were below the respective world averages of 45 and 420 Bq kg^{-1} with that of ^{226}Ra for all the samples higher than the world average value of 32 Bq kg^{-1} as set by UNSCEAR [35].

It can also be seen from the table that the activity concentration of ^{226}Ra in clay for all the samples were about 1.5–2 times higher than the world average value of 32 Bqkq^{-1} with the highest value of 63 Bq kg^{-1} each recorded in Katsina and Kebbi sand samples respectively. Furthermore, the activity concentrations of ^{232}Th and ^{40}K in all the analyzed samples were all below the world average values of 45 and 420 Bq kg^{-1} respectively. With the highest for ^{232}Th recorded in Katsina clay sample with a value of 32 Bq kg^{-1} and that of ^{40}K was recorded in Katsina and Kebbi sand samples with respective value of 257 Bq kg^{-1} each. The mean value of thorium to uranium concentration ratio (^{232}Th: ^{226}Ra) for all the samples were found to be below the world value of 1.4 given by UNSCEAR [35], this means that ^{232}Th activity concentration is slightly lower than that of ^{226}Ra.

Generally, it can be observed from Table 1 that, the activity concentration of ^{40}K was found to be the highest in all the analyzed samples of the four commonly used building materials within the study area, then followed by ^{226}Ra with ^{232}Th having the lowest. This clearly indicates that ^{226}Ra and ^{40}K are the major sources of gamma radiation in the studied samples and this is in good agreement with a similar study conducted in Kerala, India [38]. Additionally, the relatively high amount of ^{40}K observed in all the samples though not above the world average value, can be attributed to the abundance of potassium silicate minerals in the geological formations of the study area and the intense agricultural activities in the environment.

In addition, a descriptive statistics of the different building materials, correlation and ANOVA analysis of the overall results was conducted and presented in Tables 2–4.

It can be seen from Table 2 (descriptive statistics results) that clay samples have a mean value of 54 Bq kg^{-1} for ^{226}Ra, 28 Bq kg^{-1}

Table 2
Descriptive statistics of activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K.

Material	Statistics	Activity Concentration (Bqkg^{-1})		
		^{226}Ra	^{232}Th	^{40}K
Clay	Average	54.4	27.9	242.4
	Min	47	24	238
	Max	59	32	247
	SD	4.3	2.5	4.5
Sand	Average	58.4	27	247.9
	Min	55	24	238
	Max	63	29	257
	SD	3.6	2.2	7.6
Kaolin	Average	55.6	27.1	241.1
	Min	51	24	238
	Max	59	29	252
	SD	2.8	1.7	5.1
Gypsum	Average	54.4	27.4	235
	Min	51	24	219
	Max	59	29	247
	SD	2.8	1.8	10.2

for ^{232}Th, and 242 Bq kg^{-1} for ^{40}K respectively. While sand, have mean value of 58 Bq kg^{-1} for ^{226}Ra, 27 Bq kg^{-1} for ^{232}Th, and 242 Bq kg^{-1} for ^{40}K respectively. In addition, kaolin and gypsum have respective mean values as; 56 Bq kg^{-1} for ^{226}Ra, 27 Bq kg^{-1} for ^{232}Th, and 241 Bq kg^{-1} for ^{40}K and 54 Bq kg^{-1} for ^{226}Ra, 27 Bq kg^{-1} for ^{232}Th, and 235 Bq kg^{-1} for ^{40}K respectively.

From Tables 3 and it can be seen that there is a weak correlation between the radionuclides, thereby indicating that they exist independently and from different sources.

The ANOVA test further corroborated the correlation analysis as it shows that there exist is significant difference between the radionuclides since the p-value is greater than 0.05, thus rejecting the null hypothesis (Table 4).

3.2. Radiological hazards assessment

The radiological hazard parameters due to the radionuclides in the studied building materials were determined using Equations (2)–(8) and presented in Table 5.

It can be seen from the table that the Ra_{eq} varies between 104 and 125 Bq kg^{-1}, with a mean value of 113 Bq kg^{-1}. The highest value of Ra_{eq} with a value of 125 Bq kg^{-1} was found to be in Kebbi sand sample (KB sand) with clay sample collected in Kano with a code KN clay, having the lowest with a value of 104 Bq kg^{-1}. The mean values for all the samples are within recommended value of 370 Bq kg^{-1}. This implies that, the annual radiation dose due to the exposure to gamma radiation from the building materials is within the recommended value of 2.4 mSv.

In this study, the mean internal and external hazard indices varied from 0.99 to 1.15, and 0.28 to 0.34 respectively. It can be seen that the mean values were all within the recommended value of unity [32], which means the building materials are not radioactive, as such, they can be used for building purposes effectively as well as other related purposes e.g. radiation shielding under well regulated radiation shielding protocol as stipulated in both international and national guidelines.

The mean absorbed dose rates due to gamma radiation in air associated with the samples was found to range between 48 and 58 nGy h^{-1} which is within the mean range of 55–59 nGy h^{-1} set by UNSCEAR [39]. Therefore, the samples are free away from radiological contamination and may pose an insignificant radiological threat to the users and the entire population.

The indoor annual outdoor effective dose due to the measured radionuclides was found to vary between 0.76 and 0.89 mSv y^{-1} which are slightly higher than the worldwide mean value of 0.5 mSv y^{-1} due to the terrestrial gamma radiation [35], with sand collected from Kebbi with a code KB sand having the highest value (0.89 mSv y^{-1}). Furthermore, the annual gonadal dose equivalent (AGDE) and the activity utilization index (AUI) were found to vary between 333 and 382 μSvy^{-1} and 0.78 to 0.96 respectively. The mean AGDE values obtained for all the samples were found to exceed the allowable limit value of 300 μSvy^{-1}, thus, signifying the existence of significant radiochemical risk to the gonads.

Finally, based on the radiological risk parameters obtained in this study, the four commonly used building materials within the study area are relatively free from radiological contamination; as such, they can be highly recommended for building purposes and are therefore been recommended for further analysis to determine their potentials to be use for radiation shielding purpose.

4. Conclusion

Radiological studies of 28 samples of 4 commonly used building materials (clay, sand, kaolin and gypsum) in Northern Nigeria was conducted with the aim of investigating their suitability to be assessed for radiation shielding application based on the determined radiological risk parameters levels. The mean activity of ^{226}Ra, ^{232}Th and ^{40}K were found ranged from 47 to 63 Bq kg^{-1} with a cumulative mean values of 56 ± 4 Bq kg^{-1} for ^{226}Ra, 24 to 32 Bq kg^{-1} with a mean value of 27 ± 2 Bq kg^{-1} for ^{232}Th, and 219 to 257 Bq kg^{-1} with a mean value of 242 ± 19 Bq kg^{-1} for ^{40}K respectively. The assessed radiological risk parameters (radium equivalent, internal and external hazard index, absorbed gamma dose rates, internal annual effective dose rates, annual gonadal dose equivalent and activity utilization index) were found to range between 104 and 125 Bq kg^{-1}, 0.99 to 1.15, 0.28 to 0.34, 48 to 58 nGyh^{-1}, 0.76 to 0.86 mSvy^{-1}, and 0.78 to 0.96 respectively. In addition, ^{226}Ra and ^{40}K were found to be the major contributor to the natural radiation in the samples. Even though, ^{226}Ra was found to have values higher than the world average, going by the radiological risk parameters, it can still be concluded that, the samples are relatively free from radiological contamination.

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Table 3
Correlation analysis between ^{226}Ra, ^{232}Th and ^{40}K.

	^{226}Ra	^{232}Th	^{40}K
^{226}Ra	1		
^{232}Th	0.025034	1	
^{40}K	0.426614	0.318752	1

Table 4
ANOVA results of the measured radionuclides.

Source of variation	SS	df	MS	F	P-value	F crit
Rows	1112.482	27	41.20304	1.339331	0.226396	1.904823
Columns	642642.9	1	642642.9	20889.52	1.51E-40	4.210008
Total	644,586	55				

Table 5
Summary of the radiological hazard parameters.

Sample Code	Radiological Hazard Parameters						
	Ra_{eq} (Bq kg^{-1})	H_{in}	H_{ex}	D_R ($nGyh^{-1}$)	IAED ($mSvy^{-1}$)	AGDE (μSvy^{-1})	AUI
KN CLAY	104	1.05	0.28	48	0.76	333	0.78
KN SAND	111	1.06	0.30	51	0.81	356	0.85
KN KAOLIN	115	1.06	0.31	53	0.82	368	0.89
KN GYPSUM	106	0.99	0.29	49	0.77	340	0.82
KD CLAY	112	1.08	0.30	52	0.82	357	0.85
KD SAND	112	1.07	0.30	52	0.83	358	0.85
KD KAOLIN	111	1.06	0.30	51	0.81	356	0.85
KD GYPSUM	110	0.99	0.30	51	0.83	350	0.85
KT CLAY	124	1.13	0.33	57	0.87	393	0.95
KT SAND	117	1.12	0.32	54	0.86	375	0.89
KT KAOLIN	112	1.08	0.30	52	0.81	357	0.85
KT GYPSUM	112	1.09	0.30	51	0.84	356	0.85
KB CLAY	111	1.06	0.30	51	0.82	356	0.85
KB SAND	125	1.15	0.34	58	0.89	397	0.96
KB KAOLIN	120	1.13	0.32	56	0.86	384	0.92
KB GYPSUM	111	1.06	0.30	51	0.81	356	0.85
JG CLAY	108	1.11	0.29	50	0.86	346	0.81
JG SAND	120	1.11	0.32	55	0.84	382	0.92
JG KAOLIN	115	1.08	0.31	53	0.87	367	0.88
JG GYPSUM	115	1.08	0.31	53	0.86	367	0.88
SK CLAY	116	1.10	0.31	54	0.83	371	0.89
SK SAND	116	1.13	0.31	54	0.84	371	0.88
SK KAOLIN	108	1.08	0.29	50	0.82	345	0.81
SK GYPSUM	107	1.06	0.29	50	0.79	344	0.81
ZM CLAY	115	1.08	0.31	53	0.82	367	0.88
ZM SAND	112	1.08	0.30	52	0.81	357	0.85
ZM KAOLIN	108	1.05	0.29	50	0.80	345	0.82
ZM GYPSUM	120	1.11	0.32	55	0.80	382	0.92
World Average	**370**	**1**	**1**	**59**	**1**	**300**	**1**

Author contribution statement

Nuraddeen Nasiru Garba, BSc, MSc, PhD: Funding acquisition; Conceived and designed the experiments; Analyzed and interpreted the data; Wrote the paper.

Rabiu Nasiru: Conceived and designed the experiments; Reviewed the paper; Supervision; Manuscript review & editing.

Alhassan Saad Aliyu: Sample collection and preparation; Performed the experiments.

Usman Musa Kankara: Performed the experiments.

Abdullahi Muhammad Vatsa, BSc, MSc; Aminu Ismaila; Suleiman Bello: Contributed reagents, materials, analysis tools or data.

Data availability statement

Data included in article/supp. material/referenced in article.

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