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Activity concentrations and radiation hazard in the soil of Ashaka Cement PLC Gombe state, Nigeria with a multivariate statistical approach

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Abstract
Humans are exposed externally and internally in their environment to natural background radiation from radionuclides such as ^{40}K, ^{238,} U and ^{232}Th which emit gamma, alpha, beta and other forms of radiation through their decay processes. These radionuclides are found in the soil, food and the water we consume. Human activities such as the mining of uranium, oil exploration, building materials, coal-fired power station can ignite the Naturally Occurring Radioactive Materials (NORMs). The aim was to assess radiation impact in Ashaka cement factory, Gombe state for their potential radiological hazards in order to maintain a safe environment. A prospective cross-sectional survey design was adopted and using a purposive sampling technique, fifteen (15) soil samples were collected using a geographical positioning system (GPS). The activity concentrations of these natural radionuclides were measured due to Gamma radiation by using a High Purity Germanium detector. Samples were analysed at the Radiation Protection Institute of Ghana Atomic Energy Commission, Kwanbeya District, at Accra, Ghana. Fifteen (15) soil samples were collected from within the Ashaka Cement factory (Zone 1) and environment (Zone 2). Data was analyzed by the use of SPSS Version 22.0 (IBM Corp. Armonk NY, 2011). Descriptive statistics was employed to obtain the Mean and Standard deviation of the radioactivity concentrations. Inferential statistics was carried out to compare the radionuclides and radiological parameters. Statistical significance was considered at $p \leq 0.05$. The mean activity concentrations of ^{238}U, ^{232}Th and ^{40}K in soil samples at Zone 1 are 42.70 Bqkg^{-1}, 29.00 Bqkg^{-1}, 375 Bqkg^{-1} and Zone 2 is 46.11 Bqkg^{-1}, 35.00 Bqkg^{-1}, 780.30 Bqkg^{-1}respectively. Radiation hazard indices at Zone 1 and 2 shows the mean for External Hazard index, Internal Hazard index, Gamma Index, External Absorbed Dose Rate, Annual Effective Does Rate, External Annual Effective Does Rate, Excess Lifetime Cancer Risk and Annual Gonadal Effective Dose is 0.31, 0.42, 0.83, 38.80nGyh^{-1}, 0.32x10^{-3}, 0.048mSvyr^{-1}, 0.17mSvyr^{-1}, 371mSvyr^{-1} and 0.42, 0.55, 1.18, 45.70nGyhr^{-1}, 0.46mSvyr^{-1}, 0.056mSvyr^{-1}, 0.20x10^{-3}, 533.80mSvyr$_{-1}$ respectively. This implies that both staffs and members of the public in the factory might be at risk due to cumulative effect of radiation exposure high activity concentration from 238U and Annual Gonal effective dose exceeding the world average limits.

Keywords: Activity concentration, natural radionuclide, soil, radiological parameters

Introduction
Radiation from Radionuclides have been an integral part of the earth since creation [1]. Gamma radiation which is part of the electromagnetic spectrum is of high frequency [2] but very short wavelength is a product of decay of high energy atomic nuclei and also energy sub-atomic particle interactions through natural processes and man-made process [2, 3]. Radionuclides can be categorized based on their source as: Naturally Occurring Radioactive Materials (NORMs), Technologically Enhanced Natural Occurring Radioactive Materials (TENORMs) and Man-made radionuclides [1]. Sources of manmade radionuclides consist of nuclear tests, nuclear power plants, sources used for health purpose, industries and agricultural applications, and those used for research. Both NORMs and TENORMs emanates from the same congenital origin, activities such as tobacco smoking, oil exploration, mining and milling of uranium, air transportation, coal-fired power station, and so on generate the TENORMs [1, 4].

Industries giving rise to TENORM discharges include fossil fuel (e.g. Coal), cement production, nuclear power stations, metal processing, oil and gas exploration, titanium Oxide, phosphate industry, pigment production, zirconium and rare earth process [5].

Cement production is in this sequence; heating and calcining of blended raw materials, especially limestone, clay or shale along with other substance to form the clinker [5]. This heating of clinker takes place at a material temperature of 1450°C in kilns. The clinker is mixed with little amounts of gypsum to obtain Portland cement. Blended cements are in addition produced from cement clinker with blast furnace slag and fly-ash [5]. Due to the significant temperature of the raw materials in the kilns, volatilisation of ^{210}Po and ^{210}Pb is the main possible source of aerial discharge [5, 6]. Stated that for several years, processing and mining industry burden the environment seriously. The volatile isotopes ^{222}Rn, ^{210}Po and ^{210}Pb are discharged in the gaseous or precipitate form and particulate matter, increasing the activity concentration around these industrial plants. The stages, according to [4] employed in the production of cement includes appraisal of location for abundance of limestone which is a key substance in cement production, gypsum and shale through a geophysical approach, quarrying, grinding and blending of the clinker, as well as packaging and transportation of the finished products. Sensitive among all the stages to human health is the quarrying stage [7].

Though exposure from natural radiation to humans is one of the inevitable phenomena on earth as released radionuclides to the soil and water are majorly from weathering of parent-rock and other geological processes [8], researchers have shown that mining and quarrying increase the level of radioactivity in an environment [7 4]. Also, quarry activities increases the dose of radiation received by humans via the distribution of the radionuclides that are associated with the natural resources to the soil surfaces [4].

In Ashaka Cement vicinity, quarrying and mining activities are high because of the ongoing cement production from the factory, there is therefore the possibility of having underground water contamination because of the gamma, beta and alpha radiation emitters. Information on radioactivity distributions could be utilized for assessing the environmental and monitoring of a contaminated areas [9].

Ashaka Cement, also called the Star of the North, was established in 1974 and located in Funakaye Local Government Area of Gombe State. The community has abundant deposits of limestone and other minerals necessary for cement production. It is bounded by Bage, Gongilla, Badabdi, Bungum, Feshingo, Maza, Ashaka-Gari and the nearest to the Ashaka Cement Site, Jalingo villagge. Ashaka factory is a subsidiary of Lafarge-France Company and is involved in the manufacturing and marketing of Cement Products in Nigeria [10]. It covers approximately 1,415 km^2, 99 kilometers from the metropolis and 21 km from the local government. It has a population of 237, 687 as at 2006 and 278, 930 as at 2011[10]. This work is aimed at assessing the radiation impact in Ashaka Cement PLC and the environment

Materials and Methods

Radiation Hazard Indices

Hazard index is a term used to describe the sum of more than one hazard quotient for multiple substance and multiple exposure pathways. The hazard index is calculated for chronic, sub chronic, and shorter duration exposures. Potentially, non-carcinogenic effects are characterized by comparing projected doses to reference doses. The hazard quotient estimates that result is a ratio. The ratio of the intake to the reference dose (hazard index) is compared to unity (1.0). If the hazard quotient is greater than one, then the effects are assumed to be of concern [11]. The radiological hazard quotients are the activity concentration of potassium-40, radium-226 and thorium-232. The radiation hazard index in this study therefore represents the summation of the effects of the activity concentration of potassium-40, thorium-232 and radium-226 in soil [11].

External Hazard Index

It is represented by equation [11, 12].

$$H_{ex}=Ra_{eq}/370=A_k/4810 + A_{Ra}/370 + A_{Th}/259 \qquad 2.14$$

Where $A_{k,}$ A_{Ra} and A_{Th} are the specific activities in $BqKg^{-1}$ respectively of K^{40}, U^{238} and Th^{232} in the equation [11, 13].

Internal Hazard index

This is represented by equation [11, 12]

$$H_{in}= A_k/4810+ A_{Ra}/185 + A_{Th}/259 \qquad 2.15$$

A_k, A_{Ra} and A_{Th} is the activity concentration of K^{40}, U^{238} and Th^{232} respectively. Also, $H_{in}\leq 1$

Representative gamma index (I_γ)

This index is introduced by [14] and calculated by the following formula

$$I_\gamma= A_k/3000 + A_{Th}/200 + A_{Ra}/300 \qquad 2.16$$

A_k, A_{Ra} and A_{Th} is the activity concentration of K^{40}, U^{238} and Th^{232} respectively

For a safe material; $I_\gamma\leq 1$. This corresponds to annual effective dose equal or less than 1 mSv while $I_\gamma\leq 0.5$ corresponds to an annual effective dose equal or less than 0.3mSv [11, 12].

External dose rate

The external dose received from radionuclide to the general public at a distance of 1m above the surface [12] is estimated from the measured specific activities of these nuclides by the following relationships.

$$D^*=0.0417_{AK}+ 0.462_{ARa} +0.604_{ATh} \qquad 2.17$$

Where D^*= Dose rate in $nGyh^{-1}$ at 1m above the ground surface. $A_{k,}$ $A_{Ra,}$ and A_{Th} are the specific activities in $BqKg^{-1}$ of K^{40}, U^{238} and Th^{232} respectively. The other radionuclides eg. ^{137}C, ^{90}Sr and those present in the ^{235}U decay series, can be neglected as they contribute very little to the total dose from the environmental background [13]. The average value of background dose rate from soil is 59$nGyh^{-1}$ [13].

Effective dose rate

The absorbed dose rate in air due to measured specific activities of these primary radionuclides in the sample has been converted to effective dose rate by the following relationship [12, 14, 15].

$E = D^{*} \text{x } O \text{ x } C \text{ x } 8760 \text{ x } 10^{-6}$……………….………….2.18

Where:
E= Effective dose rate in $mSvy^{-1}$
D^{*}= Absorbed dose rate in air at 1m above the sample surface, $nGyh^{-1}$
O= 0.2 (Outdoor occupancy factor)
C= 0.7 $SvGy^{-1}$ (conversion factor from the absorbed dose in air to the effective dose received by an adult person). The average background outdoor effective dose rate from soil for an adult person is 0.07mSv [12, 15].

Excess lifetime cancer risk (ELCR)
The ELCR was evaluated using the AEDE values as shown in equation according to [16, 17, 18].

$ELCR = AEDE\ (mSvy{-}1)\ x\ DL\ x\ RF$……………..… (2.19)

Where DL is average duration of life (70 years) and RF is the fatal cancer risk factor per Sievert (Sv^{-1}). For low dose background radiation, which is considered to produce stochastic effects [16],

Annual Gonadal Equivalent Dose
It is essential to estimate the annual gonadal equivalent dose (AG), because it is the parameter that predicts whether the gonad, bone cells and marrow of humans are safe after exposure to γ-radiation or not [4]. The AG as a result of contributions from the activity concentrations of uranium, thorium and potassium was determined using equation [4].

AGED (μSV y-1) = $(A_U \times 3.09) + (A_{Th} \times 4.18) + (A_K \times 0.314)$ ...(2.21)

A_k, A_{Ra} and A_{Th} is the activity concentration of K^{40}, U^{238} and Th^{232} respectively

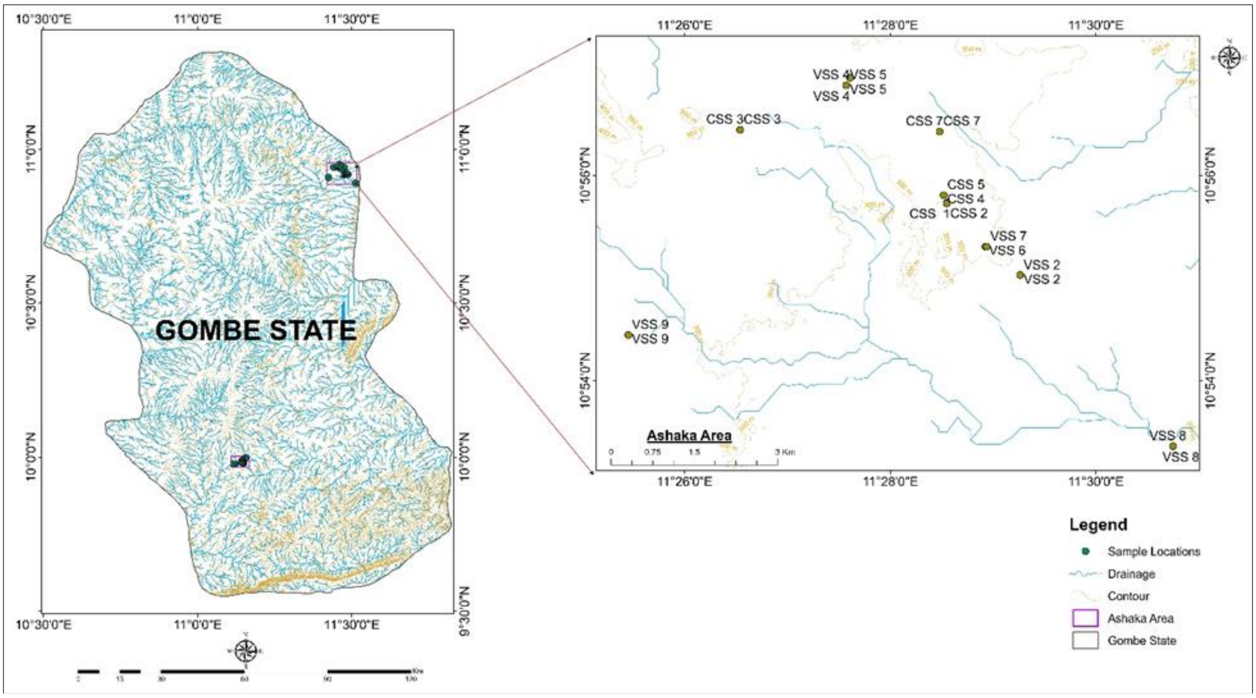

Fig 1: Ashaka Cement PLC. The GPS coordinates of sampling points were used to produce a Location map of the study areas as shown above

Results and Discussion
The mean concentration of ^{238}U in both Zones are higher comparable to the world mean value (33 $BqKg^{-1}$) while that of ^{232}Th and ^{40}K are less than the world mean value in zone 1 which is 45 Bq kg^{-1}, and 420 $Bqkg^{-1}$ respectively [15]. This may be as a result of the abundant nature in this region of the country as was recently discovered. This is in agreement the work of [18, 19, 21] where the mean uranium content reported was higher than the world average value in soil. But this is contrary to [22] who recorded a lower value and this may be attributed to the differences in location where the samples were collected or compared with other studies was believed to be because these radiation elements are not evenly spread in the earth crust [4]. Also the mean value of ^{40}K is markedly high in zone 2 with the highest concentration of ^{40}K in this study and was obtained at VSS6. The high value for ^{40}K agrees with the several studies conducted locally [23, 7], also noticed in their work on the assessment of radioactivity concentration in soil of some mining areas in central Nasarawa state, Nigeria, that ^{40}K has the largest contribution in specific activities of all the samples analyzed. The high level in ^{40}K may be attributed to the influence of different farm practices like the use of fertilizers for improving soil fertility due to poor yield of farm products around these areas [24].
The radiation hazard indices show low value for external hazard index, internal hazard index and representative gamma index. They are all less than unity, implying the radiation impact is not significant for the population living in the investigated vicinity as far as soil source is concerned. These findings are in agreement with [26, 27, 28] in their studies, the calculated indices were less than unity. The result obtained indicates that all sample locations are within acceptable limit, been less than unity. The result obtained for the external absorbed dose rate, Annual External Effective Dose Rate, Annual Effective Dose Rate, The Excess Lifetime Cancer Risk obtained indicates that all sample locations are within acceptable limit which shows no obvious threat in the both zones. The values for the Excess Lifetime Cancer Risk ranges were found to be less than limit of 0.29×10^{-3} set by [15] and also 0.05 recommended by the

International Commission on Radiological Protection (ICRP) for low-level radiations. For the Annual Gonadal Equivalent Dose from these study, results show 371.077 $mSvyr^{-1}$ and 533.80 $mSvyr^{-1}$ from both zones are more than the world acceptable limit of 300$mSvyr^{-1}$ [13]. These high values are in agreement with work of [4] on radioactivity concentration and dose assessment of soil samples in cement factory and environs in Ogun state, Nigeria where virtually all the points at the factory and environment showed elevations more than the global mean. The result for the annual gonadal doses at the factory and the environment reveal that the workers may be exposed to radiological risk, which might pose bad effect on the functionality of organs [29].

Strong positive relationship was found to exist between ^{238}U and ^{232}Th (r^2=0.53 in zone 1 and r^2=0.620 in zone 2), while very weak degree of association existed between ^{40}K and ^{238}U (r^2=0.127 in zone 1 and r^2=-0.115 in zone 2), and between ^{40}K and ^{232}Th (r^2=0.180 in zone 1 and r^2= 0.009). The very strong positive correlation existing between ^{238}U and ^{232}Th may not be unconnected with the fact that Uranium and thorium decay series have a common origin, and exists together in nature [20, 28, 30]. Furthermore, all the estimated radioactive variables were strongly correlated with one another, and also with Uranium and ^{232}Th. On the other hand, ^{40}K exhibited very weak relationship with most of the radiological variables. This indicated that the emission of gamma radiation is principally due to ^{238}U and ^{232}Th contents in soil samples from Ashaka Cement Factory and environs.

Results

Table 1: Natural Radionuclides and their activity concentration (Bq/kg) for soil within the Ashaka Cement Plant (Zone 1)

S/N	Code	U-238 (Bq/kg)	Th-232 (Bq/kg)	K-40 (Bq/kg)
1	CSS 1	31	16	244.1
2	CSS 2	25	18	121.8
3	CSS 3	43	53	386.6
4	CSS 4	36	18	285.2
5	CSS 5	62	28	945.0
6	CSS 7	59	41	272.4
		42.7±15.1	29±15.1	375.8±29.5

Key: Cement Soil Sample (CSS)

Table 2: Radiation Hazard Indices within Ashaka Cement Plant (Zone 1)

S/N	Code	H_{ex} (External Hazard Indices)	H_{in} (Internal Hazard Indices)	Representative Gama index (Ir)	Ext Absorbed Dose Rate (DE)	Annual Effective Dose Rate (E)	Ext. Annual Effective Dose (AEDE)	Excess Lifetime Cancer Risk (ELCR)	Annual Gonadal Equivalent Dose (AG)
1	CSS 1	0.20	0.28	0.53	25.00	0.21	0.031	0.12	239.32
2	CSS 2	0.16	0.23	0.43	22.93	0.17	0.028	0.10	190.74
3	CSS 3	0.40	0.52	1.07	53.49	0.42	0.066	0.23	475.80
4	CSS 4	0.23	0.32	0.61	28.69	0.24	0.035	0.12	276.03
5	CSS 5	0.47	0.64	1.32	49.50	0.52	0.061	0.21	605.35
6	CSS 7	0.37	0.53	0.99	53.16	0.39	0.065	0.23	439.22
		0.31	0.42	0.83	38.80	0.32	0.048	0.17	371.08

Key: Cement Soil Sample (CSS)

Table 3: Natural Radionuclides and their activity concentration (Bq/kg) for soil the Ashaka factory environment (ZONE 2)

S/N	Code	U-238 (Bq/kg)	Th-232 (Bq/kg)	K-40 (Bq/kg)
1	VSS 1	58	42	1033.3
2	VSS 2	25	33	696.0
3	VSS 3	31	40	853.5
4	VSS 4	59	45	749.6
5	VSS 5	30	28	295.4
6	VSS 6	57	43	1124.8
7	VSS 7	56	31	839.2
8	VSS 8	23	15	1098.2
9	VSS 9	76	38	332.7
	mean	46.1±18.99	35.0±18.99	780.3±303.3

KEY; Village Soil Sample (VSS)

Table 4: Radiation Hazard Indices in Ashaka factory environment (Zone 2)

S/N	Code	H_{ex} (External Hazard Indices)	H_{in} (Internal Hazard Indices)	Representative level index(I_r)	Ext Absorbed Dose Rate (DE)	Annual Effective Dose Rate (E)	Ext Annual Effective Dose (AEDE)	Excess Lifetime Cancer Risk (ELCR)	Annual Gonadal Equivalent Dose (AG)
1	VSS 1	0.53	0.69	1.50	56.47	0.58	0.069	0.24	679.24
2	VSS 2	0.34	0.41	0.96	34.38	0.37	0.042	0.15	433.73
3	VSS 3	0.42	0.50	1.18	42.04	0.45	0.052	0.18	530.99
4	VSS 4	0.49	0.65	1.34	57.56	0.53	0.071	0.25	605.78
5	VSS 5	0.25	0.33	0.68	32.00	0.26	0.039	0.14	302.50
6	VSS 6	0.55	0.71	1.56	56.99	0.61	0.070	0.25	709.06
7	VSS 7	0.45	0.60	1.24	48.10	0.49	0.059	0.21	566.13
8	VSS 8	0.35	0.41	1.04	24.27	0.40	0.030	0.11	478.60
9	VSS 9	0.42	0.63	1.11	59.45	0.44	0.073	0.26	498.15
		0.42±0.10	0.55±0.14	1.18±0.27	45.70±13.08	0.46±0.11	0.056±0.016	0.20±0.56	533.80±125.46

Key; Village Soil Sample (VSS)

Table 5: Correlation matrix Ashaka Zone 1

	U-238 (Bq/kg)	Th-232 (Bq/kg)	K-40 (Bq/kg)	H_{ex} (Ext Hazard Indice)	H_{in} (Internal Hazard Indices)	Ext Absorbed Dose Rate (DE)	Representative Gama index (Ir)	Annual Effective Dose Rate (E)	Ext. Annual Effective Dose (AEDE)	Excess Lifetime Cancer Risk (ELCR)	Annual Gonadal Equivalent Dose (AG)
U-238	1										
Th-232	0.533	1									
K-40	0.127	0.180	1								
H_{ex}	0.912	0.715	0.794	1							
H_{in}	0.950	0.681	0.791	0.995	1						
D	0.915	0.664	0.836	0.997	0.993						
(DE)	0.863	0.885	0.538	0.939	0.936	1					
(Ir)	0.909	0.675	0.831	0.998	0.992	0.916	1				
(E)	0.915	0.666	0.834	0.998	0.994	0.914	1.000	1			
(AEDE)	0.861	0.886	0.543	0.942	0.937	1.000	0.918	0.916	1		
(ELCR)	0.915	0.664	0.836	0.997	0.993	0.913	1.000	1.000	0.915	1	
(AG)	0.911	0.648	0.850	0.995	0.991	0.902	0.999	1.000	0.904	1.000	1

Table 6: Correlation Matrix for Ashaka Zone 2

	U-238 (Bq/kg)	Th-232 (Bq/kg)	K-40 (Bq/kg)	H_{ex} (Ext Hazard Indices)	H_{in} (Internal Hazard Indices)	Ext. Absorbed Dose Rate (DE)	Representative Gama index (Ir)	Annual Effective Dose Rate (E)	External Annual Effective Dose (AEDE)	Excess Lifetime Cancer Risk (ELCR)	Annual Gonadal Equivalent Dose (AG)
U-238	1										
Th-232	0.620	1									
K-40	-0.115	0.009	1								
H_{ex}	0.682	0.706	0.588	1							
H_{in}	0.857	0.730	0.371	0.961	1						
D	0.619	0.643	0.667	0.995	0.934						
DE	0.931	0.854	0.024	0.823	0.930	1					
Ir	0.588	0.636	0.690	0.991	0.919	0.739	1				
E	0.624	0.642	0.664	0.995	0.936	0.763	0.998	1			
AEDE	0.930	0.854	0.028	0.825	0.931	1.000	0.742	0.766	1		
ELCR	0.651	0.607	0.655	0.991	0.943	0.766	0.993	0.996	0.768	1	
AGED	0.576	0.612	0.708	0.987	0.912	0.722	0.999	0.998	0.725	0.993	1

Conclusion

The natural radionuclides present are ^{238}U, ^{232}Th and ^{40}K with higher activity concentration of ^{238}U exceeding the world permissible limits of 33 Bq/kg in all zones of Ashaka Cement PLC and environs. All Radiation Hazard indices are within world permissible limits except the Annual Gonadal effective dose with values consistently higher than the world limits of 300mSvyr^{-1} in all zones. Inferential statistics of Pearson correlation among the natural radionuclide and other radiological variables have shown strong positive relationship between ^{238}U, and ^{232}Th indicating they are of same origin and the greatest contributor to gamma radiation in the all the zones. These two radionuclide has also shown weak positive and negative relationship with ^{40}K and the other variables. The radiation hazards revealed that all the parameters are below the global standard, but the annual gonadal equivalent doses and mean Uranium value from the factory and environment, which are higher than the world average values. Therefore, there might be a stochastic effect on over exposure to γ-radiation to staff and dwellers as a result of contributions from ^{40}K and AGED respectively.

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