EVALUATION OF RADIOACTIVITY DISEQUILIBRIUM AND LIFETIME EXCESSIVE CANCER PROBABILITY DUE TO NATURALLY OCCURRING RADIONUCLIDES IN WASTES DUMPSITES SOILS IN AGBARA, SOUTHWEST, NIGERIA.

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Abstract

The concentration and spatial distribution of the natural and artificial gamma ray emitting ^{238}U, ^{232}Th, ^{40}K and ^{137}Cs radionuclides in dumpsite soils in Agbara were analyzed with the aim of evaluating the radiation hazards and excessive Lifetime cancer risk using well calibrated HPGe γ-ray spectrometry technique. The ranges of activity concentrations of ^{238}U, ^{232}Th, ^{40}K and ^{137}Cs are 11.45 ± 1.02 – 165.73 ± 39.92 $Bqkg^{-1}$, 15.63 ± 1.81 – 31.36 ± 2.28 $Bqkg^{-1}$ 20.39 ± 1.32 -365.62 ± 29.82 $Bqkg^{-1}$ and 0.52 ± 0.07 – 4.39 ± 0.06 $Bqkg^{-1}$ respectively. Radiological parameters such as absorbed dose rate, radium equivalent, annual effective dose equivalent, internal and external hazard indices, gamma level index, activity utilization index, annual genetic significant dose equivalent, exposure rate and excessive lifetime cancer risk were calculated to know the complete radiological hazardous nature of the dumpsite soils to the inhabitants of the sites. The calculated radiological parameters were higher than the world average value in most of the sample locations. The mean activity concentrations, radium equivalent and dose rate of measured radionuclides were compared with other literature values. The ratio of the detected radioisotopes was calculated for spatial distribution of natural radionuclides in the study area. RESRAD computer code was applied to calculate the total effective dose equivalent (TEDE). The code was also used to calculate the probability of excess lifetime cancer incurred by dwellers/inhabitants of the dumpsites, the level of which was determined to be 3.2×10^{-4} for Ibijola dumpsite soils and 2.7×10^{-4} for Idowale dumpsite soils over a period of 30 years respectively. Therefore, the radiological risks to the general populations from Waste enhanced naturally occurring radioactive materials (WENORM) from the selected dumpsites soils are considered to be significant. Multivariate analyses (Pearson Correlation, Principal component analysis and Factor analysis) were carried out between parameters obtained from radioactivity to know the existing relations between the radionuclide and it associated radiological parameters.

Keywords: Spatial distribution, radiation hazards, radioisotopes and radiological risk.

1.0 Introduction

There has long been concern about the issue of soil pollution by radionuclides and heavy metals because of their toxicity for plant, animal and human beings and their lack of biodegradability. Soil is a primary recipient of many of the waste products and chemical used in modern society. Soil is the primary reservoir of radionuclides and other pollutants in the atmosphere, hydrosphere and biota, and thus plays a fundamental role in the overall nuclide cycle in nature (Cao *et al.,* 2010). Radioactive materials in soil pose potential threats to the environment and can damage human health through various absorption pathways such as direct ingestion, dermal contact, and diet through the soil–food chain, inhalation, and oral intake (Lu *et al.,* 2011). Soil is an important environmental media that sustain life, one of the ways soil can be polluted is through improper disposal of hazardous wastes.

Human activities create wastes and the way that these are handled, stored, collected and disposed can give rise to impacts on the environment and public health (Ademola, Morounfolu and Peter, 2015). Hazardous waste can cause pollution, damage to health and even death. The environmental problem posed by solid wastes which are improperly disposed in most cities and town has been of concern to federal, state and local authorities in Nigeria. Poor waste management poses several challenges to the well-being of city residents due to the potential of the waste to pollute water, food sources, land, air and vegetation (Njoroge, 2007), when the wastes are not properly managed or disposed (Porteous, 1985).

Hazards posed by such waste dumpsite are not only in term of odour and presence of disease causing micro-organism, but can arise from the radiation emanating from such dumpsite (Ojoawo *et al.*, 2011), which occur as results of accumulation and reaction of different radioactive materials in the waste indiscriminately dumped on open waste site. At waste dumpsites, there are possibilities for radiation to be emitted due to the presence of radioactive waste in the landfills as well as naturally occurring radionuclides in the soil. The radioactive contamination of soil, water and air can be transferred to human through the soil via plants (^{40}K) or through inhalation (^{222}Rn and ^{220}Rn). These radionuclides even at low concentrations can have potential impacts on the environmental quality and human health and may pose a long term risk (Ademola, Morounfolu and Peter, 2015). Moreover, risk assessment which is an effective scientific tool will enables decision makers to manage sites so contaminated in a cost-effective manner while preserving public and ecosystem health (Zhao and Kaluarachchi, 2002). Therefore, objectives of this study was to determine the concentration of waste enhanced naturally occurring radionuclides (WENORM) present in representative soil

samples from some selected waste dumpsites in Agbara by gamma-ray spectrometry in order to estimate the, radioactivity disequilibrium, excessive lifetime cancer and other radiological hazard indices from these dumpsites to the general public and provide a reliable baseline data for future radionuclide evaluation in the area.

2.0 Material and Methods

2.1 Description of the Study area

Agbara which is an industrially populated town in Ogun State is located within latitude $6^0 20\,0$ N and $6^0 35\,0$ North of the Equator and longitude $3^0 5\,0$ E and $3^0 10\,0$ East of the Greenwich Meridian (Figure 1). The area stands on a low–lying gent undulating terrain with altitude ranging between 30 and 80 m above sea level. The area is characterized by high annual temperature, high rainfall, high evapotranspiration and high relative humidity which make it to be classified as humid tropical region (Akanni, 1992). The prevailing wind direction was Southwesterly at the period of study (wet season period–June–October) while the prevailing wind speed ranged range between 2.52 to 3.55 ms^{-1} and in the dry season. The prevailing wind direction in the dry season (November to March) is Northeasterly. Two major municipal dumpsites soils were used and they are Ibijola dumpsite along Ibijola hospital way and Idowale Road dumpsite.

2.2 Sample coding

The soil samples were coded as follows in order to prevent identification error. The sample codes consist of four alphabets. The first alphabet stands for the study area (Agbara), the second alphabet stands for representative soil samples (A, B, & C), and the last two alphabets stands for the soil layer type e.g. TS which connotes Top Soil and SS which connotes Sub Soil. All soil samples were controlled with a control sample different from the above stated soil samples, this was done in order to know the contamination level and dose rate of radionuclide in these dumpsite soils compared to that of the anthropogenic free soil (control soil samples).

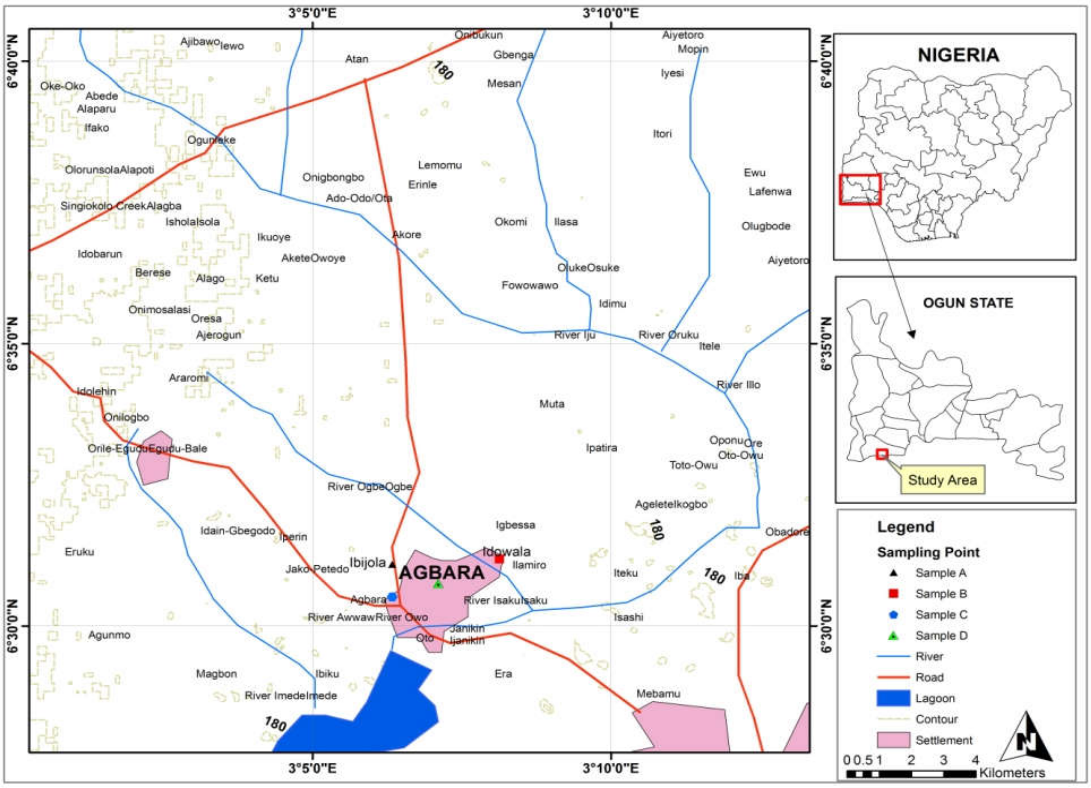

Figure 1: Map of the study area showing sample locations

2.3 Sample Collection and Preparations for Radiochemical Analysis

With a garden rake, the waste was removed to expose the soil under the waste dumps from where samples were collected. Five sets of soil samples were collected from each of the dump sites. The soil samples were taken at about 0 – 15cm depth (using a meter rule) by the use of hand-driven auger, and taken to the laboratory in labeled polythene bags stored and were air-dried at laboratory temperature in order to avoid cross contamination or pollution of the samples. The samples were then pulverized by grinding, and filtered through a 2mm mesh sieve. Two hundred grams (200 g) of pulverized soil samples was subsequently measured using an analytical weighing balance with a precision of ± 0.01 g and packed into cylindrical containers (beaker) which were carefully labeled. These samples were safely conveyed to Natural Institute of Radiation Protection and research, University of Ibadan, Ibadan, South – West Nigeria, at the laboratory the plastic were hermitically sealed with adhesive tape and kept for minimum of 30 days to ensure that the parent and daughter nuclide in the sample were at secular equilibrium between radium and its gaseous decay progenies. At the end of

the four weeks in-growth period, the samples were subjected to gamma-ray spectroscopy counting.

3. Radioactivity measurements

3.1 System used for measurements

Activity concentration ^{238}U, ^{232}Th and ^{40}K were measured by gamma-ray spectrometry using NaI (TI) detector. The counting system used in the determination of the natural radionuclide contents of the soil consists of 7.6 cm x 7.6 cm NaI (TI) detector (Model Bicron) couple to Canberra 10 multi-channel analyzer with adequate lead shielding which reduced the background by a factor of about 95%. The spectrometer was tested for its linearity and calibrated for energy and efficiency using the well calibrated standard gamma source obtained from an International Atomic Energy Agency (IAEA), laboratories, Vienna, Austria (Ademola, Hammed & Adejumobi, 2008). Efficiency is the measure of percentage of radiation at a given detector detect from the overall yield that is emitted from the source into a solid angle of usually 4π in the photo-peaks (Hossain *et al.*, 2012). Accuracy of efficiency calibration of detector is necessary to obtain the high precision measurements with radioactive samples. The resolution of the detector is 7.5 % at 0.662 MeV of ^{137}Cs. This resolution is capable of distinguishing the gamma ray energies of interest in the study. All the samples were counted for 36000 sec in order to obtain good statistics for uranium, thorium daughter products and ^{40}K. Also measurements were repeated at intervals for quality assurance purposes as well as to ascertain the stability of the measuring system. The background radiation due to the naturally occurring radionuclides in the environment around the detector was measured by using an empty plastic container; the empty plastic container was measured in the same manner as the soil samples for the same counting time of 3600s (10 hours). The background spectrum was subtracted from the measure spectra to obtain the net radionuclides activities. The background, reference sample and the soil samples were measured under the same conditions (Jibiri *et al.,* 2014). Three regions of interest in the spectrum were identified. These were centered on the three characteristic photopeaks at approximately 1.460 MeV for (^{40}K), 1.760 MeV for (^{214}Bi) and 2.614 MeV for (^{208}TI) in the samples was obtained. These were used for evaluating the activity levels of ^{40}K, ^{238}U and ^{232}Th series respectively. The activity concentration (A_c) of ^{238}U, ^{232}Th and ^{40}K in $Bqkg^{-1}$ were obtained using the relation in equation 1 below (Amrani & Tahtat, 2001):

$$\text{Sample activity } (A_C)\ (\text{Bq } kg^{-1}) = \frac{C_i}{\varepsilon(E)\ x\ t\ x\ m} \quad (1)$$

Where C_i is the net peak area after subtraction of background of the gamma-ray line at energy E, ε (E) is the detector efficiency of such gamma-ray line, t is the time of measurement in seconds and m is the mass of the sample in kg. The below detectable limit (BDL) of each radionuclide were determine from the background radiation spectrum for the same counting time for the dumpsites soil samples.

The detection limits (DL) which is required to estimate the minimum detectable activity in a sample and will be obtained using Equation 2

$$LLD\ (Bqkg^{-1}) = 4.65\frac{\sqrt{C_b}}{t_b}\ f \quad (2)$$

Where C_b is the net background count in the corresponding peak, t_b is the background counting time (s), and f is the factor that converts cps (counts per second) to activity concentration (Bq kg^{-1}).

Radiation Hazard Parameters

In order to estimate the radiation hazards incurred by the population due to the activity levels of the measured waste enhanced naturally occurring radionuclides (WENORM) in the selected dumpsite soils in Agbara Industrial town, some radiation hazard indices were calculated and the details calculations are given in Table 2. These are used to reassess the statistical information about excessive lifetime cancer risk (ELCR), γ-ray absorbed dose rate (D_R), Outdoor and Indoor Annual Effective dose equivalent ($AEDE_{outdoor}$ & $AEDE_{indoor}$), Annual gonadal dose equivalent (AGDE), Radium equivalent (Ra_{eq}), activity utilization index (AUI), Exposure rate (ER), internal and external hazard indices (H_{int} & H_{ext}) and External (γ-radioactivity) level index ($I_{\gamma r}$) for the present study. Even though the total activity concentration of radionuclides is calculated, it does not provide the exact indication about the total radiation hazards due to uneven distribution of the waste enhanced naturally occurring radionuclide (^{40}K, ^{238}U and ^{232}Th) in the soil. Statistics of all the calculated radiological parameters and their recommended levels by UNSCEAR (2000) are given in table 3. Distributions of the above radiological parameters are shown in Fig 2 and 3.

Table 1: Summary of the radiological parameters of all the dumpsite soil samples

S/N	Radiological parameters	Units	Used formula[a, b]
1	Absorbed Dose Rate (D_R)	$nGyhr^{-1}$	$D_R = (0.462A_U + 0.604A_{Th} + 0.0417A_K)$
2	Radium equivalent (Ra_{eq})	$Bqkg^{-1}$	$Ra_{eq} = (A_u + 1.43A_{Th} + 0.077A_k)$
3	External Hazard index (H_{ext})	-	$H_{ext} = {A_U}/{370} + {A_{Th}}/{259} + {A_K}/{4810} \leq 1$
4.	Internal Hazard index (H_{int})	-	$H_{int} = {A_U}/{185} + {A_{Th}}/{259} + {A_K}/{4810} \leq 1$
5	Annual effective dose equivalent ($AEDE_{outdoor}$)	$\mu Svyr^{-1}$	AEDE (outdoor) $= D_R \times 8766 \times 0.7\ Sv/Gy \times 0.2 \times 10^{-3}$
6	Annual effective dose equivalent ($AEDE_{indoor}$)	$\mu Svyr^{-1}$	AEDE (indoor) $= D_R \times 8766 \times 0.7\ Sv/Gy \times 0.8 \times 10^{-3}$
7	Annual gonadal dose equivalent (AGDE)	$\mu Svyr^{-1}$	AGDE $= 3.09\ A_U + 4.18\ A_{Th} + 0.314\ A_K$
8	Gamma level index (Iγr)	-	$I\gamma r = {A_U}/{300} + {A_{Th}}/{200} + {A_K}/{3000} \leq 1$
9	Activity utilization index (AUI)	-	$AUI = \left\lfloor {A_U}/{50} \right\rfloor f_U + \left\lfloor {A_{Th}}/{50} \right\rfloor f_{Th} + \left\lfloor {A_K}/{500} \right\rfloor f_K \leq 2$
10	Exposure Rate (ER)	μRhr^{-1}	ER $(\mu Rhr^{-1}) = 1.90\ A_U + 2.82\ A_{Th} + 0.179\ A_K$
11	Excess lifetime cancer risk (ELCRoutdoor)	-	ELCR = AEDEoutdoor x DL x RF

[a] UNSCEAR (2000) and [b] (Al-Trabulsy, Khater & Habbani, 2011). Where A_U, A_{Th} and A_K are the activity concentrations of ^{238}U, ^{232}Th and ^{40}K in ($Bqkg^{-1}$) present in waste dumpsites soil respectively. f_U (0.462), f_{Th} (0.604) and f_K (0.0417) are the fractional contributions to the total dose rate due to γ-radiation from the actual radionuclide of ^{238}U, ^{232}Th and ^{40}K, respectively. DL and RF is duration of life (70 years) and risk factor (Sv^{-1}), fatal cancer risk per sievert. For stochastic effects, ICRP 60 uses values of 0.05 for the public.

4. Results and Discussion

The results obtained from NIRPR shows different activity concentration of the radionuclides as shown below.

Table 2. Geological Location and Activity Concentration of Radionuclides (A_c) & Absorbed Dose Rate value for different soil samples

			Activity Concentrations ($Bqkg^{-1}$)				
Sample ID	Latitude	Longitude	^{238}U	^{232}Th	^{40}K		
^{137}Cs	D_R ($nGyhr^{-1}$)	Raeq ($Bqkg^{-1}$)					
AATS1	06°11.140′N	03°52.735′E	165.73 ± 39.92	28.76 ± 2.49	365.62 ± 29.82		
7.13 ± 0.14	109.18 ± 21.19	235.01 ± 45.78					
AATS2	06°11.151′N	03°33.618′E	139.52 ± 30.06	29.18 ± 1.96	276.56 ± 22.46		
8.44 ± 0.16	93.62 ± 16.01	202.54 ± 34.59					
ABTS1	06°11.562′N	03°31.614′E	17.80 ± 1.16	29.94 ± 2.57	38.84 ± 2.37		
2.94 ± 0.15	27.93 ± 2.19	63.60 ± 5.02					
ABTS2	06°11.729′N	03°21.641′E	11.45 ± 1.02	15.63 ± 1.81	36.10 ± 2.18		
3.82 ± 0.11	16.24 ± 1.66	36.58 ± 3.78					
AASS1	06°11.140′N	03°52.735′E	12.93 ± 0.61	27.14 ± 2.01	79.48 ± 4.57		
2.83 ± 0.16	25.68 ± 1.69	57.86 ± 3.84					
AASS2	06°11.213′N	03°41.636′E	17.01 ± 1.14	26.28 ± 2.24	85.97 ± 4.91		
3.93 ± 0.14	27.32 ± 2.08	61.21 ± 4.72					
AASS3	06°11.186′N	03°48.596′E	14.58 ± 0.83	28.09 ± 2.52	76.06 ± 4.38		
4.39 ± 0.06	26.87 ± 2.09	60.61 ± 4.77					
ABSS1	06°11.638′N	03°10.558′E	22.15 ± 2.07	31.36 ± 2.28	57.96 ± 3.44		
1.31 ± 0.31	31.59 ± 2.48	71.46 ± 5.60					
ABSS2	06°11.729′N	03°21.641′E	12.01 ± 0.92	20.98 ± 2.08	39.86 ± 2.40		
1.89 ± 0.22	19.88 ± 1.78	41.08 ± 4.08					
ABSS3	06°11.562′N	03°15.526′E	16.32 ± 0.94	31.06 ± 2.30	54.07 ± 3.26		
0.52 ± 0.70	28.55 ± 1.96	64.90 ± 4.48					
Control	-	-	13.45 ± 0.57	17.87 ± 1.66	20.39 ± 1.32		
1.93 ± 0.11	17.86 ± 1.32	40.57 ± 3.05					
Min.			11.45 ± 1.02	15.63 ± 1.81	20.39 ± 1.32		
0.52 ± 0.07	16.24 ± 1.66	36.58 ± 3.78					
Max.			165.73 ± 39.92	31.36 ± 2.28	365.62 ± 29.82		
4.39 ± 0.06	109.18 ± 21.19	235.01 ± 45.78					

Range				10.70 ± 1.50	15.73 ± 0.91	76.17 ± 4.14	3.87 ± 0.00	92.94 ± 19.53 198.43 ± 42.00
Mean				42.95 ± 7.87	26.84 ± 2.20	111.05 ± 7.98	2.74 ± 0.21	40.69 ± 5.31 89.49 ± 11.67
World Average				33[a]	45[a]	420[a]		
				55[a]		370[a]		

[a] UNSCEAR (2000); [b] (Al-Trabulsy, Khater & Habbani, 2011).

Table 2 above shows the activity concentration of naturally and artificially occurring radioactive elements ^{238}U, ^{232}Th, ^{40}K and ^{137}Cs determined in the dumpsite soil samples. The activity concentration ranges for ^{238}U, ^{232}Th, ^{40}K and ^{137}Cs are 11.45 ± 1.02 – 165.73 ± 39.92 $Bqkg^{-1}$, 15.63 ± 1.81 – 31.36 ± 2.28 $Bqkg^{-1}$, 20.39 ± 1.32 – 365.62 ± 29.82 $Bqkg^{-1}$, and 0.52 ± 0.07 – 4.39 ± 0.06 $Bqkg^{-1}$ respectively. When comparing the activity concentration of radionuclides with the world average value, ^{238}U is higher by a factor of 6.2 whereas ^{232}Th, ^{40}K and ^{137}Cs were below the average value. It was observed from the results that the activity concentration values are in the order ^{137}Cs ^{232}Th ^{238}U ^{40}K in all sampling sites. Similar observation has been reported by Ademola, Morounfolu and Peter (2015). Higher activity concentrations of the radionuclides were obtained in the top soil samples from Ibijola dumpsites (Fig 2). Higher concentration of ^{40}K might be due to the higher silica content that generally occurs in the soil (Navarrete, Zuniga, Espinosa and Golzarri, 2014).

Radium equivalent (Ra_{eq}) which is used to describe the γ-output from different mixtures of ^{238}U, ^{232}Th and ^{40}K in the soil samples from the study area and it is calculated using equation in Table 1. The calculated values of Ra_{eq} ranges from 36.58 ± 3.78 – 235.01 ± 45.78 $Bqkg^{-1}$ with a mean value of 89.49 ± 11.67 $Bqkg^{-1}$. The values from all the sampling sites are below the maximum permissible level of 370 $Bqkg^{-1}$(UNSCEA, 2000). The absorbed dose rate (D_R) which is the energy imparted per unit weight of the irradiate material is also calculated using the equation in Table 1 and results shown in Table 2. The absorbed dose rate (D_R) vary from 16.24 ± 1.66 to 109.18 ± 21.19 $nGyhr^{-1}$ with an average value of 92.94 ± 19.53 $nGyhr^{-1}$.

From the present study, it is clearly revealed that the average absorbed dose rate (D_R) is higher than the recommended level of 55 nGyhr^{-}1 (UNSCEAR, 2000) by a factor of 2.04.

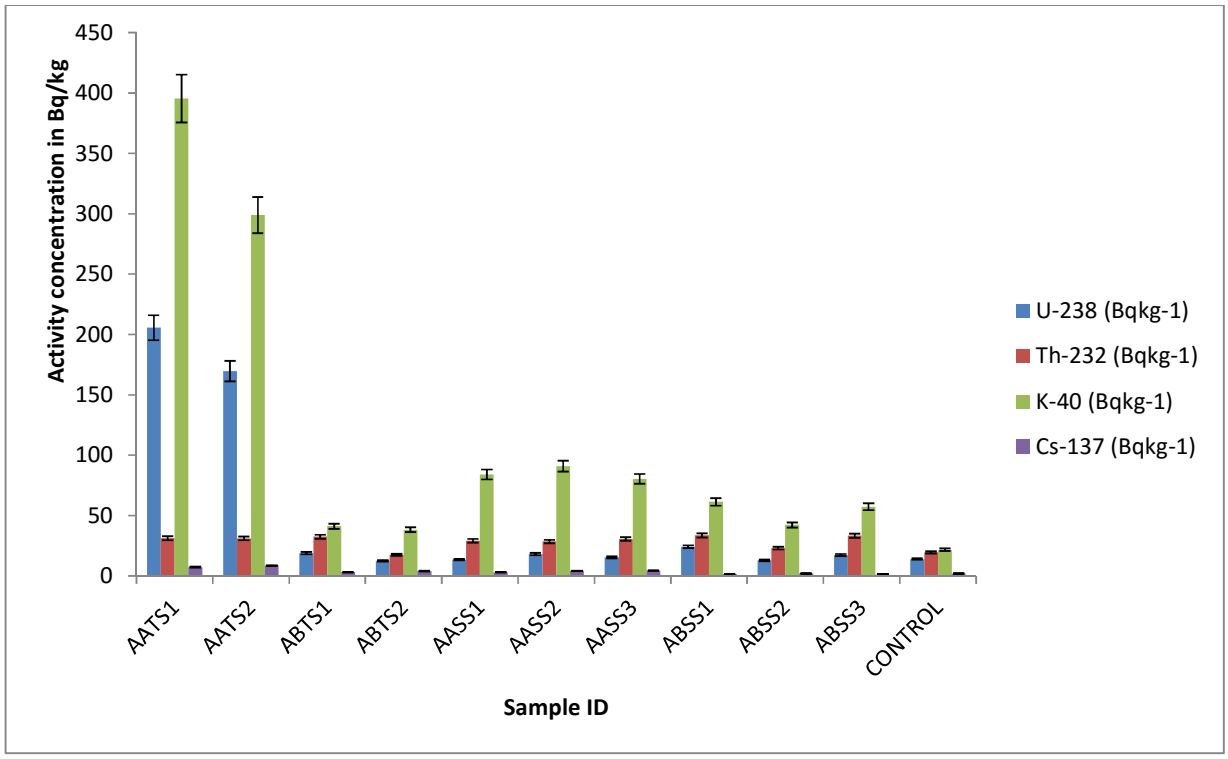

Fig. 2: Activity concentration of ^{238}U, ^{232}Th, ^{40}K and ^{137}Cs.

Table 3. Radiological parameters of Dumpsites Soils in Agbara

Sample ID	ER (μRhr^{-1})	AGDE ($\mu Svyr^{-1}$)	H_{ext}	H_{int}		Iγr
	AUI	AEDEout	ELCR x10^{-3}			
AATS1	461.90 ± 88.21	747.13 ± 143.12	0.63 ± 0.12	1.08	±	0.23
	0.82 ± 0.16	1.91 ± 0.04	133.99 ± 26.01	0.47±0.07		
AATS2	396.88 ± 66.66	639.93 ± 108.13	0.55 ± 0.09	0.92	±	0.17
	0.70 ± 0.12	1.66 ± 0.30	114.89 ± 19.65	0.40±0.06		
ABTS1	125.20 ± 9.88	192.35 ± 15.07	0.17 ± 0.01	0.22	±	0.02
	0.22 ± 0.02	0.53 ± 0.04	34.28 ± 2.69	0.12±0.01		
ABTS2	72.29 ± 7.43	112.05 ± 11.40	0.10 ± 0.01	0.13	±	0.01
	0.13 ± 0.01	0.30 ± 0.03	19.93 ± 2.04	0.07±0.01		
AASS1	115.33 ± 7.65	178.36 ± 11.72	0.16 ± 0.01	0.19	±	0.01
	0.21 ± 0.01	0.45 ± 0.03	31.52 ± 2.07	0.11±0.01		
AASS2	121.82 ± 9.36	189.41 ± 14.43	0.17 ± 0.01	0.21	±	0.02
	0.22 ± 0.02	0.48 ± 0.04	33.53 ± 2.55	0.12±0.01		
AASS3	120.53 ± 9.47	186.35 ± 14.47	0.16 ± 0.01	0.20	±	0.02
	0.21 ± 0.02	0.48 ± 0.04	32.98 ± 3.04	0.12±0.01		
ABSS1	140.90 ± 10.98	217.73 ± 17.01	0.19 ± 0.02	0.25	±	0.02
	0.25 ± 0.02	0.59 ± 0.05	38.77 ± 3.04	0.14±0.01		
ABSS2	89.12 ± 8.02	137.32 ± 12.26	0.12 ± 0.01	0.15	±	0.01
	0.16 ± 0.01	0.37 ± 0.03	24.40 ± 2.18	0.09±0.01		
ABSS3	128.28 ± 8.86	197.24 ± 13.54	0.18 ± 0.01	0.22	±	0.01
	0.23 ± 0.02	0.53 ± 0.04	35.04 ± 2.41	0.12±0.01		
Control	79.60 ± 6.00	122.66 ± 9.11	0.11 ± 0.01	0.15	±	0.01
	0.14 ± 0.01	0.34 ± 0.03	21.92 ± 1.60	0.08±0.00		
Min.	72.29 ± 6.00	112.05 ± 9.11	0.10 ± 0.01	0.13	±	0.01
	0.13 ± 0.03	0.30 ± 0.03	19.93 ± 1.60	0.07±0.00		
Max.	461.44 ± 88.21	747.13 ± 143.12	0.63 ± 0.12	1.08	±	0.23
	0.82 ± 0.16	1.91 ± 0.40	133.99 ± 26.01	0.47±0.07		
Range	389.15 ± 82.21	635.08 ± 134.01	0.53 ± 0.11	0.95	±	0.22
	0.69 ± 0.13	1.61 ± 0.37	114.06 ±24.41	0.40±0.07		
Mean	168.31 ± 21.14	265.50 ± 33.66	0.23 ± 0.03	0.34	±	0.05
	0.30 ± 0.04	0.70 ± 0.09	47.38 ± 6.12	0.17±0.02		

World average 600[a] 1000[a] ≤ 1[a] ≤ 1[a] ≤ 0.5[a] ≤ 2[a] 70[a] 0.29[a]

[a] UNSCEAR (2000); [b] (Al-Trabulsy, Khater & Habbani, 2011).

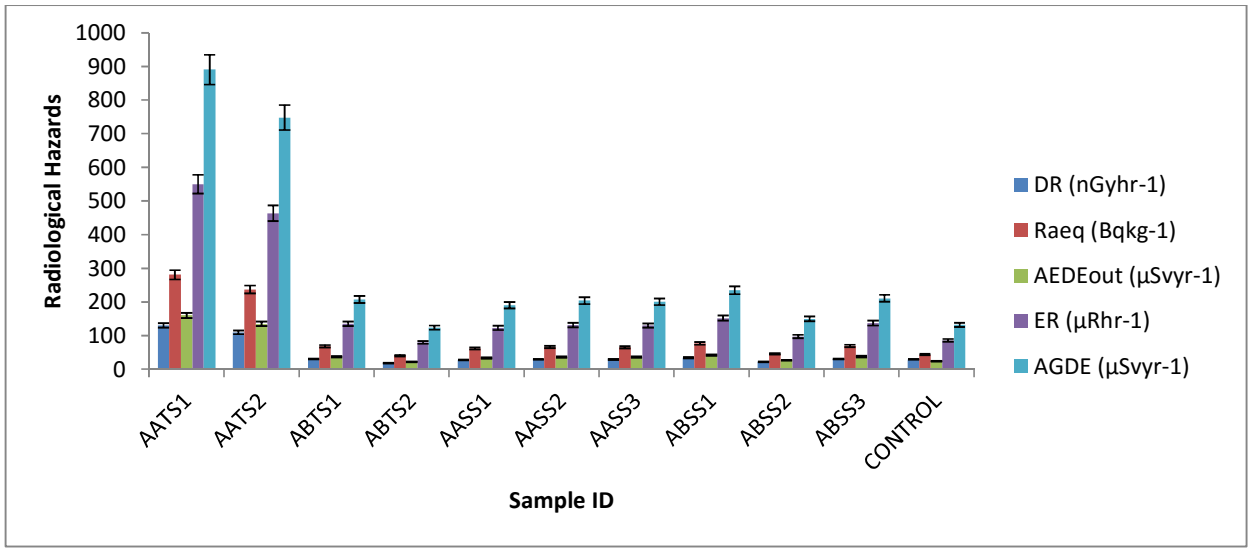

Fig. 3: Radiological Parameters

Table 4: Comparison of activity concentration and radiological hazard indices with those found in similar studies

Sample	^{238}U (BqKg^{-1})	^{232}Th (BqKg^{-1})	^{40}K (BqKg^{-1}	D_R (nGyhr^{-1})	Ra_{eq} (Bqkg^{-1})	References
Control	13.45 ± 0.57	17.87 ± 1.66	20.39 ±1.32	17.86 ± 1.32	40.57 ± 3.05	
Present study						
Agbara	42.95 ± 7.87	26.84 ± 2.20	111.05 ± 7.98	40.69 ± 5.31	89.49 ± 11.67	Present study
Lagos	69.19 ± 19.10	14.49 ± 3.22	409.44±86.08	57.80 ± 14.36	121.44 ± 30.33	Oladapo *et al.,* 2012
Sango Ota	122.10 ± 20.60	3.0 ± 1.23	3.30 ± 9.8	58.36 ± 10.67	126.64 ± 23.11	Ademola *et al.,* 2014
Ojota (Lagos)	23.1 ± 2.5	35.1 ± 2.1	318.9 ± 27.4	48.8	97.8	Ademola *et al.,* 2015

Ado Ekiti	36.57 ± 2.70	25.73 ± 5.60	758.51 ± 132.93	64.07 ±10.17
			131.77±	20.94
Isinkaye & Faweya, 2006				
Ibadan (Oritaperin)	27.93 ± 10.52	44.93 ± 7.24	488.91 ± 217.24	60.43 ± 7.41
			92.26 ± 5.60	
			Jibiri et al., 2014	
Osogbo	52 ± 6	22 ± 2	186 ± 6	45.07 ± 4.23
			97.78 ± 9.32	
Faweya & Babalola, 2010				
Port Harcourt	41.96 ± 5.53	62.61 ± 18.97	643.10 ± 5.94	84.02 ± 14.26
			181.01±33.12	Avwiri
& Olatubosun, 2014				
Crustal average	35	35	370	59
370				
			UNSCEAR, 2000.	

The ^{238}U, ^{232}Th and ^{40}K activity concentrations, Raeq and D_R for the samples from the terrestrial environment were compared with the values found in a similar study (Table 4).

4.1 Multivariate Statistical Analysis

Multivariate statistical analysis was employed to assess the relationship and interdependency among the soil characteristics because of its usefulness as a tool to reduce and organize large data sets into groups with similar characteristics without losing much information. It is widely accepted and effectively used in radioactive analysis. In the present study, the dumpsite soils naturally occurring radionuclides characteristics and its associated radiological parameters were subjected to multivariate statistical analysis (i) Pearson's correlation (ii) principal component analysis (PCA) and (iii) Hierarchical cluster analysis (HCA) was used to draw a valid conclusion regarding the interrelation among the variables using the commercial statistics software package SPSS (version 17.0). This statistical analysis was carried out in order to have a multivariate view and a good representation of the overall level of waste enhanced naturally occurring radionuclides (WENORM) and its radiological indices in the dumpsite soils. Correlation, PCA, and cluster analysis is used to explain the correlation amongst a large number of variables in terms of small number of factors without losing any information. Cluster and Pearson correlation were carried out in order to clarify the

relationship among the variables, especially the influence of soil radiological parameters on the distribution of natural radionuclides. PCA is the most common technique used to summarize patterns among variables in multivariate dataset (Sivakumar *et al.,* 2014). The PCA is a way of identifying patterns in variables, and expressing data in such a way to highlight their similarities and differences. Both PCA and Factor analysis (FA) seek to simplify data by projection from n dimensions into a line, plane or 3D graph to reduce the number of dimensions without losing information.

4.1.1 Pearson's correlation coefficients analysis

Correlation analysis was carried out in order the strength of association and direction of the linear relationship between pairs of variables through the calculation of the linear Pearson product moment correlation coefficient (r). According to the values of Pearson moment correlation coefficients (r) obtained (Table 5). The radioactive variable ^{238}U and ^{40}K shows high positive correlation coefficients with all the calculated radiological parameters in this study at 0.01 level of significant (Table 5), while there is a weakly correlation between radioactive ^{232}Th and all the radiological parameters in this study. This shows that the increase in one leads to an increase in the other calculated radiological parameters. It is also observed that all the calculated radiological parameters determined correlated highly at ($P \leq 0.01\%$) with others as except with ^{232}Th (Table 5).

Table 5: Pearson's correlation matrix for naturally occurring radionuclides and its radiological hazard indices in the dumpsite soil samples

	^{238}U	^{232}Th	^{40}K	^{137}Cs	D_R	Ra_{eq}	AEDEoutdoor	ER	AGDE	H_{ext}	H_{int}	Iγr	AUI	ELCRout
^{238}U	1	0.295	0.983**	0.859**	0.996**	0.994**	0.995**	0.994**	0.995**	0.995**	0.998**	0.994**	0.975**	0.995**
^{232}Th		1	0.349	0.152	0.347	0.394	0.383	0.392	0.383	0.389	0.347	0.398	0.447	0.388
^{40}K			1	0.869**	0.982**	0.987**	0.988**	0.988**	0.988**	0.987**	0.987**	0.988**	0.968**	0.989**
^{137}Cs				1	0.841**	0.848**	0.850**	0.847**	0.849**	0.846**	0.855**	0.845**	0.863**	0.853**
D_R					1	0.997**	0.997**	0.996**	0.997**	0.997**	0.998**	0.996**	0.975**	0.996**
Ra_{eq}						1	1.000**	1.000**	1.000**	1.000**	0.999**	1.000**	0.987**	1.000**
AEDEoutdoo							1	1.000**	1.000**	1.000**	0.999**	1.000**	0.986**	1.000**
ER								1	1.000**	1.000**	0.999**	1.000**	0.987**	1.000**
AGDE									1	1.000**	0.999**	1.000**	0.986**	1.000**
H_{ext}										1	0.999**	1.000**	0.987**	1.000**
H_{int}											1	0.998**	0.982**	0.999**
Iγr												1	0.987**	1.000**
AUI													1	0.988**
ECLCRoutdoor														1

(*) Correlation is significant at the 0.05 level (2-tailed) (**) Correlation is significant at the 0.0l level (2-tailed).

4.1.2 Principal component Analysis (PCA)

Principal component Analysis (PCA) was performed to establish the possible factors that contribute towards the radionuclides concentrations with its radiological parameters and sources apportionment. PCA was applied between the studied variables on the basis varimax orthogonal rotation with Kaiser Normalization with eigenvalue greater than 1 after Kaiser-meyer Olkin (KMO) measuring of sampling adequacy and Bartlett's test of sphericity was adequate and significant for the variables. The rotated component matrix is given in Table 6, and illustrated in Fig. 4.

Table 6: Factor analysis (after varimax rotation) showing contribution of statistically dominant variables measures in this study

Variables	Component Matrix	
	PC1	PC2
^{238}U	**0.989**	0.134
H_{int}	**0.981**	0.195
D_R	**0.977**	0.190
AEDEout	**0.974**	0.224
AGDE	**0.973**	0.226
^{40}K	**0.973**	0.184
ELCR	**0.973**	0.229
H_{ext}	**0.972**	0.231
ER	**0.971**	0.234
Ra_{eq}	**0.971**	0.235
Iγr	**0.970**	0.240
AUI	**0.950**	0.286
^{137}Cs	**0.902**	-0.050
^{232}Th	0.170	**0.979**
Eigenvalue	12.822	1.002
% of variance explained	87.140	11.004
Cumulative (%)	87.140	98.144

Significant value are given in bold

From the rotated component matrix, the eigen values and eigen vectors are extracted to explain the number of significant factors and the percent of variance explained by that factors. Table 6, shows the results of the factor loadings with a varimax rotation, as well as the eigen values and communalities. The results showed that there were two eigen values higher than one and that these two factors could explain over 98.144 % of the total variance.

Normally, an ordination result was good if the value was 75% (Zhang *et al.,* 2005). As seen from Table 6, the first component (PC1) explained 87.140 % of the total variance and loaded heavily on ^{238}U (0.989), ^{137}Cs (0.902) and all the radiological parameters. The second component (PC2) which account for 11.004 % of the total variance was correlated very strongly with ^{232}Th with a high loading value of (0.979)

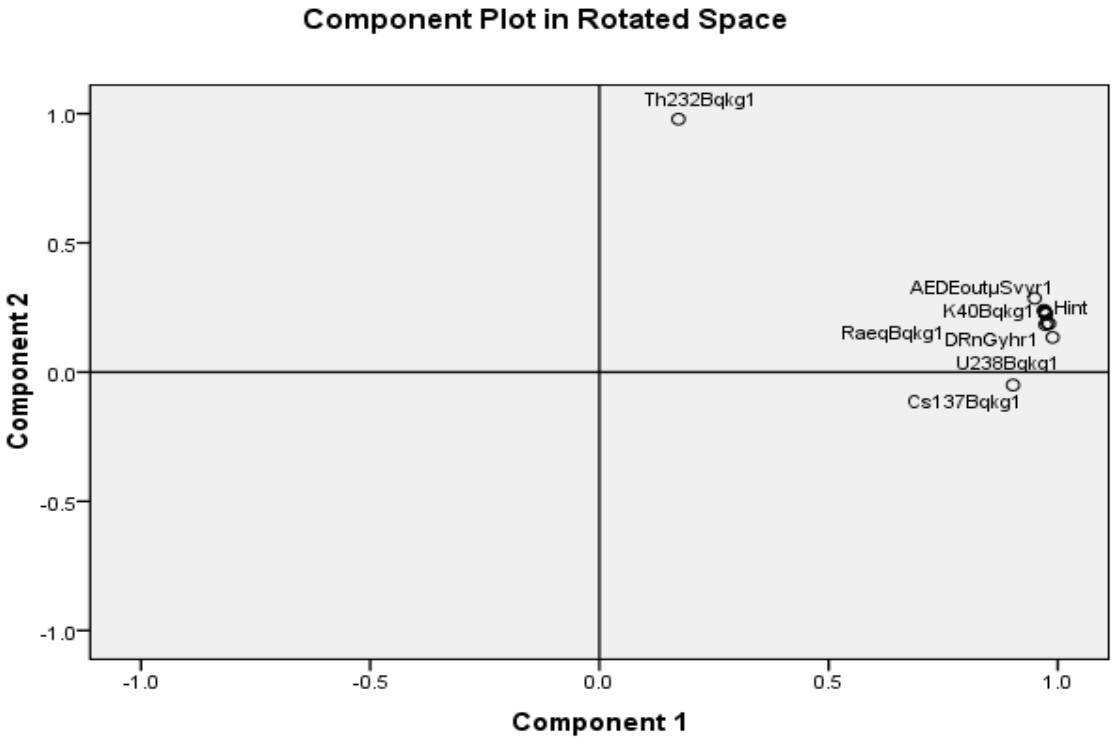

Fig. 4: Graphical representation of components PC-1 (87.140 %) and PC-2 (11.004 %).

4.1.3 Hierarchical Cluster Analysis (HCA)

Hierarchical Cluster analysis (HCA) is a multivariate technique which is used to provide important information about the grouping of variables on the basis of similarity. Cluster analysis deal with identification and classification of groups with similar characters in a new group of observations or objects. Each observation within each cluster is same but the clusters are dissimilar from each other. Hierarchical Cluster analysis was carried out through axes to identify similar characteristics among naturally occurring radionuclides and its radiological parameters in the dumpsite soils.

In HCA, the average linkage method along with coefficient distance was applied and the dendrogram was shown in Fig 5. In this dendrogram, all the three radionuclide and its

associated twelve radiological parameters were grouped into three statistically significant clusters. Clusters-I consisted of Hext, ELCRout, Iγr, AUI, ^{238}U ($Bqkg^{-1}$), AEDEout and D_R ($nGyhr^{-1}$). Cluster-II consisted of ^{137}Cs ($Bqkg^{-1}$), ^{232}Th ($Bqkg^{-1}$) series and AEDEout ($\mu Svyr^{-1}$) which is linked to Cluster-I with moderate similarity. Cluster-III consisted of ^{40}K and other radiological parameters determined that was not in Cluster-I and Cluster II above as shown in Fig 5 below. From the results obtained in this cluster analysis, it shows that ER (μRhr^{-1}), D_R ($nGyhr^{-1}$) and Ra_{eq} (Bq/kg^{-1}) depend on the radioactivity variables. Cluster II is also linked with cluster III variables with moderate similarity. This vividly shows that all other radiological parameters in cluster III depends on naturally occurring radionuclides analyzed which does not contributes to any radiological parameters in the dumpsites soils. The close relation between ^{238}U and ^{232}Th series members but not with ^{40}K was in accordance with the results obtained by (Sivakumar *et al.,* 2012; Chen, Huang and Yeh, 2001 and Elejalde *et al.,* 1996).

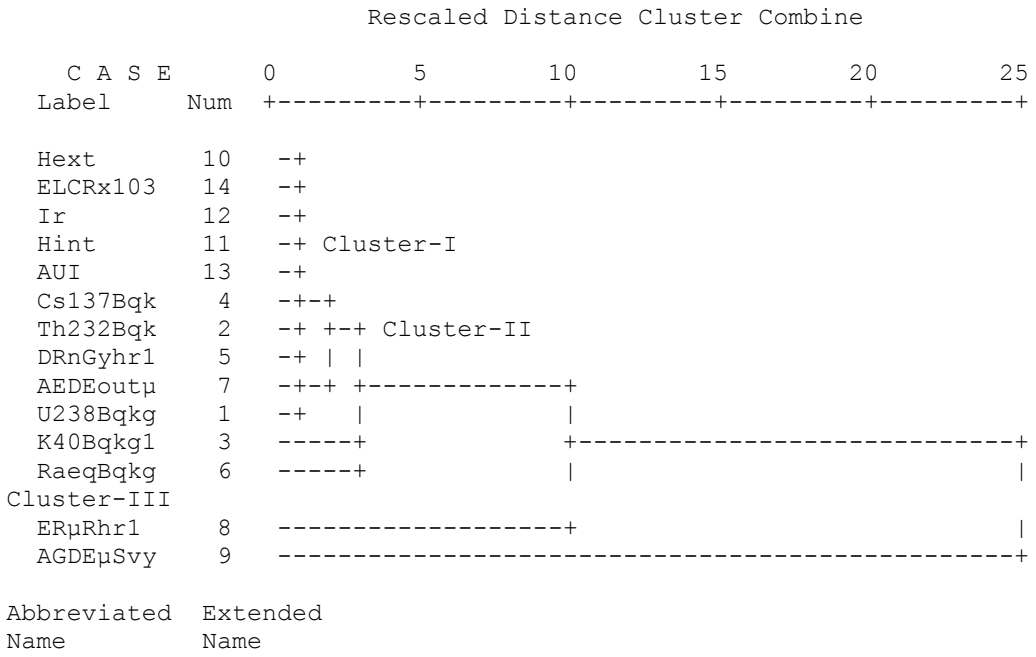

Fig. 5: Dendrogram shows cluster formation of radionuclides and their radiological parameters of tar-sand soil samples

5. Spatial Distributions

The investigation of ^{238}U:^{232}Th activity concentrations in the dumpsite soils revealed that ^{238}U activity concentrations are seen to be of 6 times higher than the ^{232}Th activity concentration in the measured soil samples from AATS1 (Ibijola dumpsite top soils). The ratio $^{238}U/^{232}Th$

ratios (Table 7) were higher than the world's average value of 1 in the two top soil samples from Ibijola waste dump sites. The activity concentration of ^{40}K shows that on the average ^{40}K are about 2 and 12 times times higher than ^{238}U and ^{232}Th activity concentration in the collected soil samples. This ratio ^{232}Th: ^{238}U, ^{238}U: ^{232}Th, ^{232}Th: ^{40}K and ^{238}U: ^{40}K gives an indication that the samples from a certain region or part have higher uranium and potassium than thorium concentrations to be economic for the ^{238}U and ^{40}K extraction. On the average, activity concentration of ^{40}K and ^{238}U is high in all the samples which may be due to (i) the presence of loamy and clay soils couple with extensive use of potassium rich fertilizer like phosphate in the soils. The $^{232}Th/^{238}U$ ratio was also calculated (Table 7), from these results, the ratio were higher than the stipulated range 0.7-0.4 mentioned by (Mitchell, Mihalynuk & Heaman, 2002; ICRP, 1976) for igneous zircon sources in all the soil samples. All the soil samples of the study area have $^{232}Th/^{238}U$ ratio above this range. This could be indicated that the present study confirmed the highest presence of xenolith zircon in the soil samples.

Table 7: Activity ratios

S/N	Sample ID	$^{238}U/^{232}Th$	$^{232}Th/^{238}U$	$^{232}Th/^{40}K$	$^{238}U/^{40}K$
1.	AATS1	6.58	0.15	0.08	0.52
2.	AATS2	5.45	0.18	0.10	0.57
3.	ABTS1	0.58	1.71	0.79	0.46
4.	ABTS2	0.72	1.40	0.46	0.33
5.	AASS1	0.46	2.15	0.35	0.16
6.	AASS2	0.64	1.57	0.31	0.20
7.	AASS3	0.50	1.99	0.38	0.19
8.	ABSS1	0.72	1.39	0.55	0.39
9.	ABSS2	0.56	1.78	0.55	0.31
10.	ABSS3	0.52	1.93	0.58	0.30
11.	CONTROL	0.72	1.39	0.90	0.65

4.3 Determination of Annual Doses & Probability of Excess Cancer risk lifetime using Residual Radiation Software (RESRAD)

Residual Radioactivity software (RESRAD, version 6.5) developed by the environmental assessment of the Argonne National laboratory was used to assess and predict the total effective dose equivalent (TEDE) and probability of excess lifetime cancer risk incurred by workers (Fouzey *et al.,* 2013), scavengers and people living around the dumpsites who are exposed to the deleterious waste enhanced naturally occurring radioactive materials (WENORM) ^{238}U, ^{232}Th and ^{40}K emanated from the selected dumpsites. The derivation for the dumpsite clean-up or remediation criteria depends on the sum of the fraction rule from multiple contamination from ^{238}U, ^{232}Th and ^{40}K radionuclides (Equation 3), This sum of rule fraction from the multiple contamination must be less than unity and if the calculated value from (Equation 3) is greater than 1, there is need for soil remediation in order to reduce the cancer risk. From this scenario, the potential exposure route examined include (i) direct exposure to external γ-radiation from the contaminated materials in the dumpsites (ii) internal radiation from inhalation of radionuclide (Radon) polluted dust by the workers, scavengers and people leaving around the dumpsites and (iii) the internal γ-radiation from ingestion of contaminated soil. While water and food materials is assumed to be from other sources and not been produced from the dumpsites. The simulation was performed for up to 70 years and that workers, scavengers and people around the selected dumpsites have 3 hours daily exposure to these substances. The probability of cancer incidence risk is shown in Fig. 5 respectively. Finding from this figures show that there is high probability that the worker will have cancer if they are exposed to these radionuclides for 70 years at 3 hours exposure per day in Ibijola dumpsite soils than in Idowale Dumpsites (Fig. 6)

Sum of the fraction rule equation (SOFRE) (Fouzey *et al.,* 2013)

$$\text{SOFRE} = \frac{IR_U}{SR_{(U)tmax}} + \frac{IR_{Th}}{SR_{(Th)tmax}} + \frac{IR_K}{SR_{(K)tmax}} \leq 1 \quad (3)$$

Where IR_U, IR_{Th} and IR_K are Initial radionuclide concentration ^{238}U, ^{232}Th and ^{40}K in the soil in (pCi/g) and $SR_{(U)t}$, $SR_{(Th)t}$ and $SR_{(K)t}$ are the concentration of the single radioactivity ^{238}U, ^{232}Th and ^{40}K (pCi/g) after a particular time t-maximum (70 years in this scenario). This Sum of fraction rule equation value must be < 1 for the soil not to be considered for remediation processes. The calculated SOFRE value for Ibijola and Idowale dumpsite soils are 0.611 and 0.536 respectively. Ibijola dumpsite has higher SOFRE value than Idowale

dumpsite. Indication of this is that Idowale dumpsite soils have greater tendency of causing harms to the inhabitants and it will be due for remediation process in the nearest future. Though both locations SOFRE value falls within the stipulated value by nuclear regulatory commission (NRC). The average cancer risk or probabilities for all the study sites (Ibijola and Idowale dumpsites) are shown in Fig. 6 respectively. This figures shows that there 3.2 x 10^{-4}, and 2.7 x 10^{-4} probability that the inhabitants of the dumpsites (Ibijola and Idowale) will have cancer. Therefore, there is higher cancer probability in Ibijola dumpsite soils than Idowale dumpsites soils respectively. The total effective dose equivalent (TEDE) received by the people or animal living or working around the dumpsites were calculated using equation (4).

$$TEDE = SOFRE * 0.25\ mSvyr^{-1} \quad (4)$$

Where 0.25 $mSvyr^{-1}$ is the basic dose limit adopt by nuclear regulatory commission (NRC) criteria and 0.3 $mSvyr^{-1}$ by (IAEA criteria). The calculated TEDE values for Ibijola and Idowale dumpsites were 0.51 $mSvyr^{-1}$ and 0.134 $mSvyr^{-1}$ respectively. This revealed that the TEDE value from the two selected dumpsites soils in Agbara is below the regulatory dose limit of 0.25 $mSvyr^{-1}$ set by NRC and 0.3 $mSvyr^{-1}$ set by IAEA.

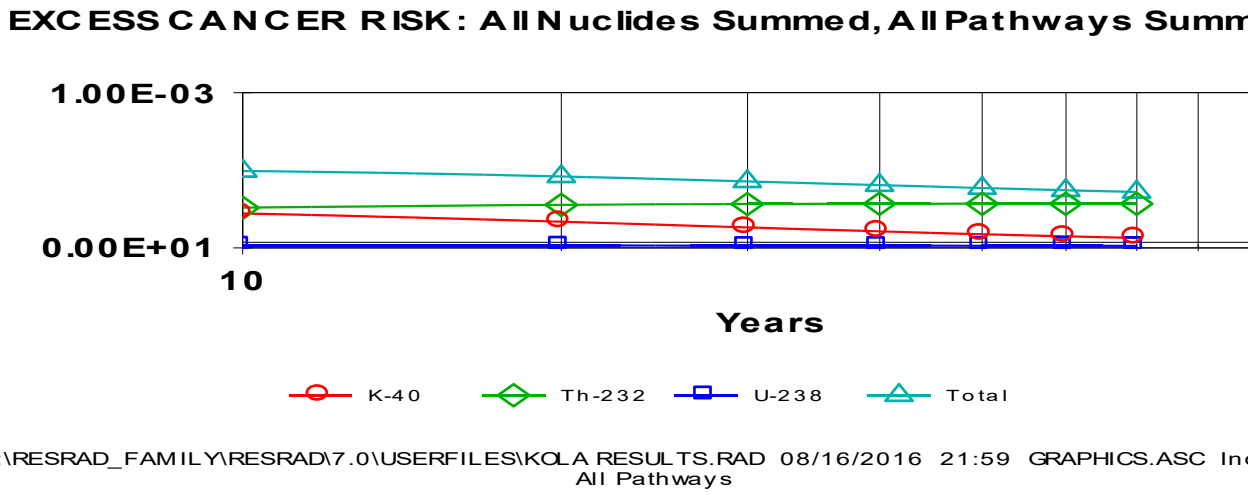

Fig. 6: Showing the probability of excessive cancer lifetime incurred by the people living around Ibijola dumpsite.

Conclusion

The activity concentration of the radionuclides and its associated radiological parameters from the selected dumpsites in Agbara were estimated. The following conclusions were reached.

1. The mean activity concentrations of the naturally and artificial occurring radionuclides ^{238}U, ^{232}Th, ^{40}K and ^{137}Cs in all the soil samples are of this order ^{40}K ^{238}U ^{232}Th ^{137}Cs. The total and average activity concentrations of the radionuclides (^{238}U and ^{40}K) were higher than the reported worldwide average values. Radiological parameters like absorbed dose rate, I_r, H_{int}, $AEDE_{outdoor}$ and $ELCR_{outdoor}$ were higher than the recommended level while others radiological parameters (AGDE, AUI, H_{ext}, ER, and Ra_{eq}) falls below the worldwide average value. This implies that the inhabitants of the study area are exposed to radiation exposure which was significantly higher than the corresponding exposure levels reported in some related studies. Statistically, there is a strong positive significant relationship between ^{40}K and ^{238}U series with all the calculated radiological parameters.
2. The soils of Ibijola and Idowale dumpsites pose little radiological threat to the people working/living around the dumpsites in which the cumulative effects can be detrimental to human health if the exposure is for a longer period of time. This finding was also corroborated with the Residual Radioactivity (RESRAD software, version 6.5) which revealed a high probability of cancer risk incidence on exposure to dumpsites soil through inhalation, ingestion or dermal contact. The total effective dose equivalent (TEDE) value were lower than the regulatory dose limit of 0.25 $mSvyr^{-1}$ and 0.3 $mSvyr^{-1}$ set by NRC and IAEA respectively.

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