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ASSESSMENT OF RADIONUCLIDES IN THE SOIL OF BONNY LOCAL GOVERNMENT AREA OF RIVERS STATE, NIGERIA AND EVALUATION OF RADIOLOGICAL RISK.

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ABSTRACT

Twenty Samples each of soil from Bonny LGA, Rivers State, Nigeria were analyzed using NaI(IT) gamma detector to estimate radiation hazard due to the anthropogenic sources. The activity concentration of ^{232}Th was found to be in the range 18.78 – 397.13 $BqKg^{-1}$, ^{40}K in the range 43.72 – 390.62 $BqKg^{-1}$ and ^{226}Ra in the range 10.77 – 57.84 $BqKg^{-1}$ all in soil. These results were used to calculate the radiological hazard parameters including the Annual Gonadal Equivalent Dose. The calculated gamma exposure rates ranged between 10.00 – 270.79 $nGyh^{-1}$ while the average value of the Excess Lifetime Cancer Risk (ELCR) was found to be 0.38 x 10^{-3} for soil which is higher than the world average of 0.29 x 10^{-3}.

Keywords: Gamma Spectrometer, NaI(IT) detector, Radionuclides, Excess Lifetime Cancer Risk, Annual Gonadal Equivalent Dose.

INTRODUCTION

Radiation is the transmission of energy in the form of heat, light, electrons etc. Nuclear radiation all over the world has become a great concern not ruling out the fact that it is part of our natural environment. The most common radionuclides in soil are ^{40}K and the three natural radioactive decay series ^{238}U, ^{226}Ra and ^{232}Th. These natural radionuclides contribute to the radiation exposure both internally and externally as a result of inhalation and gamma emission respectively [International Atomic Energy Agency (IAEA)., 1989]. Virtually everything on earth is somehow exposed to radiation [Avwiri,G.O. *et al*., 2010] but can be harmful when the degree of exposure is at an exceedingly high level. Thus, radionuclides could be enhanced when anthropogenic activities (artificial radionuclides) are on the increase. This artificial radionuclide enters the environment largely as a result of these activities e.g. oil and gas recovery processes etc and then spreads out into distant locations through atmospheric convection [Chowdhury, M.I. *et al*., 1999].

Researchers on the effect of radiation have shown that radiation exposure could lead to lung, pancreas, hepatic, bone, skin and kidney cancers, cataracts, sterility, atrophy of the Kidney and leukemia [Taskin, H. *et al*., 2009; Amiri, M. *et al*., 2011]. Quantities such as the absorbed dose and annual effective dose from the activity concentration of the radionuclides have been introduced to specify the dose

received and the total dose the tissue is exposed to.

The coastal areas are dominated with the oil production and exploration industries by making it the highest importer and consumer of radioactive materials [Oni, O.M. *et al.*, 2011]. During exploration and exploration processes, various operational practices contribute to an increased NORM concentration namely drilling equipment and activities, down-the-hole geophysical logging methods etc [Avwiri, G.O. and Ononugbo, C.P. 2012].

This paper therefore seeks to measure the radiation exposure rates around Bonny Local Government Area (LGA) of Rivers State. Bonny LGA of Rivers State, Nigeria is located in the South East Senatorial District in Rivers State political zone. The Island lies on the longitude E7°10' and latitude N4°27 with an estimated population of 214,983 [National Population Commission Report, 2006]. It plays host to multinational oil and gas companies such as Shell Petroleum Development Company (SPDC) export terminal, Chevron Nigeria, NLNG etc. The Island has a relatively flat topography on an elevation of 3.05 atmospheric mean sea level with a total land area of 214.52 m^2 [Nigerian Liquefied Natural Gas Report, 2005].

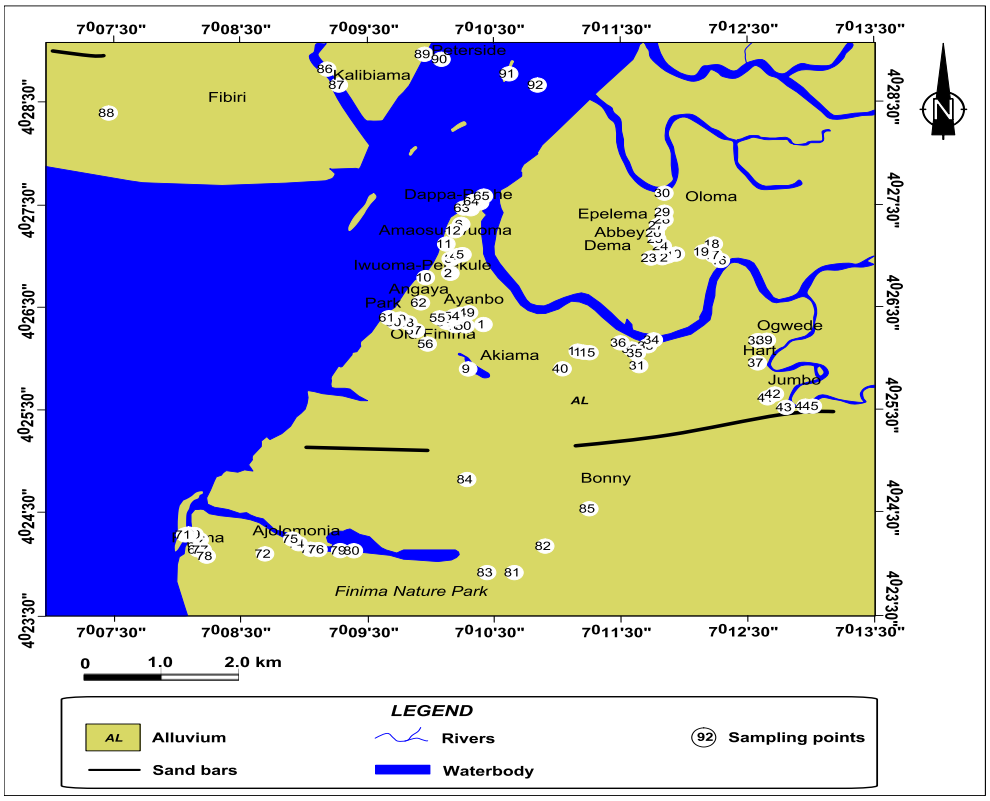

Fig. 1: Map of Bonny Island showing sampling points

MATERIALS AND METHODS

Samples Collection and Preparation

Twenty (20) soil samples were collected from the study area. Each was subjected to spectrometric analysis using Sodium Iodide (NaI(TI))Spectrometer. The samples were prepared for gamma analysis by drying overnight in the electric oven at 115^{0}C. The samples were mechanically crushed and sieved through a 0.8mm mesh sieve. The sieved portion of the sample was transferred into a 100ml Marinelli beaker for gamma spectrometry and sealed for four weeks to attain secular equilibrium between the radium contents of the samples and their decay

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product [Diab, H.M. *et al*., 2008] before analysis using the NaI(TI) gamma spectrometer. The NaI(TI) is a 2″×2″ Sodium iodide detector coupled to an ORTEC 456 digiBase multichannel analyzer (MCA). The digiBase is connected to a computer with a USB cable. Accumulation and analysis of the gamma-ray spectrum were carried out with the computer using the ORTECMestro Software. The counting (accumulation) time was 10800 seconds. The detector was installed in a 15cm thick cylindrical lead shield to reduce the influence of background radiation.The standard materials from the International Atomic Energy Agency (IAEA) were used for calibration. From the counting spectra, the activity concentrations of radium ^{226}Ra, thorium ^{232}Th and potassium ^{40}K were determined using the software.

Activity Concentrations and Radiological Hazard Parameter Calculation

An essential requirement for the measurement of gamma emitters is the exact identity of photo peaks present in a spectrum produced by a detector system. The energy calibration of detector system is made by measuring mixed standard sources of known radionuclide with well- defined energies provided by the IAEA.

The specific activity concentrations of the samples were determined using the net area under the photo peaks from the energy and efficiency calibration.

$$C\ (BqKg^{-1}) = K\ C_n \qquad (1)$$

where C ($BqKg^{-1}$) is the specific activity concentration of the radionuclides in the sample, C_n is thecount rate under the corresponding peak, $K = 1/\varepsilon P_\gamma M_S$, ε is the detector efficiency at the specific gamma ray energy, P_γ is the absolute transition probability of the specific gamma ray and M_S is the mass of the sample [Jibiri, N. N. and Okeyode, I.C. 2012].

The peak corresponds to 1460 KeV (^{40}K) for 40K, 1764.5 KeV (Bi-214) for ^{226}Ra, and 2614.5 KeV (Ti-208) for ^{232}Th was considered in arriving at the activity levels ($Bqkg^{-1}$). The activity concentration (C) of the radionuclide was calculated after subtracting decay correction using the following expression [Jibiri, N. N. and Okeyode, I.C. 2012; Belyaeva, O. *et al*., 2019].

$$C_S = \frac{C_a}{P_\gamma (M_s/V_s)\varepsilon_\gamma t_c}\left(BqKg^{-1}\right) \qquad (2)$$

where Cs = Sample concentration, Ca = net peak area of a peak at energy, ε_γ= Efficiency of the detector for a γ-energy of interest, Ms/Vs = Sample mass/volume for soil/sediment, tc = total counting time, Pγ is the abundance of the γ-line in a radionuclide.

Natural radionuclides ^{226}Ra, ^{232}Th, and ^{40}K in soil are observed to vary from place to place. So, the assessment of radiation hazards associated with these radionuclides in soil was calculated by the following formula.

Absorbed Dose (D)

Is a measure of the energy deposited in a medium by ionizing radiation. It is equal to the energy deposited per unit mass of medium, and so has the unit J/kg or gray (Gy) where $1Gy = 1Jkg^{-1}$. The absorbed dose rates (D) due to gamma radiations in the air at 1m above the ground surface for the uniform distribution of the naturally occurring radionuclides (^{238}U, ^{232}Th and ^{40}K) will be calculated using equation [Chowdhury, M.I. *et al*., 1999].

$$D\ (nGyh^{-1}) = 0.462A_{Ra} + 0.621A_{Th} + 0.0417A_K \qquad (3)$$

Where A_U, A_{Th} and A_K are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K respectively [UNSCEAR, 2000].

Annual Effective Dose Equivalent (AEDE)

The annual effective dose equivalent received outdoors by the inhabitants of an area is evaluated from the absorbed dose rate (D) using the conversion factor of 0.7Sv/Gy and the outdoor occupancy factor of 0.2. The annual effective dose equivalent ($mSvyr^{-1}$)

was calculated using the formula below Ramasamy, V. *et a*l., 2009; Etuk, S.E. *et al*., 2017].

$$AEDE\ (mSvyr^{-1}) = D(nGyrh^{-1}) \times 8760\ h \times 0.7Sv/Gy \times 0.2 \quad (4)$$

where D is the absorbed dose rate, 0.7Sv/Gy as the conversion coefficient from absorbed dose in air to effective dose [UNSCEAR, 2000] and 0.2 as the value for the outdoor occupancy factor [Harb S. *et al*., 2010].

Excess LifetimeCancer Risk (ELCR)

Excess Lifetime Cancer Risk is the probability of developing cancer over a lifetime at a given radiation exposure level. It is presented as a value representing the number of extra cancers expected in a given number of people on exposure to a carcinogen at a given dose. It is calculated using the relation:

$$\text{Excess lifetime cancer risk (ELCR)} = AEDE \times DL \times RF \quad (5)$$

Where AEDE is the Annual Effective Dose, DL is the average Duration of Life (estimated to be 70years) and RF is the Risk Factor (Svy^{-1}), i.e. fatal cancer risk per Sievert. For stochastic effects, ICRP uses RF as $0.05Sv^{-1}$ forpublic exposure [[Taskin, H. *et al*., 2009; Amiri M. *et al*., 2011; AgbalagbaO.E. 2017; (Khandaker *et al*., 2018).].

Annual Gonadal Equivalent Dose (AGED)

According to UNSCEAR (1982), the gonads (an organ in which eggs or sperm are produced especially, the ovary and testis), the active bone marrow andthe bone surface cells are considered the organs of interest. An increase in Annual Gonadal Equivalent Dose (AGED) has been known to affect the bone marrow, destroying the red blood cells that are then replaced bywhite bloodcells. This situation results in a blood cancer called leukemia which is fatal [Amiri, M. *et al*., 2011]. Therefore, the AGED ($\mu Sv\ y^{-1}$) for the residents of the study area due to the specific activities of ^{226}Ra, ^{232}Th and ^{40}K was calculated using equation 6 given by as: [Arafa, W. 2004]

$$AGED\ (\mu Svy^{-1}) = 3.09\ A_{Ra} + 4.18\ A_{Th} + 0.314\ A_K \quad (6)$$

Where A_{Ra}, A_{Th} and A_K are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K, respectively.

RESULTS AND DISCUSSION

Table 1: Activity Concentration in Soil Samples

S/N	LOCATION	LOCATION CODE	ACTITIVY CONCENTRATION ($BqKg^{-1}$) ^{40}K	^{226}Ra	^{232}Th
1	Ajolomonia	SAAJ 1	90.86	57.84	15.86
2	Iwuoma	SAOG 1	385.78	17.5	397.13
3	Kalabiama	SAK 1	78	35.27	54.35
4	Park Community 2	SAPC 2	185.14	41.99	18.78
5	Fibiri	SAF 2	94.49	13.18	bdl
6	Dappa-Poshe	SAAD-P	270.96	40.55	28.02
7	Light House	SALH 1	171.84	41.03	83.61
8	Main Bonny Town	SAMB 1	215.36	33.83	bdl
9	Ayanbo 2	SAAY 2	348.31	29.03	295.72
10	Park Community 1	SAPC 1	101.74	21.82	374.7
11	Akiama	SAAK 1	243.16	33.35	271.34
12	Minima	SAM 1	274.58	36.23	bdl
13	Ayanbo 1	SAAY 1	43.72	30.95	27.56
14	Epelema	SAE 1	252.83	25.18	174.8
15	Oloma	SAO	173.05	20.86	51.94
16	Agaya	SAAG	229.86	55.44	75.34
17	New Finima	SAFM 2	306.01	16.06	214.78
18	Hart/LongJohn	SAL 1	243.16	32.39	339.6
19	Peterside	SAP 1	390.62	37.67	250.86
20	Abalamabie	SAAB 1	285.46	10.77	54.35

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Mean	**234.64±10.34**	**31.55±1.75**	**160.51±13.83**
UNSCEAR (2000)	**400**	**35**	**30**

bdl = below detectable level

Table 2: Radiation Hazard Parameters in Soil

Location	D ($nGyh^{-1}$)	AEDE ($mSvy^{-1}$)	Gonadal ($mSvy^{-1}$)	ELCR x 10^{-3}
SAAJ 1	40.36	0.05	2369.51	0.12
SAOG 1	270.79	0.33	1154.01	0.83
SAK 1	66.13	0.08	314.61	0.20
SAPC 2	38.78	0.05	135.65	0.12
SAF 2	10.03	0.01	40.73	0.03
SAAD-P	47.43	0.06	134.10	0.15
SALH 1	78.04	0.10	321.24	0.24
SAMB 1	24.61	0.03	358.30	0.08
SAAY 2	211.58	0.26	92.86	0.65
SAPC 1	247.01	0.30	940.20	0.76
SAAK 1	194.05	0.24	356.46	0.59
SAM 1	28.19	0.03	111.95	0.09
SAAY 1	33.24	0.04	142.03	0.10
SAE 1	130.73	0.16	1719.34	0.40
SAO	49.11	0.06	80.77	0.15
SAAG	81.98	0.10	194.97	0.25
SAFM 2	153.56	0.19	117.07	0.47
SAL 1	236.00	0.29	1084.14	0.72
SAP 1	189.48	0.23	550.85	0.58
SAAB 1	50.63	0.06	387.59	0.16
Mean	**109.09**	**0.13**	**530.32**	**0.33**
UNSCEAR (2000)	**57**	**1**	**300**	**0.29**

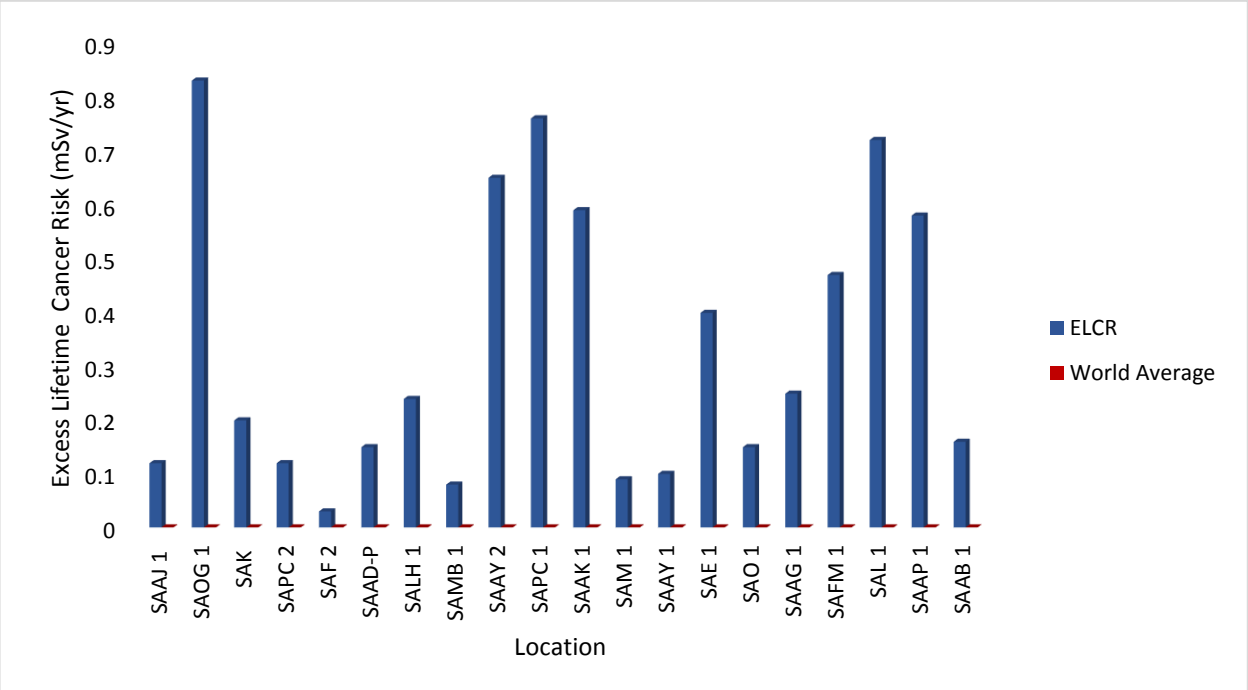

Fig. 2: Comparison of Excess Lifetime Cancer Risk ($mSvy^{-1}$) in Soil with world average in all the locations.

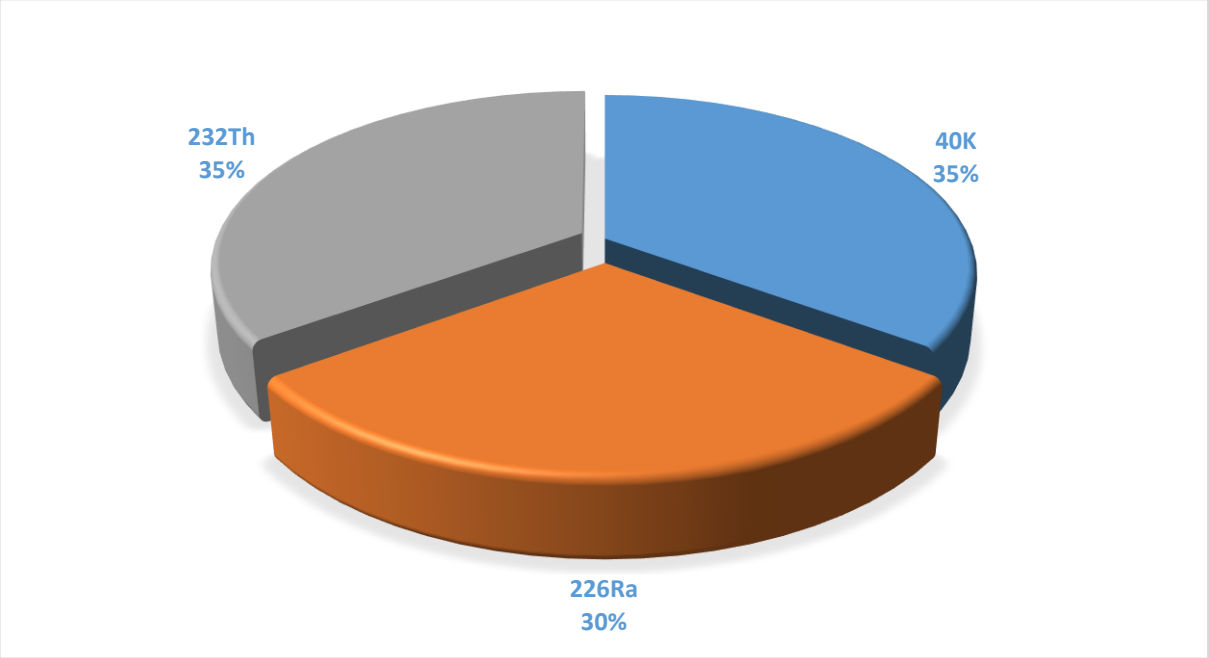

Fig. 3: Percentage contribution of ^{226}Ra, ^{232}Th and ^{40}K activity in Soil

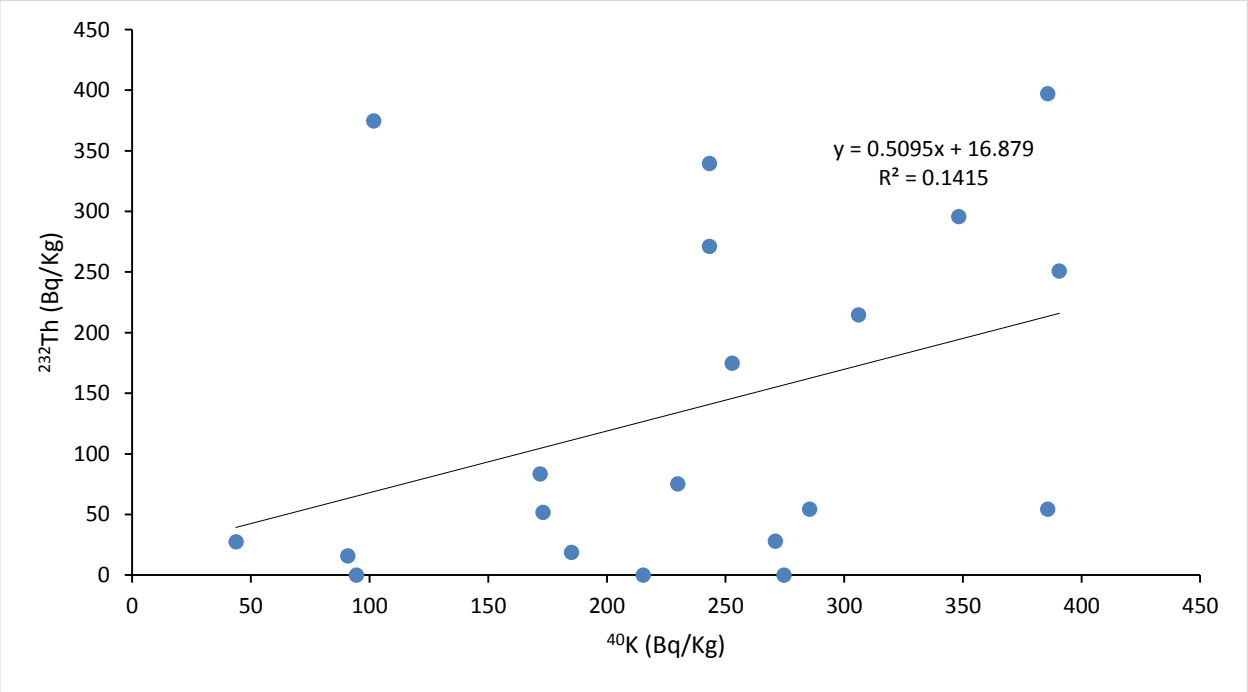

Fig. 4: Correlation of ^{232}Th with ^{40}K in soil samples

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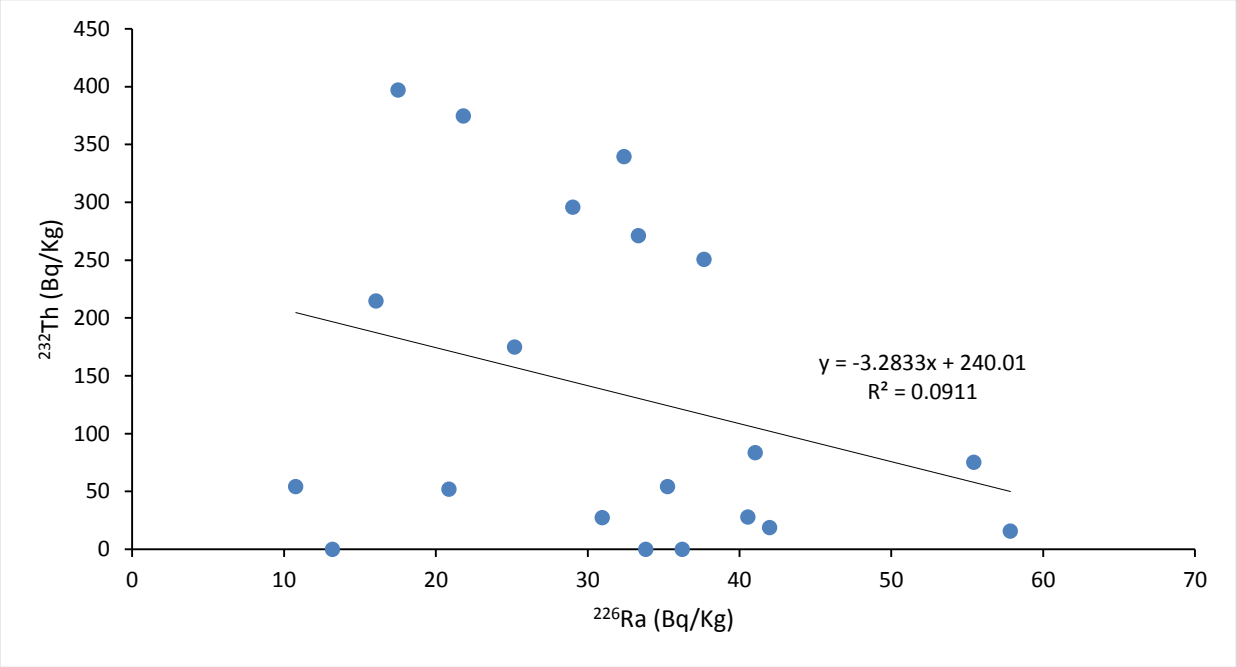

Fig. 5: Correlation of ^{232}Th with ^{226}Ra in soil samples

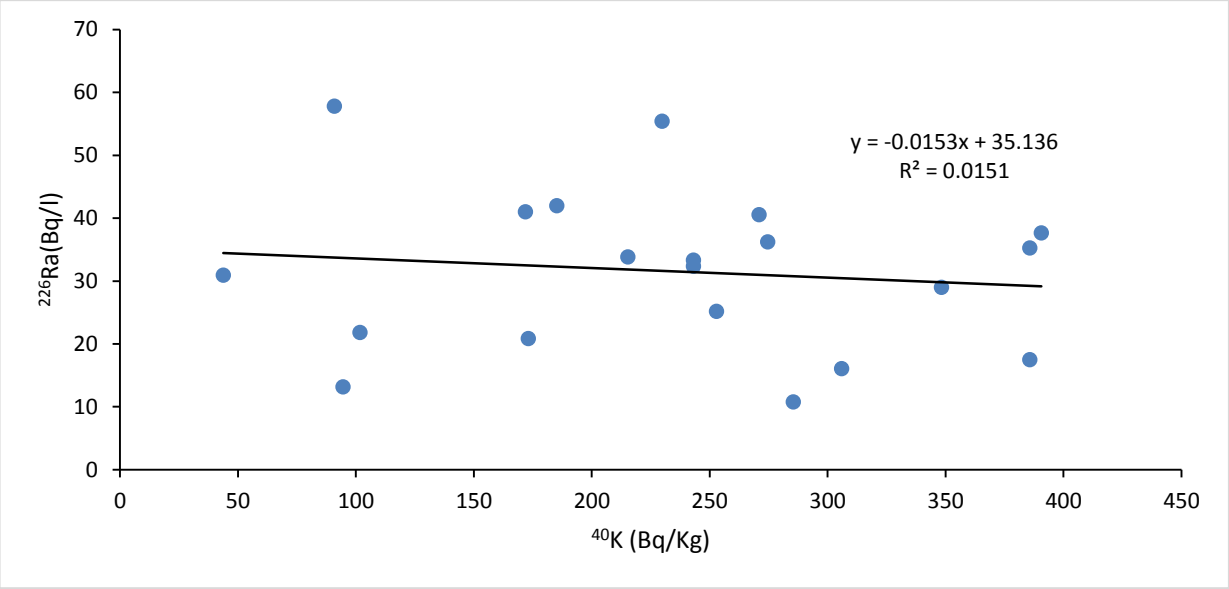

Fig. 6: Correlation of ^{226}Ra with ^{40}K in soil samples

Three naturally occurring radionuclides^{40}K, ^{226}Ra and ^{232}Th were determined in all the soil as shown in Table 1. The mean activity concentration of ^{40}K, ^{226}Ra and ^{232}Th in soil from the studied area are 234.64 ± 10.34 Bqkg^{-1}, 34.55 ±1.75 Bqkg^{-1} and 100.51 ± 13.83 Bqkg^{-1}respectively. The largest contribution of the overall activity concentrations in soil comes mainly from ^{40}K.

The obtained mean values of ^{40}K, ^{226}Ra and ^{232}Th, when compared with [UNSCEAR, 2000], exceeded the standard value limit of thorium (30 Bqkg^{-1}) and were lower than radium (35 Bqkg^{-1}) and potassium (400 Bqkg^{-1}) as shown in Table 1. From the obtained

results, the mean values exceeded the reported results of Avwiri*et al.,* (2014) and Benjamin *et al.,* (2023). The high concentrations of the obtained results can be attributed to the geological formation of the area and also to drilling chemicals (drilling mud), well logging,etc used during oil and gas exploration, exploitation and production by the companies operating.

Radiological Hazard Parameters

The absorbed dose rate was calculated using Equation 3 and obtained results are shown in Table 2. The soil samples have their mean result as 109.09 $mSvy^{-1}$The obtained result when compared with the world average value of 1.5 $mSvy^{-1}$ [UNSCEAR, 2000] revealed that the mean value of the absorbed dose rate is higher than world average value. This can be attributed to drilling chemicals, well logging equipment etc used during oil and gas activities and the maritime activities in the area.

Table 2 and Figure 2 also show the mean result of the calculated excess lifetime cancer risk which is 0.88×10^{-3} $mSvy^{-1}$ for soil. When compared with the permissible allowed world average (0.29×10^{-3}) [Amiri M. *et al*., 2011, Ramasamy, V. *et al*., 2009] as shown in Figure 2 it is observed that the obtained value of soil is higher than the world allowable average. This could be attributed to the high activity concentrations of ^{40}K, ^{226}Ra and ^{232}Th that are present in the measured soil samples. These high concentrations pose a significant threat to both the human system and the environment.

Table 2 shows the result of the calculated annual gonadal equivalent dose. The values ranged from 40.73 to 2369.51 $mSvy^{-1}$ with a mean value of 530.32 $mSvy^{-1}$. When compared with thepermissible allowed world average of 300 $mSvyr^{-1}$ [Xinwei, L. *et al*., 2006], it is observed that the obtained mean value is higher than the world allowable average. This could be attributed to the high activity concentrations of ^{40}K, ^{226}Ra and ^{232}Th that are present in the measured sample. These high concentrations pose a significant threat to both the human system and the environment. This is attributed to the oil and gas exploitation activities.

Graphically the percentage contribution of each of these radionuclides is represented by a pie chart in Fig. 3.

The correlation coefficient among measured ^{226}Ra, ^{232}Th and ^{40}K activity concentrations in soil (Figures 4 to 6) were shown for the studied area. It can be observed that a weak positive correlation exists among the three radionuclides ^{226}Ra, ^{232}Th and ^{40}K. ^{226}Ra and ^{40}K has a coefficient value r = 0.0151, ^{226}Ra and ^{232}Th has correlation coefficient r = 0.0911 and ^{40}K and ^{232}Th has correlation coefficient r = 0.1415. This weak relationship among the three radionuclides shows that they may have the same origin but their behavior in the soil environment differs from one another.

CONCLUSION

Radionuclides from the twenty samples collected in Bonny were measured using a Sodium Iodide (NaI(TI)) spectrometer. The Gamma analysis result shows that the radionuclide concentration measured in soil for ^{232}Th exceeds the world average but below the world average for radium and potassium. The calculated hazard indices and their associated potential radiological health risks in soil samples are higher than their world average permissible limit. This pose danger to the community.

Competing Interests

Authors have declared that no competing interests exist.

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