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Inventory of Occupational Risk from Natural Occurring Radioactive Materials (NORMs) in Soil Samples of the Coastal Region of Nigeria After Six Decades of Oil and Gas Exploration

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ABSTRACT

Background and Objective: The occupational risks associated with exposure from radioactivity concentration of soil from oil and gases producing coastal areas of Nigeria have been assayed after sixty years of exploration activities in the area, to ascertain the radiological impact on the oil field workers and host communities. This study observes the occupational risk from natural occurring radioactive materials in soil samples. **Materials and Methods:** Gamma spectrometric technique was used to determine the radiometric in the soil samples collected using highly efficient thallium activated 3"×3" gamma spectrometer. **Results:** The mean activity concentration of the three natural radionuclides of ^{40}K, ^{238}U and ^{232}Th are 1586.82±17.97, 27.26±8.38 and 37.67±9.55 Bq kg^{-1}, respectively. The occupational risk estimates from three exposure routes, external, inhalation and ingestion were calculated using the specific activities of the radio nuclides detected in the soil samples. Radioactivity concentrations measured in the communities were found to be higher than their corresponding standard values. The estimated total effective dose received by the residents and oil field workers through the three exposure routes ranged from 0.2-14.8 mSv y^{-1} with a mean value of 4.77 mSv y^{-1}. The values of each of the total effective doses estimated in the different communities exceeded the 1.0 mSv y^{-1} permissible limit. The dose equivalent estimate obtained for external exposure is 4.27 µSv y^{-1} which represents 89.0% of the permissible limit. **Conclusion:** These results obtained show that both residents and oil field workers are significantly exposed to radiation from the soil which may be of radiological health concern.

KEYWORDS

Occupational risk, effective dose, radionuclide pathway, oil exploration

INTRODUCTION

Consequent to the discovery of crude oil and gas in the Niger Delta Region of Nigeria in 1956, the exploration and exploitation of these natural resources have been a continuous one unabated. Not the civil war between 1966 and 1970 could hinder the operations of the multinationals involved in this oil and gas exploration. The human and environmental impact of the exploration activities of these natural

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resources has been a subject of debate in the national discourse. However, research findings in recent times have revealed that the rural dwellers and oil field workers in the region are continually exposed to radiation hazards from gamma radiation and internal exposure resulting from the inhalation of radon gas from gas flairs, radiation from industrial radiography, automated ionizing radiation gauge, well logging, use of radiotracers in pipes, mapping and evaluation of geological formation[1-4]. Ironically, most of these studies are restricted to the onshore area of the Niger Delta Region leaving the swamp/coastal areas unreached due to lack of ease of access occasioned by the difficult nature of the terrain. This research work is set out to reach these hitherto unassessed areas of the coastal communities for proper evaluation of the occupational risk associated with oil and gas activities on the soil radioactivity, after sixty years of oil and gas exploration activities in the region.

Natural radioactive elements are found in the natural mineral and mineral-laden materials deposits in suitable geological formations and they manifest in outcrops thereby elevating the background ionizing radiation(terrestrial and extra-terrestrial sources)[5,6]. Thus, Naturally Occurring Radioactive Material (NORM) and the associated external exposure due to gamma radiation depended largely on the geological formation while artificial sources are due to medical and industrial activities[7-10]. Research has revealed that man and its environment are exposed to ionizing radiations emission knowingly or unknowingly. And when the man is exposed to radiation at a high level, it can lead to a wide range of health challenges which includes cancer of the lung, bone and skin, kidney diseases and blood infections. Chronicle effects include alteration in the human genetic make-up, deterministic effect, stochastic effects, irritations, sensitization, embryonic effects, etc.[11,12]. To evaluate man's safety to the exposure to radiation, there is a need to understand the distribution of radio nuclide's in the soil is one of the chief lives supports of man and other living things in the environment[13].

The Coastal Region of Nigeria where oil and gas exploration started in 1957, is characterized by the sedimentary rock of the Niger Delta Basin which is mainly Cenozoic in age[14]. Most sedimentary mineral deposits such as limestone, shale and hydrocarbon are associated with radionuclides like uranium, thorium and their progenies[15,16]. The drilling of oil and gas in the region is through the use of a beam pump, which utilizes the pressure of the gas in the reservoir, pressuring the oil up and into the well[16]. A similar process of crude oil extraction that is being proposed and may soon be deployed for oil drilling includes water and steam injections, shale gas extraction like fracking[16]. All these crude oil and gas exploration techniques are the probable cause of the increase in radioactive materials at the surface, as part of the flow back. These in most times find their way into the oil field soil through oil spills and gas flares. Besides these, operating companies deploy some radioactive materials during good drilling and work over (mud, well logging gauges), which may also add to the rise in the activities of the radionuclides beyond the normal background level[9.17]. Accidental discharges, equipment failures and pipelines vandalization may cause oil spilled, sludge, produced water, effluents, leachates and scales to pollutethe coastal environment[1]. These pollutants are transported into the soil compartment of the coastal ecosystem gravitationally due to seepage, which can also increase the specific activities of the soil in the environment[18]. The Technologically Enhanced Naturally Occurring Radioactive Materials (TENORM) in the soil in the oil and gas industry has the potential to cause both internal and external exposures during production due to the accumulation of gamma emitted radiation exposure hazard to both workers and members of the public through the inhalation and ingestion of radionuclides[9,19,20].

The assessment of the levels of environmental and human exposure to radiations from soil and other naturally occurring radioactive materials have been reported in previous studies[21-25]. Literature abound also on research works that have been undertaken to precisely quantify the number of radioactivity levels in different soil and solid minerals found in Nigeria in recent time[1,3,4,26,27] and in some countries of the world for radiation protection[28-36]. In Northern and Western Nigeria, a sizeable number of research works have also been conducted in this regard[37-39]. It is worth mentioning that investigation on the level of dosage and excess level of radiation in the risk of cancer in this area has been reported in literatures[40,41]. It, however, appears that not much work has been done on occupational risk estimation from the soil, especially in these study areas, as kinds of literature on the occupational risk assessment are lacking. This work, therefore aimed at estimating the occupational risk due to radionuclides alongside other risk parameters in the soil from these oil and gas producing coastal environments of the Niger Delta Region of Nigeria after six decades of oil and gas exploration activities.

MATERIALS AND METHODS

Description of study area: The study area is located between latitudes 5°18' and 5°30' North and longitudes 5°54 and 5°45' East of the Niger Delta Region of Nigeria with an elevation of 3.0 m above sea level. It bounded to the south by the Atlantic Ocean. It is one of the highest producers of oil and gas in the Niger Delta Region of the country.

Sample collection and preparation: In the study areas, sixty soil samples (four from each community) were collected at different sampling points from fifteen communities using a standard method[1,40-42]. About 3 kg of soil samples were collected and stored in a sealed and labelled black polythene container to avoid cross-contamination before and during transport to the laboratory. They were sprayed on stainless steel trays after which they were separately crushed and sieved through a 2 mm mesh sieve. The sieved smooth samples were weighed into special sampling containers which were then tightly sealed and stored in the laboratory for not less than four weeks to reach secular equilibrium.

Sample analysis: The activity concentrations of each soil sample were determined using gamma spectrometry at the National Institute of Radiation Protection and Research Centre (NIRPR), University of Ibadan, using a Canberra HpGe detector, with a relative efficiency of 29% and an energy resolution of 1.8 keV for ^{60}Co γ-ray energy line at 1332 keV.

The detector was enclosed in a graded lead shield (Model 747, USA) and connected to DSA-1000 (Canberra, USA) for data acquisition and the spectrum was analyzed by GENIE- 2000 software. The energy calibration was performed using the standard reference radionuclide sources: ^{60}Co, ^{137}Cs and ^{154}Eu, while the efficiency calibration was performed using the reference soil (^{238}U, ^{226}Ra, ^{228}Ra, ^{232}Th and ^{40}K) obtained from the IAEA laboratory, Vienna. The activity concentration of ^{40}K was directly determined using the 1460.8 keV photo peak. For ^{232}Th, the photo peaks of ^{212}Pb (238.6 keV), ^{208}Tl (583.1 kev) and ^{228}Ac (911.1 keV) were used. The ^{226}Ra concentration was derived from ^{214}Bi (609.3 keV) and ^{214}Pb (295.2 and 352.0 keV) in the same pattern. The well-known interference between the gamma line of 186.2 keV of ^{226}Ra and 185.7 keV emitted by ^{235}U is inevitable, especially in the presence of a high uranium concentration[43,44]. Therefore, the above-mentioned lines were not used for the determination of ^{226}Ra. High-level shielding against the environmental background radiation was achieved by counting in the Canberra 100 mm thick lead castle. Since the accuracy of the quantitative measurements is dependent on the calibration of the spectrometry system and adequate energy, background measurement and efficiency calibration of the system was made possible using Cs-137 standard source from IAEA, Vienna. Spectrum was accumulated for background for 29000 sec at 900 volts to produce strong peaks at gamma-emitting energies of 1461 keV for ^{40}K, 609 keV of ^{214}Bi and 911 keV of ^{228}Ac, which were used to estimate the concentration of ^{238}U (^{226}Ra) and ^{232}Th (^{228}Ra), respectively. The energy resolution of the detector using Cs-137 from the International Atomic Energy Agency (IAEA) is 18% at 662 keV Cs-137 line, while the activity of the standard at the time of calibration is 25.37 K Bq. The background spectrum measured under the same conditions for both the standard and sample measurements were used to correct the calculated sample activities concentration in accordance with[3,4,27,45-47]. The activity concentrations in Bq kg^{-1} of the radio nuclides in the samples were calculated after subtracting decay correction using the expression:

$$C_s = \frac{N_{Ey}}{\varepsilon_{Ey} x M_s x t_c x P\gamma} \ (\text{Bq kg}^{-1}) \quad (1)$$

C_s = Sample concentration
N_{Ey} = Net peak area of a peak at energy
ε_{Ey} = Efficiency of the detector for a γ-energy of interest
M_s = Sample mass
t_c = Total counting time
P_γ = Emission probability of radionuclide of interest[1]

Total effective dose: The total effective dose parameters representing the occupational risk to host communities, oil and gas workers and the public' were estimated employing relevant conversion coefficients available in the literature (Table 2) using the Eq.[48]:

$$D_{ext} = \Sigma A_i C_{ext} T_e \quad (2)$$

$$D_{inh} = \Sigma A_i C_{inh} \eta_{inh} D_f T_e \quad (3)$$

$$D_{ing} = \Sigma A_i C_{ing} \eta_{ing} T_e \quad (4)$$

Where:
A_i = Specific activity of nuclide i in Bq kg^{-1}
C_{ext} = Effective dose coefficient for the nuclide in the contaminated surface measured in Sv h^{-1}/Bq g^{-1}
C_{inh} = Dose coefficient for inhalation of the nuclide measured in Sv Bq^{-1}
η_{inh} = Breathing rate measured in m^3 h^{-1}
D_f = Dust loading factor
C_{ing} = Dose coefficient for ingestion of the nuclide measured in Sv Bq^{-1}
η_{ing} = Ingestion rate for adults, measured in kg h^{-1}
T_e = Exposure duration in the number of years

RESULTS AND DISCUSSION

The mean results of radionuclide concentrations and the calculated effective dose from the three exposure routes of soil samples are presented in Table 1 and 2, is the standard dose coefficients used in the estimation of the effective dose in the soil sample and Table 3 is the computed effective dose values while the statistical comparison results with standard and contour maps are presented in Fig. 3-7.

The average specific activity concentrations of the natural radionuclides measured in soil samples collected from the Coastal Area of Nigeria are as shown in Table 1. The obtained average values of ^{40}K ranged from 40.7±5.81 Bq kg^{-1} in Koko community to 4255.73±123.26 Bq kg^{-1} in the Okpele-ama community with a mean value of 1586.82±17.97 Bq $kg^{-1}$and a control value of 36.54±7.11 Bq kg^{-1}obtained from a non-oil producing community from Aladja community of the same geological formation. The elevated ^{40}K value obtained in the Okpele-ama community may be attributed to the spill of hydrocarbon that has impacted the environment and the presence of oil sludge and scrabs material from the oil well. The ^{238}U average values ranged from Below Detection Level (BDL) in Okerenkoko to 51.54±12.41 Bq kg^{-1} in Abigborodo with a mean value of 27.26±12.41 Bq $kg^{-1}$and a control value of 12.19±0.43 Bq kg^{-1}. The low value of ^{238}U recorded may be attributed to the low activities of oil and gas exploitation and exploration in the

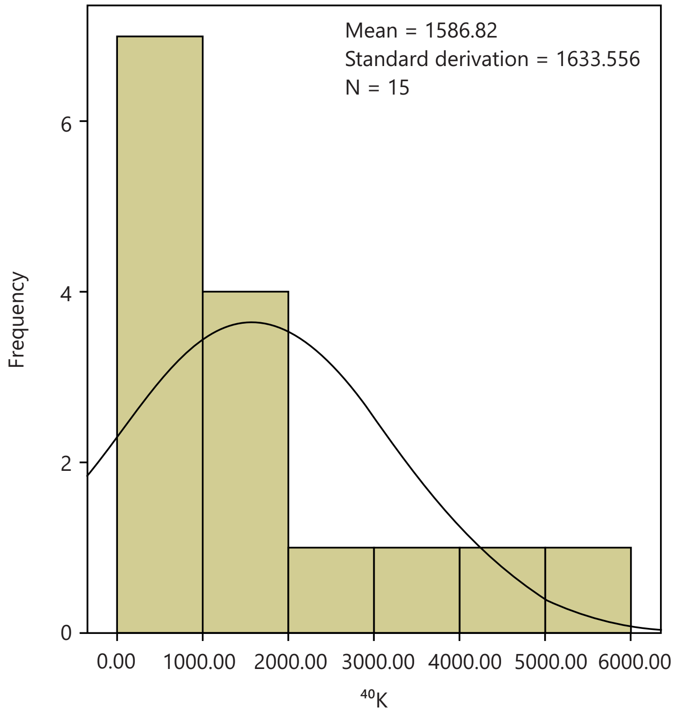

Fig. 1: Frequency distribution histogram of ^{40}K Bq kg^{-1} in soil samples

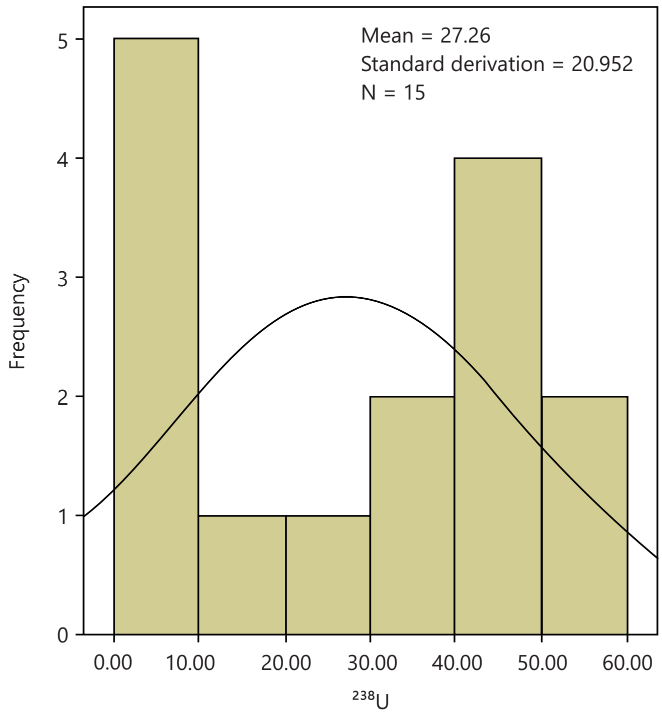

Fig. 2: Frequency distribution histogram of ^{238}U Bq kg^{-1} in soil samples

Okerenkoko community fringe which only host only crude and gas delivering pipelines. However, the elevated ^{238}U value recorded in Abigborodo may not be unconnected with the combined activities of trucking of crude oil and the flow station operational activities which normally through up radon gas and NORM from these facilities coupled with oil spill within the flare stack and bund wall area where measurements were taken. The ^{232}Th average values ranged from 5.92±0.6 Bq kg^{-1} in Oporoza community to 99.60±19.81 Bq kg^{-1} in the Akpele-ama community with a mean value of 37.67±9.55 Bq kg^{-1} and a control value of 3.61±1.09 Bq kg^{-1}. From the ^{232}Th values recorded, the elevated values observed at

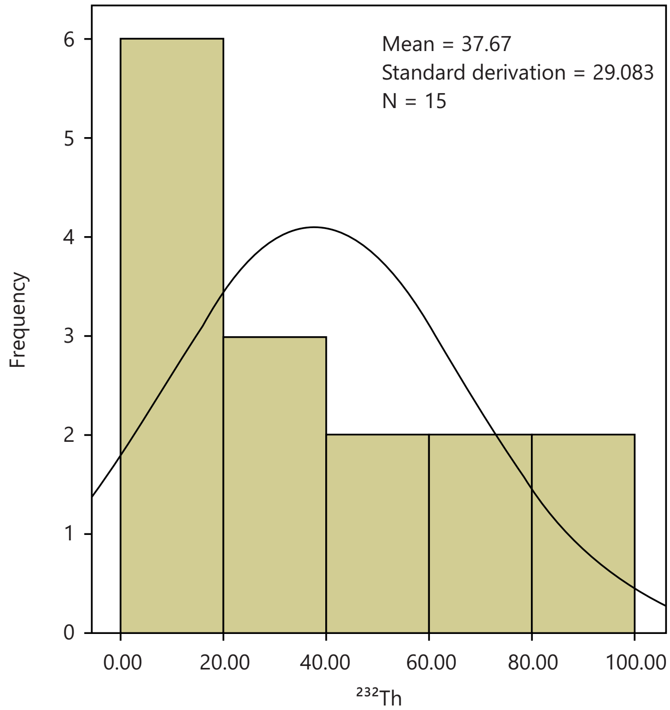

Fig. 3: Frequency distribution histogram of ^{232}Th Bq kg^{-1} in soil samples

Table 1: Average specific activity concentration of ^{40}K, ^{238}U and ^{232}Th (Bq kg^{-1}) results in soil samples

	Average activity		
Communities	^{40}K (Bq kg^{-1})	^{238}U (Bq kg^{-1})	^{232}Th (Bq kg^{-1})
Burutu	1031.58±62.14	24.44±5.40	28.63±2.78
Yeye	535.3±297.9	48.87±22.02	60.60±16.8
Ogulagha	2187.86±25.53	54.42±12.07	32.33±3.17
Forcados	1428.18±33.39	45.10±20.35	38.75±9.06
Odimodi	5566.18±304.74	30.93±7.38	87.22±8.04
Okerenkoko	608.81±40.81	BDL	12.9±1.27
Kunukunuma	1095.14±66.89	2.85±0.68	18.76±1.89
Benikurukuru	770.52±49.71	36.50±8.10	11.21±1.21
Oporoza	1201.81±72.90	0.16±0.04	5.92±0.61
Okpele-Ama	4255.73±123.26	4.11±1.09	99.60±19.81
Koko	40.70±5.81	3.02±0.77	15.82±1.55
Abigborodo	904.15±64.84	51.54±12.41	41.64±13.29
Tebu	466.01±32.73	47.59±21.42	62.67±5.84
Tisum	3634.31±52.42	42.04±9.90	44.07±17.38
Kolokolo	76.03±6.15	17.35±4.04	4.88±0.55
Mean	1586.82±17.97	27.26±8.38	37.67±9.55
Control	36.54±7.11	12.19±0.43	3.61±1.09

Akpele-ama community can be attributed to operational activities of the oil multinational companies including tank farm and pigging manifold that are present and being maintained.

Table 2 are some of the constants used for the computation of effective doses while Table 3 presents the calculated effective doses in soil incurred by residents of the coastal area of the region from ^{238}U and ^{232}Th which are the parent's radionuclides.

The calculated effective dose values of soil samples due to external exposure to contaminated surfaces (D_{ext}) ranged from 0.1557-13.6953 µSv y^{-1} with an average value of 4.2706 µ Sv y^{-1}. The effective dose values due to inhalation pathway (D_{inh}) ranged from 0.0282-0.8033 µSv y^{-1}, with an average value of 0.3674 µSv y^{-1} while that from accidental ingestion (D_{ing}) values ranged from 0.0229-0.3118 µSv y^{-1}, with

Table 2: Dose coefficients and other risk parameters adopted in this study[48]

Parameters	Values	^{226}Ra	^{232}Th	^{40}K
Breathing rate, η_{inh} (m^3 h^{-1})	1.69			
Dust loading factor, D_f (g m^{-3})	1×10^{-3}			
Ingestion rate, η_{ing} (kg h^{-1})	5×10^{-6}			
Duration of exposure, T (h y^{-1})	2000			
Effective dose coefficient, C_{ext} (nSv h^{-1} Bq kg^{-1})		9.929	0.003	1.175
Dose coefficient for inhalation, C_{inh} (Sv Bq^{-1})		2.2E-06	2.9E-05	3.0E-09
Dose coefficient for ingestion, C_{ing} (Sv Bq^{-1})		2.8E-07	2.2E-07	6.2E-09

Table 3: Calculated effective dose in soil samples from the three exposure routes due to radio nuclide activity

		Effective dose (μSv y^{-1})			
S/No	Communities	D_{ext}	D_{inh}	D_{ing}	Total effective dose (mSv y^{-1})
1	Burutu	2.9097	0.2892	0.0977	3.2966
2	Yeye	2.2288	0.4238	0.1517	2.8043
3	Ogulagha	6.2223	0.6298	0.1796	7.0317
4	Forcados	4.2521	0.4841	0.1500	4.8862
5	Odimodi	13.6953	0.8033	0.3118	14.8103
6	Okerenkoko	1.4308	0.0630	0.0331	1.5269
7	Kunukunuma	2.6303	0.1341	0.0586	2.8230
8	Benikurukuru	2.5356	0.3507	0.0873	2.9736
9	Oporoza	2.8275	0.1237	0.0440	2.9951
10	Okpele-Ama	10.0832	0.4722	0.2472	10.8026
11	Koko	0.1557	0.0282	0.0229	0.2068
12	Abigborodo	3.1485	0.4792	0.1460	3.7736
13	Tebu	2.0405	0.4075	0.1500	2.5980
14	Tisum	9.3757	0.6856	0.2200	10.2813
15	Kolokolo	0.5232	0.1372	0.0320	0.6925
Mean		4.2706	0.3674	0.1288	4.7668
Min		0.1557	0.0282	0.0229	0.2068
Max		13.6953	0.8033	0.3118	14.8103

the average value of 0.1288 μSv y^{-1}. The total effective dose received by the residents through the three exposure routes ranged from 0.2068 mSv y^{-1} at Koko community to 14.8103 mSv y^{-1} at Odimodi community with an average value of 4.7668 mSv y^{-1} in Table 3.

The bell-shaped frequency distribution of the measured radionuclide's was analyzed and the histograms are as shown in Fig. 1-3. The histograms distributions explained the even distribution of the measured radionuclides in the coastal area of the Niger Delta Region of Nigeria. The histograms show that ^{238}U, ^{232}Th and ^{40}K exhibited some degree of multi-modality. The multi-model feature of the radioelements demonstrates the complexity and variation in the origin of radioactive laden minerals in soil samples that are the sources of the natural radioactivity concentrations.

The contour maps of mean specific activity concentrations of the radionuclides (^{238}U, ^{232}Th and ^{40}K) distribution in soil samples in the coastal communities studied are as shown in Fig. 4-6. The dense contour lines of the locations show generally higher activity values, while the less dense areas show low activity values and these can be observed in the contour maps of the three natural radionuclides. The observed relatively high radionuclide concentrations of the three radionuclides in almost all the communities investigated indicate that radionuclide emitters may have been introduced over time into the environment which probably has elevated the natural radioactivity level, which can be traced to the oil and gas exploration, exploitation and production activities in the area, which deploy and use some of the radiation generators on a large scale for industrial radiography, automatically ionizing radiation gauge, well logging, use of radiotracers in pipes, mapping and evaluation of geological formation and the extraction of other natural hydrocarbon resources[49].

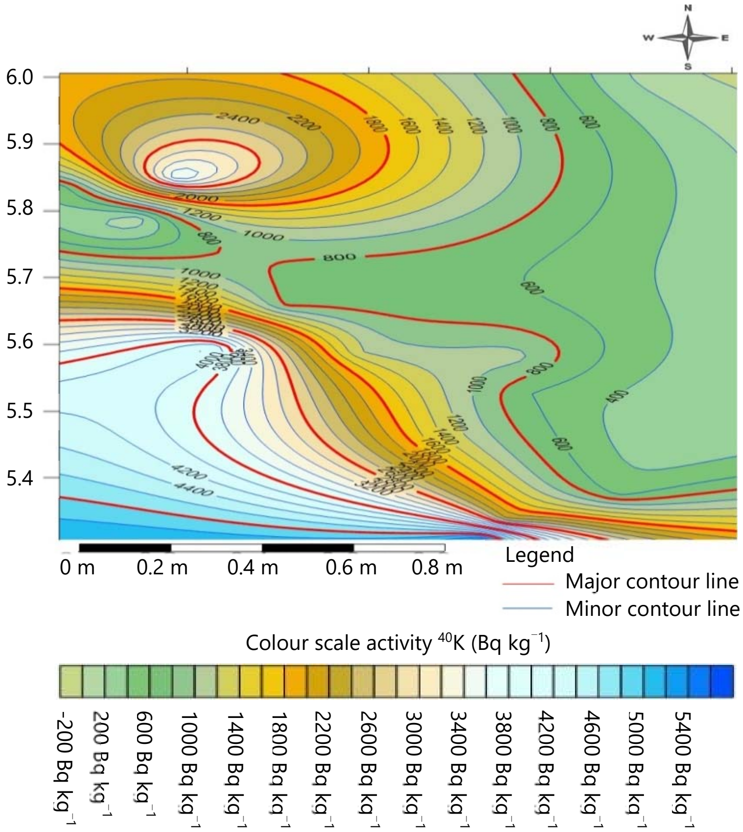

Fig. 4: Mean specific activity concentration results from ^{40}K Bq kg^{-1} for soil

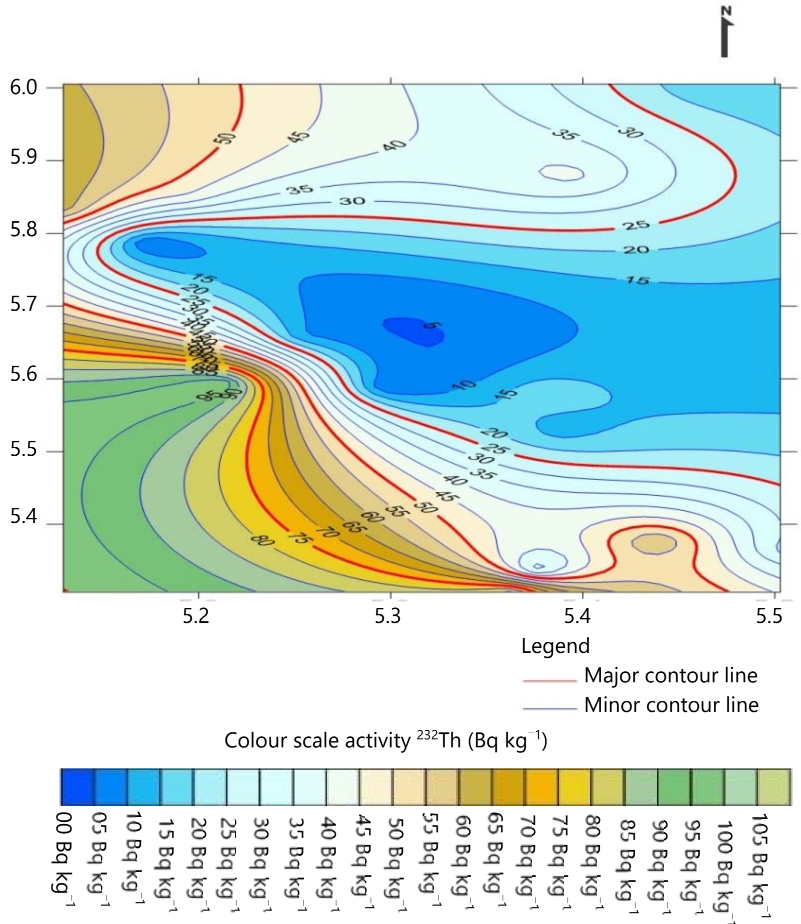

Fig. 5: Mean specific activity concentration results from ^{232}Th Bq kg^{-1} for soil

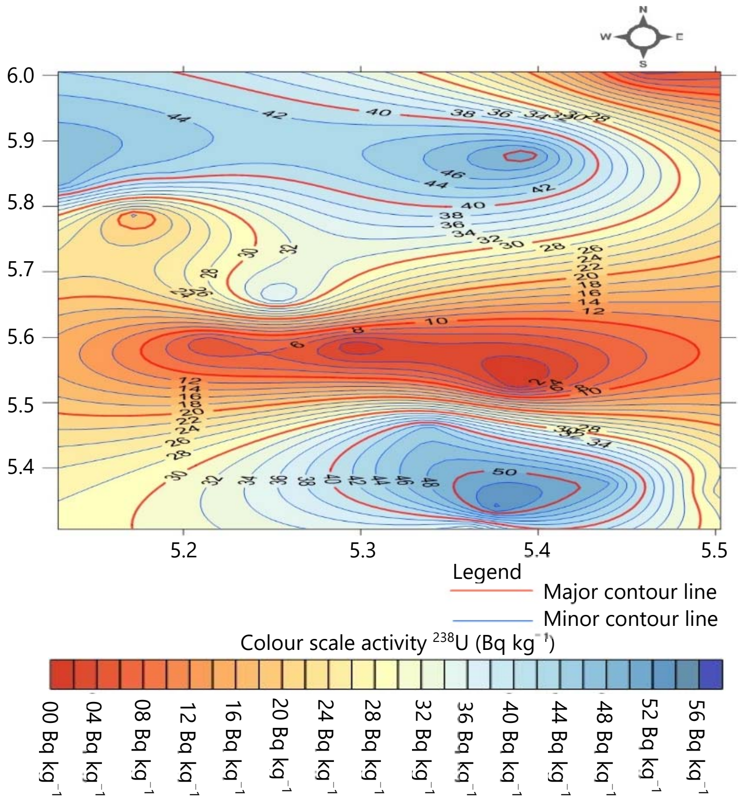

Fig. 6: Mean specific activity concentration results from ^{238}U Bq kg^{-1} for soil

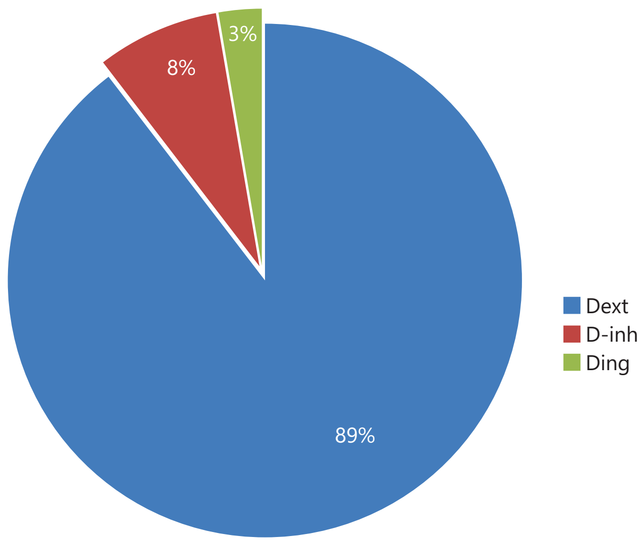

Fig. 7: Percentage contribution of total effective dose in soil samples

The results of the calculated ingested doses from the soil by residents of the coastal area of the oil-producing area for these six decades from these natural radionuclides revealed that the total effective doses obtained in all the communities' soil samples are higher than the recommended permissible limit of 1.0 mSv y^{-1} [50]. These values are also higher than previously estimated values reported by Kolo *et al.*[48]. The results showed that the most significant exposure pathway in soil samples is the external exposure which accounted for about 89.0%, inhalation 8.0% and the least significant is the ingestion which accounted for 3% as shown in Fig. 7. These high effective dose values may also be traced to oil and gas exploration, exploitation and production activities which includes oil spillage, discharge of oil and gas waste (produced water, used drilling mud) into the environment.

The results of the average specific activity concentrations of the natural radionuclides measured in soil samples collected from the coastal area of Nigeria are as shown in Table 1, which shows that the mean values obtained for ^{40}K and ^{232}Th are higher than the standard recommended permissible limit for the public while ^{238}U is within its permissible limit for the public[9]. The average specific activity concentrations of the samples in the studied areas varied significantly from one community to the others across the fifteen communities investigated. This is an indication of the varying degree of impact of human activities on the soil environment. But these results obtained are in agreement with previous studies on soil and sediments in the oil and gas exploration area of the Niger Delta Region of Nigeria and other soil of solid mineral deposit[1,3,4,26,27,37-39,49]. A comparison of the average radioactivity concentration in soil samples in the oil and gas exploitation activities of the Coastal Region with the control sample (within the same area but without any oil and gas exploration activities) reveals that all soil samples of the studied areas are well above the control samples in the three natural radionuclides investigated. Thus, the use of soil in the oil and gas exploitation areas of the Coastal Region for either building materials or other industrial/domestic purposes may lead to radiation exposure beyond the ambient levels which can pose some levels of radiological health impact to the residents and dwellers of the areas. According to the European Commission document on reference levels for workplace processing Naturally Occurring Radioactive Materials (NORMs)[49], high level of regulation and/or individual doses assessment of workers/dwellers may be required if these soil samples are used as building materials.

The radioelements histograms of ^{238}U, ^{232}Th and ^{40}K exhibiting some degree of multi-modality revealed the complexity of radioactive laden minerals in soil samples found in this oil-rich region of Nigerian.

The obtained contour maps of the radionuclide's (^{238}U, ^{232}Th and ^{40}K) showing the distribution of some dense contour lines and less dense areas show variation in the radionuclide distribution in the area which goes beyond NORM in geological formations. The alteration of the environmental NORM may have been introduced over time into the environment through the oil and gas exploration activities which probably have elevated the natural radioactivity level[49,51]. This research work has shown that the most significant exposure pathway of soil samples into man is the external exposure which accounted for about 89.0%, inhalation 8.0% and the least significant is the ingestion which accounted for 3.0%. The high radioactive dose values are attributable to the activities of oil and gas exploration, exploitation and production activities over the years in the study environment.

CONCLUSION

The evaluation of the radioactivity concentration of soil samples from the oil and gas producing area of the Coastal Niger Delta has been carried out after sixty years of oil and gas exploration activities. Soil samples from the Coastal Region of Nigeria were characterized for their activity concentrations (^{238}U, ^{232}Th and ^{40}K) using a gamma spectrometer. The obtained average radio nuclide concentrations values were higher than the standard except for ^{238}U which is lower than the permissible limit for the public. And all measured radio nuclides activity concentrations in the studied areas were higher than the control samples. The estimated total effective dose received by residents and oil and gas workers in the coastal communities through the three exposure routes were found to be higher than the recommended permissible safety limit of 1.0 mSv y^{-1}. External exposure accounted for 89.0% which place it as the most significant exposure pathway for the field workers and the residents. The overall results indicate that the coastal communities' soil is radiologically impaired and thus not safe for building constructions. There is, therefore, the need for proper treatment of oil and gas waste in the coastal area before disposal and processional handling of possible radiation-emitting machines, reduction in gas flair and other activities of the oil and gas multinationals that may tend to aggravate the radiation levels of the coastal areas.

SIGNIFICANT STATEMENT

The study examined the radiological impact of oil and gas exploration activities on the host community residents and the oil field workers in the study area after intense oil and gas exploration for sixty years in the area, using soil as the test case. The results of the radioactivity analyses on the soil samples examined shows that the residents and the field workers in these oil and gas fields environment are significantly and radiologically impacted by the oil and gas activities in the area.

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