

ASSESSMENT OF SPECIFIC ACTIVITY CONCENTRATION AND PERCENTAGE CONTRIBUTION OF ^{226}Ra , ^{232}Th AND ^{40}K TO ABSORBED DOSE RATE OF THE PORT HARCOURT REFINERY COMPANY HOST COMMUNITY

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ABSTRACT

A survey of radioactivity concentration in soil and water from the Port Harcourt refinery company host community was carried out using the gamma ray spectrometer with NaI(Tl) detector to determine the concentration of the primordial radionuclides ^{226}Ra , ^{232}Th and ^{40}K . 20 samples (10 each of soil and water) were collected at random from various parts of the community. The mean specific activity obtained for ^{40}K was $76.64 \pm 22.93 \text{ Bq kg}^{-1}$ (soil), with a range of 41.54 ± 14.80 - $175.54 \pm 39.53 \text{ Bq kg}^{-1}$ and $58.88 \pm 21.66 \text{ Bq l}^{-1}$ (water) with a range of 36.09 ± 14.06 - $121.18 \pm 28.75 \text{ Bq l}^{-1}$. For ^{226}Ra (soil), the mean specific activity was $7.43 \pm 3.47 \text{ Bq kg}^{-1}$ with a range of 6.62 ± 2.24 - $10.10 \pm 4.16 \text{ Bq kg}^{-1}$ and $4.16 \pm 1.97 \text{ Bq l}^{-1}$ (water) with a range of 2.61 ± 1.23 - $5.46 \pm 1.66 \text{ Bq l}^{-1}$ and the mean specific activity for ^{232}Th (soil) was $4.71 \pm 2.46 \text{ Bq kg}^{-1}$ with a range of 3.25 ± 2.10 - $6.26 \pm 4.01 \text{ Bq kg}^{-1}$ and $3.30 \pm 1.90 \text{ Bq l}^{-1}$ (water) with a range of 2.46 ± 2.08 - $4.16 \pm 2.07 \text{ Bq l}^{-1}$. The radionuclide concentrations in the soil are all lower than the ICRP standards, while those for the water samples are significantly higher (six times for ^{40}K ; eight times for ^{226}Ra and 15 times for ^{232}Th). The percentage contribution of each radionuclide to absorbed dose rate in soil and water were also determined. The absorbed dose rates calculated for both samples vary between 24.60 to 79.74 nGy/hr in soil and 18.52 to 54.98 nGy/hr in water. The results of this work indicate that effluents from the refinery have had some impact on the radionuclide concentration of the water of its host community.

Key words: Radionuclide concentration, specific activity, absorbed dose rate, refinery effluent, Gamma spectrometry,

INTRODUCTION

Naturally occurring radioactive materials (NORMS) are acknowledged as the largest sources of exposure to human health (UNSCEAR, 1993, 2000). It is also established that ionizing radiation can cause damage to human tissues and other biological systems (Arafa, 2004; Darkoet al., 2005). Wastes from the oil and gas industry and crude disposal techniques have created serious environmental pollution havoc in many

parts of the Niger Delta region of Nigeria (Chad-Umoren, 2012). Petroleum production companies such as refineries use a wide range of chemicals in their operations as additives. These input chemicals can pollute the soil and groundwater system in the areas where such operations are carried out, especially if proper control mechanisms are not enforced and disposal of waste in accordance with guidelines and standards set by regulatory agencies like the Department of

Petroleum Resources (DPR) and Federal Environmental Protection Agency (FEPA) now Federal Ministry of Environment are not adhered to. The harmful radiological health hazards resulting from anthropogenic activities, especially in the production of energy such as oil and gas extraction and production have attracted some concern over the years in the field of radiation protection (Arogunjo et al., 2004; Chad-Umoren, 2012). A previous study of the ionizing radiation distribution in Rivers state had shown that areas of the state with high density industrial activity, especially oil and gas, tended to exhibit higher radiation levels than those areas not so endowed (Chad-Umoren and Briggs-Kamara, 2010). The work of Jibiri et al (2007) showed that staple food stuffs consumed in some parts of Nigeria contain traces of radionuclides due to anthropogenic activities in the study area. It has also been established that human activities in another part of Nigeria resulted in the presence of traces of radionuclides in the vegetation (Akinloye and Olomo, 2005).

Jibiri and Emelue (2009) conducted radioactivity measurements using gamma-ray spectroscopy to determine the radionuclide concentrations in soil samples at the premises of the Warri Refining and Petrochemical Company located in the Niger Delta region of Nigeria and also in communities around it and reported that activity concentrations of ^{40}K , ^{226}Ra and ^{228}Th in the soil samples ranged from 261.3 to 932.3Bq/kg, 4.2 to 23.0Bq/ kg and 5.1 to 10.2 Bq/kg, respectively. They concluded that the radionuclide concentrations were due to petroleum additives, but that the long duration of refining activities did not seem to have affected radionuclide concentrations in the environment therefore the value are less than the world average outdoor value given by UNSCEAR.

A recent study investigated the activity concentration of naturally occurring radionuclides ^{40}K , ^{226}Ra and ^{232}Th measured in soil samples from six commercial cities in south western Nigeria where different brands of fertiliser are used (Jibiri and Fasae, 2010). Average activity concentration of ^{40}K in the nitrogen, phosphorus and potassium fertilisers across the cities varied from 3972.0 ± 416.9 to 5089.3 ± 111.3 Bq kg^{-1} , 9.9 ± 7.3 to 450.6 ± 14.3 Bq kg^{-1} for ^{226}Ra , while for ^{232}Th it varied from less than lower limit of detection to 15.1 ± 2.8 Bq kg^{-1} . However, high activity concentrations of ^{226}Ra were obtained in the SSP fertiliser and phosphate rocks and in particular, two brands of the fertilisers from two different companies. The study further established that the values of the activity concentration of the radionuclides in the brands of fertilisers evaluated were within the range of values reported in some other countries. Ademola and Atare (2011) report that natural radionuclide concentrations in soil samples collected within and around crude oil flow and gas compression stations in the Niger Delta show that the range of activity concentrations of ^{40}K , ^{238}U and ^{232}Th were 30.1 ± 3.0 to 59.0 ± 17.1 , below detectable limits (BDL) to 8.8 ± 2.3 and 7.9 ± 3.7 to 10.9 ± 1.9 Bq. kg^{-1} , respectively and concluded that the contents of these radionuclides in the soil samples were very low compared with the world average for natural background, hence showing that there is no significant radiation hazard in the study areas.

This present study seeks to determine the impact of effluents from the Port Harcourt Refinery Company on the specific activity concentration and radionuclide doses of the naturally occurring radionuclides: ^{226}Ra , ^{232}Th and ^{40}K in soil and water from the host community, Okochiri of Okirika Local Government Area of Rivers State, Nigeria. During production and operation processes the refinery discharges large

volumes of water and associated treatment chemicals into neutralization basins. Subsequently, the untreated water is pumped into the general drain and then into the Okrika Creek. This conscious and indiscriminate pumping of waste pollutes soil and water with attendant unsustainable and wasteful utilization of resources giving rise to land degradation, non-availability of safe drinking water and a threat to human health (Odunaike et al, 2008).

MATERIALS AND METHODS

Study Area:

This research was conducted at Okochiri, Okirika Local Government Area of Rivers State at sites polluted by wastes discharged from the Port Harcourt Refinery Company (PHRC). Figs. 1 and 2 show parts of the Okirika polluted farm lands. *Okrika is a port town and is the headquarters of the Local Government Area of the same name. The town which lies to the north of the Bonny River is situated on a small island in the mangrove swamps of the eastern Niger Delta just south of Port Harcourt, the capital of Rivers State* (en.wikipedia.org/wiki/Okrika).



Fig.1: Polluted Field 1



Fig. 2: Polluted Field 2

Sample Collection and Preparation

Ten soil samples and ten water samples making a total of twenty samples were collected from ten locations selected at random from the community. The soil samples were dried in an oven maintained at a temperature of 50⁰ C, then grounded and passed through a mesh size of 2mm, sealed for about 28 days in plastic containers previously washed and rinsed with diluted sulphuric acid before analysis with the gamma-spectrometer. The water samples were stored in containers previously acidified with 11 M of HCL at the rate of 10 ml per litre of sample to avoid absorption of radionuclides on the walls of the container. The incubation time was 28 days for a state of secular equilibrium to be reached.

Gamma spectrometry:

The gamma spectrometer used for this work consists of a high purity germanium (HPGe) detector coupled to a multichannel analyser (MCA) with software for data acquisition coupled to a Canberra 1510 signal processing unit containing an analogue to digital converter. Spectrum acquisition and analysis were performed with the APTEC software. The detector and measuring assembly were calibrated for energy and efficiency in order to determine the type of radionuclides and the quantities present.

The activity concentration (Bq/kg) in each of the samples in the spectrum was calculated using the analytical expression:

$$A_c = \frac{N_{sam}}{P(E)*O(E)*T_c*M_{sam}} \quad (1)$$

where M_{sam} (kg) is the mass of sample, N_{sam} (cps) is the net peak area for the sample in peak range, $P(E)$ is the gamma emission probability, T_c is the counting time in seconds, $O(E)$ is the photopeak efficiency.

The activity concentrations of the parent nuclides were obtained using their daughter nuclide activity concentrations assuming attainment of secular equilibrium within the period of storage. The transition lines (609.34, 1764.51keV) of ²¹⁴Bi and (583.32, 2614.56keV) of ²⁰⁸Tl were used to determine the activity concentration of ²²⁶Ra and ²³²Th respectively; while the concentration of ⁴⁰K was determined directly with its own 1460.7keV peak transition line.

Absorbed Dose Rate

The absorbed dose measures the radiation energy absorbed by unit mass of a substance. And the hazard increases with increase in the absorbed dose. The gamma radiation population dose of those living in the area is given as:

$$D=0.462 C_{Ra}+ 0.621 C_{Th}+ 0.0417C_k \quad (2)$$

Where D is the dose rate in nGyh⁻¹ and C_{Ra}, C_{Th} and C_k are the concentrations of radium, thorium and potassium, respectively.

RESULTS**Table 1:** Specific activity concentration of ^{40}K , ^{226}Ra and ^{232}Th (Bq kg^{-1}) in soil

S/No	Sample	^{40}K	^{226}Ra	^{232}Th
1	A	175.54 ± 39.53	7.04 ± 3.12	3.97 ± 1.16
2	B	54.98 ± 19.57	8.35 ± 4.65	3.25 ± 2.10
3	C	52.56 ± 17.23	7.51 ± 3.01	5.16 ± 3.13
4	D	41.54 ± 14.80	6.92 ± 2.84	6.26 ± 4.01
5	E	62.06 ± 21.82	10.10 ± 4.16	4.98 ± 2.78
6	F	50.54 ± 16.80	6.10 ± 2.85	5.72 ± 2.10
7	G	64.08 ± 24.08	7.62 ± 4.64	4.56 ± 1.18
8	H	46.57 ± 36.52	6.62 ± 2.24	5.93 ± 3.13
9	I	52.97 ± 17.10	7.10 ± 3.10	4.01 ± 2.10
10	J	165.60 ± 21.86	6.94 ± 4.10	3.30 ± 2.90
Mean Value		76.64 ± 22.93	7.43 ± 3.47	4.71 ± 2.46
Standard		400	35	30

Table 2: Specific activity concentration of ^{40}K , ^{226}Ra and ^{232}Th (Bq kg^{-1}) in water

S/No	Samples	^{40}K	^{226}Ra	^{232}Th
1	A	52.03 ± 19.35	4.42 ± 2.13	2.97 ± 1.16
2	B	121.18 ± 28.75	4.03 ± 1.45	3.25 ± 2.10
3	C	50.56 ± 17.32	2.61 ± 1.23	4.16 ± 2.07
4	D	49.76 ± 20.08	5.03 ± 2.84	2.67 ± 1.22
5	E	42.06 ± 16.28	5.46 ± 1.66	3.51 ± 1.88
6	F	36.09 ± 14.06	3.84 ± 2.94	2.46 ± 2.08
7	G	52.24 ± 26.65	4.06 ± 1.18	4.01 ± 1.03
8	H	46.58 ± 17.70	2.84 ± 2.10	3.24 ± 1.27
9	I	58.10 ± 22.26	4.02 ± 1.26	2.96 ± 2.06
10	J	80.18 ± 34.10	5.26 ± 2.86	3.75 ± 4.10
Mean Value		58.88 ± 21.66	4.16 ± 1.97	3.30 ± 1.90
Standard		10	0.5	0.2

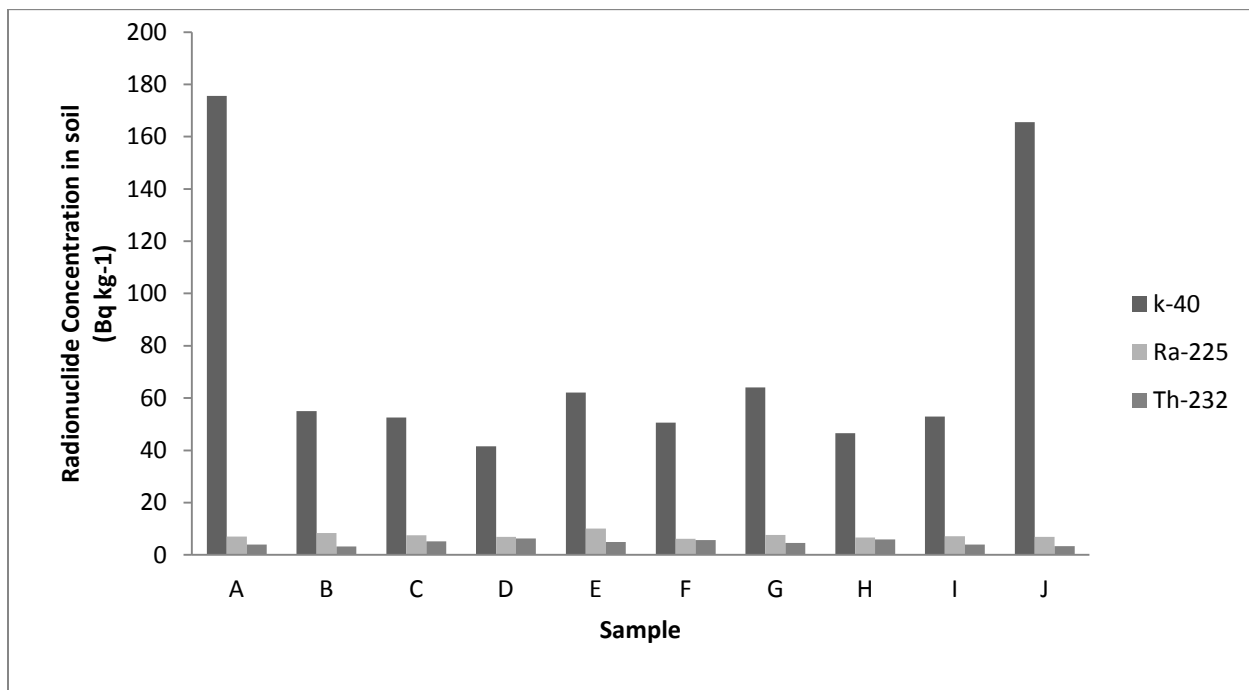


Fig. 3: Radionuclide concentrations in soil

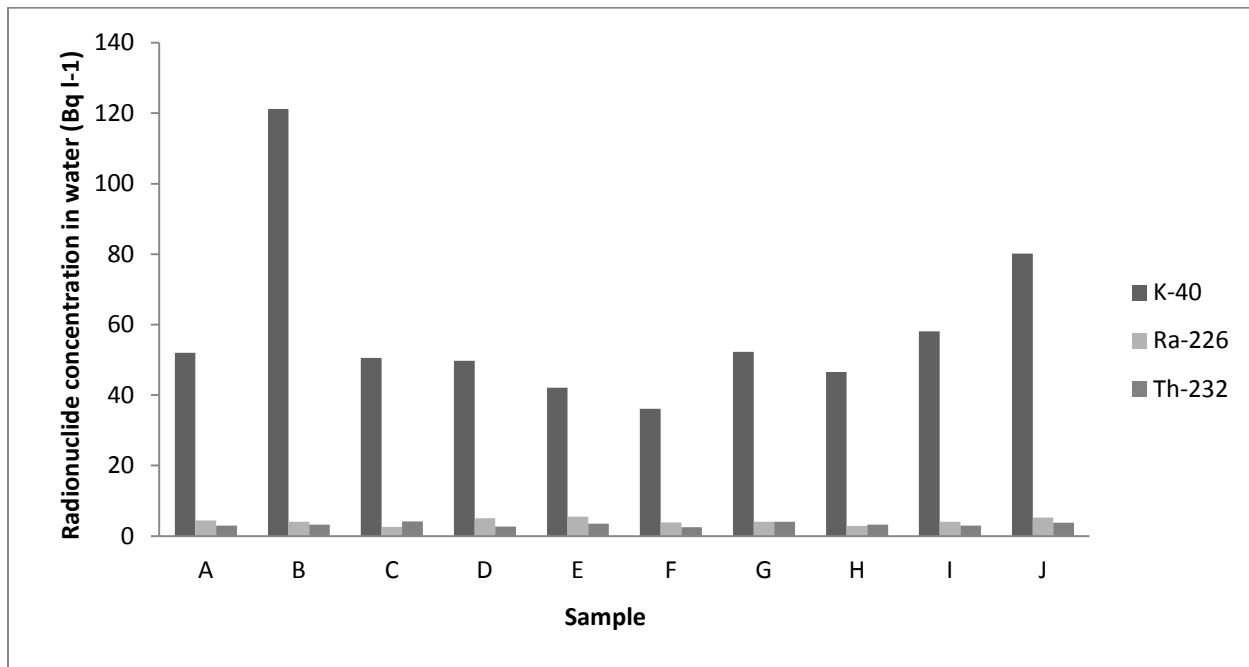


Fig.4: Radionuclide concentration in water

Absorbed dose rate in soil**Table 3:** Calculated Absorbed Dose Rate (D) in Soil.

S/No	Sample	Absorbed Dose Rate (D) (nGy/h)
1	A	79.74
2	B	29.06
3	C	28.84
4	D	24.60
5	E	33.93
6	F	27.68
7	G	33.38
8	H	26.38
9	I	28.11
10	J	75.09
Mean Value		38.68± 12.88
Standard		55nGy/h

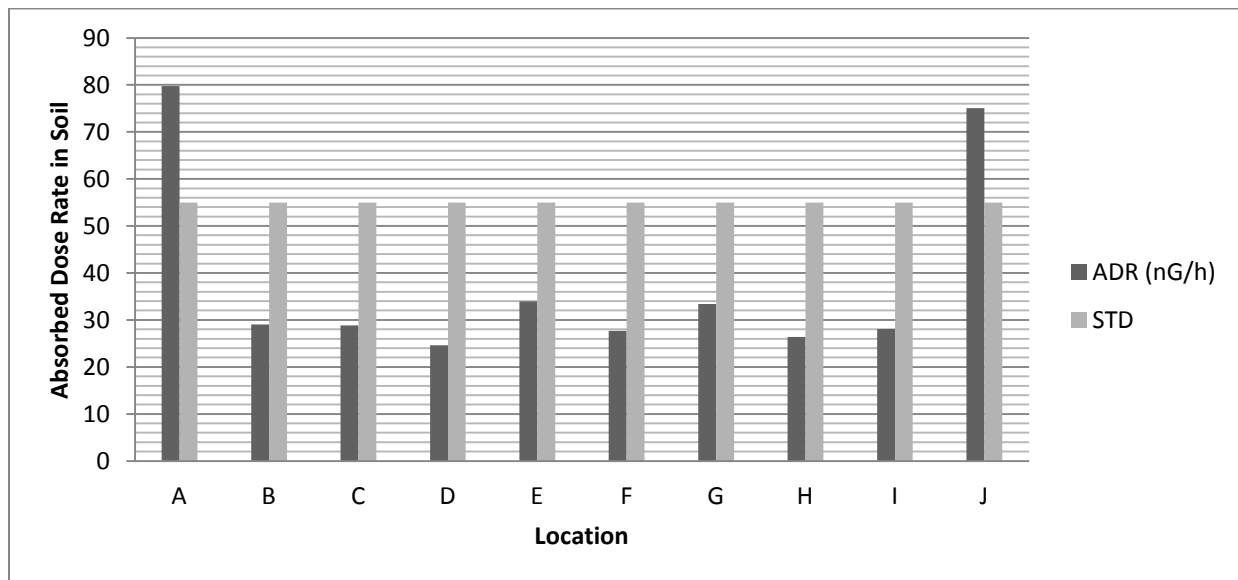


Fig 5: Comparison of Absorbed Dose rate with Standard in soil

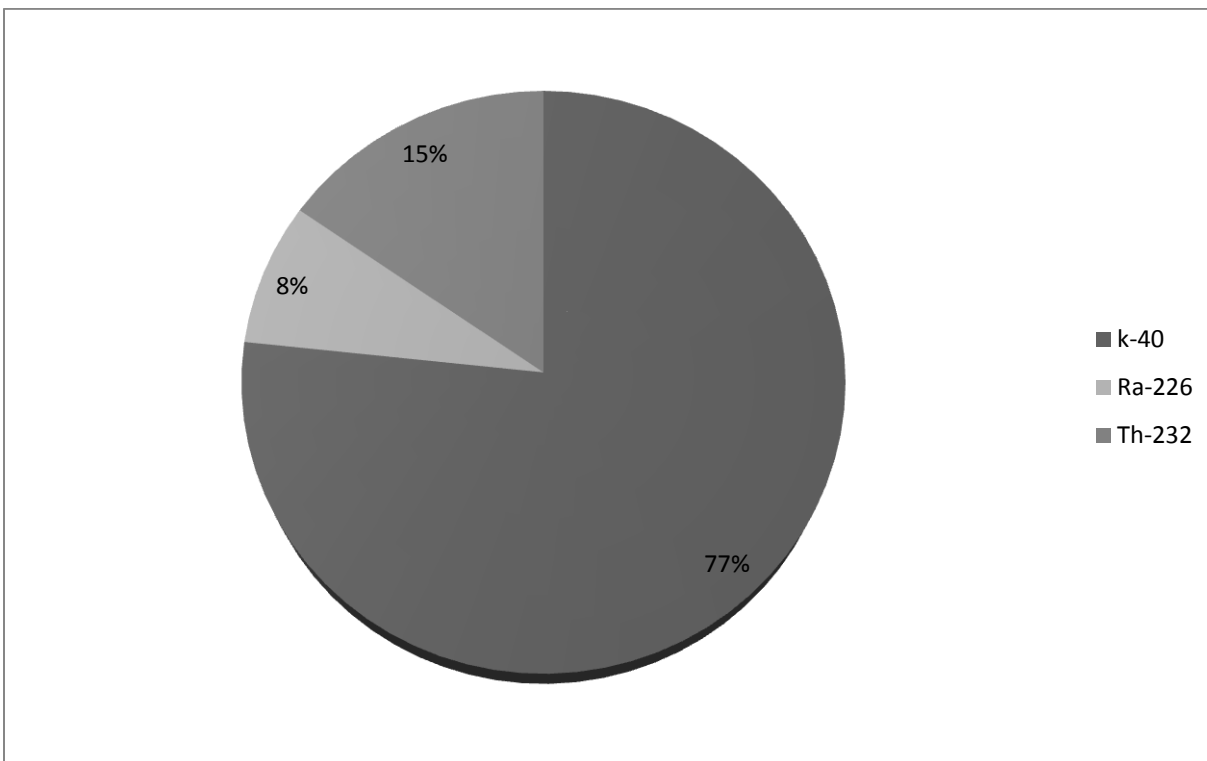


Fig. 6: Percentage Contributions of the Radionuclides to Absorbed Dose Rate in Soil

Absorbed dose rate in water

Table 4: Calculated Absorbed Dose Rate (D) in Water

S/No	Sample	Absorbed Dose Rate (D) (nGy/hr)
1	A	25.83
2	B	54.98
3	C	25.11
4	D	24.97
5	E	22.441
6	F	18.52
7	G	26.40
8	H	22.93
9	I	28.20
10	J	38.57
Mean Value		28.79 ±11.10
Standard		55 nGy/hr

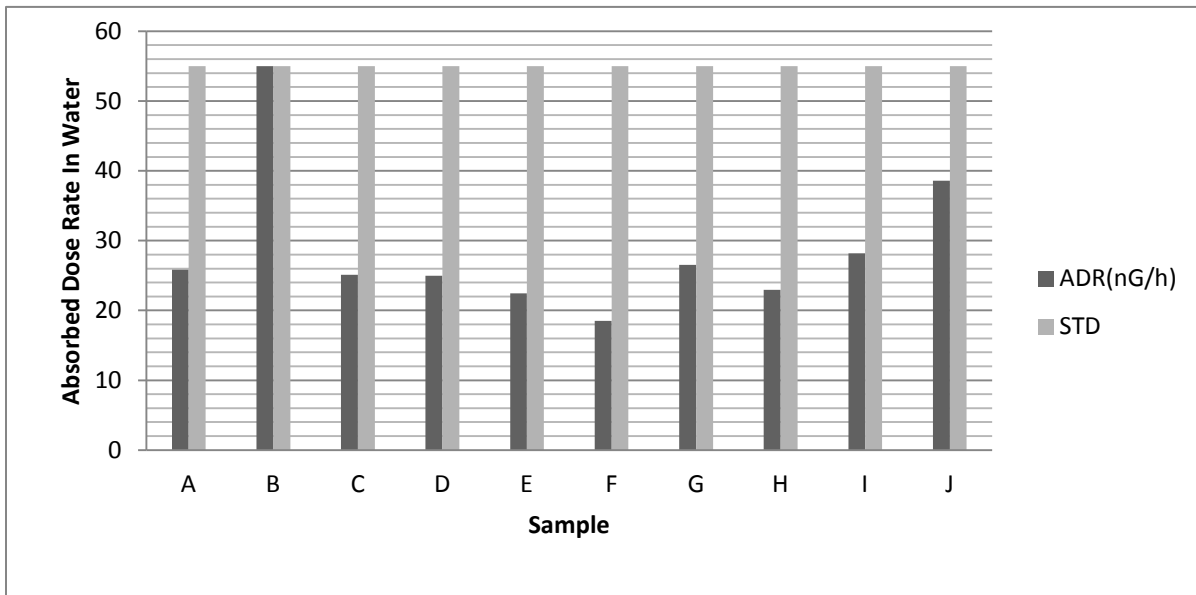


Fig 7: Comparison of Absorbed Dose rate with Standard in water

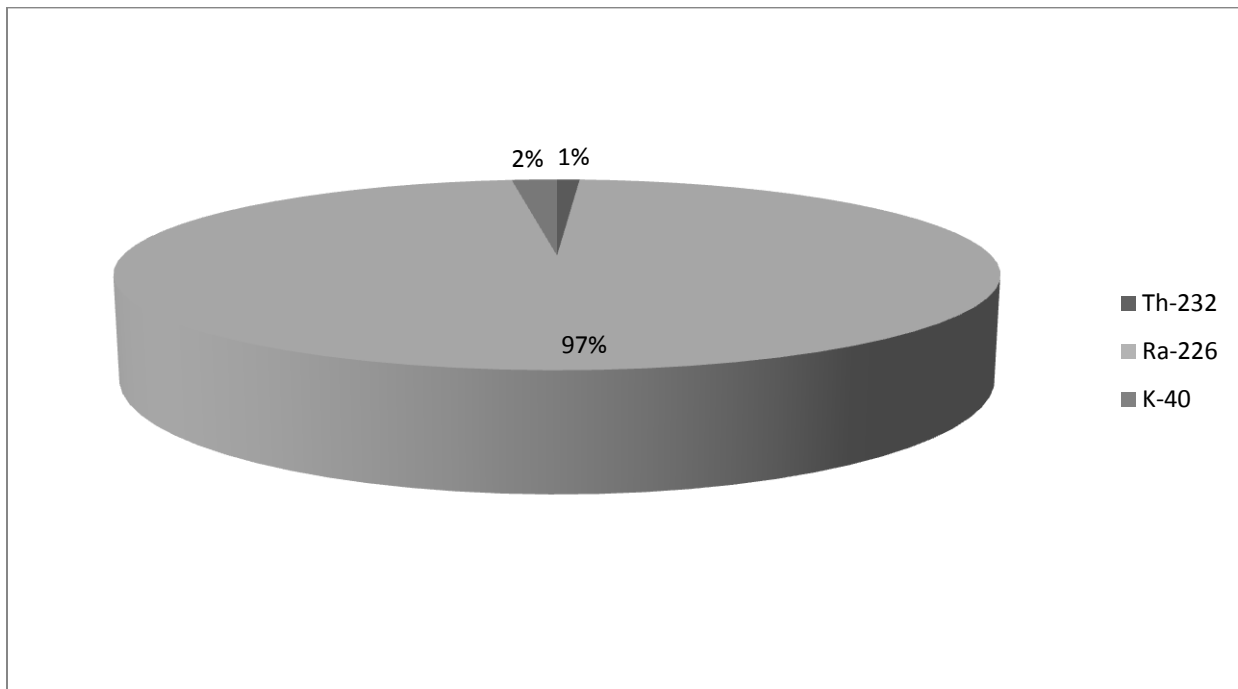


Fig. 8 : Percentage contributions of the radionuclides in water

DISCUSSION

Radionuclide concentration in soil

The radioactivity concentration of the radionuclides in soil is shown in Table 1. Three naturally occurring radionuclides ^{40}K , ^{226}Ra and ^{232}Th were detected in all the samples. The specific activity values obtained in the soil range from 41.54 ± 14.80 to 175.54 ± 39.53 Bq kg⁻¹ with a mean of 71.35 ± 22.93 Bq kg⁻¹ for ^{40}K ; between 6.62 ± 2.24 to 10.10 ± 4.16 Bq kg⁻¹ with a mean value of 5.73 ± 3.47 Bq kg⁻¹ for ^{226}Ra and from 3.25 ± 2.10 to 6.26 ± 4.01 Bq kg⁻¹ with a mean of 4.73 ± 2.46 Bq kg⁻¹ for ^{232}Th . The relative concentrations are shown in Fig. 3 where it is seen that the concentration of ^{40}K is consistently higher than the concentration of the other radionuclides for all the samples and locations. This present results show lower concentrations compared to a previous study at another refining and petrochemical company in Warri, Delta State of Nigeria (Jibiri and Emelue, 2009). A recent study by Agbalagba et al (2012) which measured the natural radioactivity of soil samples from some oil fields in Delta State, Nigeria gave much higher values than the present study (a mean of 412.5 ± 20.0 Bq kg⁻¹ for ^{40}K , 41.0 ± 5.0 Bq kg⁻¹ for ^{226}Ra and 29.7 ± 4.0 Bq kg⁻¹ for ^{232}Th).

An evaluation of natural radioactivity in a cultivated area around a fertilizer plant in Abu-Zabal in Cairo, Egypt (Diab et al., 2008) gave the activity concentration for soil to be in the range of 71.8 ± 24 to 543.2 ± 26.5 Bq/kg with an average value of 264.1 ± 11.94 Bq/kg for ^{40}K ; a range of 6.0 ± 1.2 to 87.5 ± 4.5 Bq/kg with an average value of 31.12 ± 2.22 Bq/kg for ^{226}Ra and a range of 3.8 ± 1.2 to 14.2 ± 3.3 Bq/kg with a mean of 10.96 ± 1.89 Bq/kg for ^{232}Th .

A Comparison of the mean radioactivity concentration of the present study with ICRP standards shows that the radionuclide content of the soil of the study area are much less than the reference standards for each corresponding nuclide.

Radionuclide concentration in water

The specific activity concentration of the radionuclides in water is shown in Table 2. For ^{226}Ra , the concentration obtained ranges between 2.61 ± 1.23 to 5.46 ± 1.66 Bq l⁻¹ with a mean value of 4.11 ± 2.00 Bq l⁻¹; between 2.46 ± 2.08 to 4.16 ± 2.07 Bq l⁻¹ with a mean of 3.30 ± 1.90 Bq l⁻¹ for ^{232}Th and between 36.09 ± 14.06 and 121.18 ± 28.75 Bq l⁻¹ with a mean value of 58.87 ± 21.66 Bq l⁻¹ for ^{40}K . The relative concentrations are shown in Fig. 4 where it is again seen that the concentration of ^{40}K is consistently higher than the concentration of the other radionuclides for all the samples and locations. An analogous study to assess the radiological impact of the petrochemical industry on the Aletto Eleme River showed a significant elevation of the ionizing radiation levels at the point of effluent discharge into the river (Avwiri and Tchokossa, 2006).

A comparison of the mean radioactivity concentration results of the present study with ICRP standards shows that the radionuclide content of the water of the study area are higher than the reference standards for the corresponding nuclide. The mean activity concentration of ^{40}K is 6 times higher; ^{226}Ra has a mean activity concentration that is 8 times higher and ^{232}Th has a mean activity concentration that is 15 times higher than the ICRP reference level, indicating that the effluents from the refinery has impacted negatively on the radionuclide profile of the water of the host community.

In soil the absorbed dose rate ranges from 24.60 to 79.74 nGy/h with a mean of 38.68 ± 12.88 nGy/h (Table 3). The values for all the locations are lower than the reference standard except at two locations where they are higher (Fig. 5). The percentage contribution of each radionuclide to the absorbed dose rate in soil is shown in Fig. 6. It is seen that ^{40}K contributed more than ^{226}Ra and ^{232}Th to the absorbed dose rate in the soil. The 77% contribution of ^{40}K may be attributed to its wide distribution in nature, the fact that it's one of the principal cations in plant cells and a cofactor for enzymes. The implication is that of the three radionuclides, ^{40}K is absorbed the most by the inhabitants of the study area. Long exposure of inhabitants can result in radiation hazards. Also, crops grown on this soil may suffer from retarded growth. The recent study by Agbalagba et al (2012) gives a higher average absorbed dose rate of 54.6 nGy/h for soils affected by oil activities in parts of Delta state, Nigeria.

In water the absorbed dose rate ranges from 18.52 to 54.98 nGy/h with a mean of 28.80 ± 11.10 nGy/h (Table 4). The values for all the locations are lower than the reference standard except at location B where they are about equal (Fig. 7). The percentage contribution of radionuclides to absorbed dose rate in water is shown in Fig. 8. It is observed that ^{226}Ra contributed more than ^{40}K and ^{232}Th . With 97% contribution, the implication is that those drinking or using the water from the Okochiri Community are more exposed to ^{226}Ra than the other radionuclides present in the water.

The following conclusions are drawn from this work:

1. In the case of soil, the mean activity concentrations of the three radionuclides detected are all less than the WHO reference levels for soil; while in the case of water they are all higher than the reference levels, showing that the effluents discharged from the Port Harcourt Refinery Company has impacted more on the radiation profile of the water of the Okochiri community than on the soil.
2. ^{40}K is the highest contributor to the absorbed dose rate in soil, 77%; while ^{226}Ra is the highest contributor to the absorbed dose rate in water with 97% contribution.
3. The calculated absorbed dose rate for both soil and water are well below the reference standard, indicating that the refinery host community is not exposed at the present time to any radiological health hazards; however further studies need to be conducted in the area to conclusively ascertain the radiological risk levels from the refinery.

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