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RESEARCH ARTICLE

NATURAL RADIOACTIVITY OF SURFACE AND GROUND WATER SAMPLES IN COASTAL COMMUNITIES OF DELTA STATE, NIGERIA

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ABSTRACT

The activity concentration of natural radionuclide's; ^{226}Ra, ^{232}Th, ^{40}K of surface and ground water samples in coastal communities of Delta State, Nigeria was measured using a well - calibrated gamma – ray spectrometry system with high – purity germanium detector. The mean activity concentration of radionuclides in surface water samples for ^{226}Ra, ^{232}Th and ^{40}K are $3.2\pm 0.2 BqL^{-1}$, $3.1\pm 0.4 BqL^{-1}$ and $12.3\pm 0.1 BqL^{-1}$ respectively. For the ground water samples the mean activity concentration for ^{226}Ra, ^{232}Th and ^{40}K are; $2.1\pm 0.2 BqL^{-1}$, $0.9\pm 0.2 BqL^{-1}$ and $4.9\pm 0.6 BqL^{-1}$ respectively. The gamma representative index, radium equivalent activity values for the water samples showed that they constitute very low radiological hazard to human and the environment.

INTRODUCTION

The natural radionuclides in the earth or soil and water of an environment are present as daughters of (^{238} U, ^{232}Th, ^{40}K) isotopes distributed by natural geological and geochemical process in addition to small quantities of fission – product residues ^{137} Cs from atmospheric weapon test (Trimble 1968). Natural radionuclides and other progenies could be radiotoxic and carcinogenic when present at enhanced levels in water, food and the environment. The most important radionuclide especially in water is ^{226}Ra. Their presence in drinking water and associated damaging biological effect can lead to serious health risk when the populace is exposed to high level of radium over a period of time, cancer of bone and nasal cavity could result (Nour 2004, Rowland 1993). Activity concentration in water samples from Nigeria have been studied and reported by other workers (Olomo *et al*., 1994; Avwiri 2005, Ajayi and Adesida, 2009). The aim of this paper is to identify and determine the activity concentrations of natural radionuclide in surface and tap water (bore holes) from five coastal communities in Delta State, Nigeria. Also, to determine the potential radiological risk of the water samples to humans and the environment.

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MATERIALS AND METHODS

Sample Collection and Preparation

Five surface (river) water Samples and five tap water samples from boreholes (depth ranged from 20m to 25m) were collected from five coastal communities in Delta State, Nigeria (Fig.1). The collection of the water was done using plastic bottles of 1.5 liters. The containers were earlier thoroughly rinsed with distilled water before the water samples were collected. Two drops of IM hydrochloric acid was immediately added to the water samples after collection to prevent adsorption of the radionuclide to the walls of the container. The bottles were filled to the brim without any head space to prevent trapping of Co_2 gas. For activity concentration measurement, the water samples were taken to the laboratory and prepared into 1 liter Marinelli beakers. The samples were not filtered prior to preparation and measurements. The beakers were thick enough to prevent the permeation of radon. The beakers were closed by screw caps and plastic tapes was wrapped over the caps and then stored for measurement. This step was necessary to ensure that radon gas is confined within the volume and that the daughters will also remain in the sample.

Sample Measurement and Analysis of Spectra

The activity concentration of the water samples were determined by a non-destructive analysis a computerized gamma ray spectrometry system with high purity germanium (HPGe). The gamma spectrometer is coupled to conventional

electronics connected to a multichannel analyzer card (MCA) installed in a desk top computer. A software program called MAESTRO- 32 was used to accumulate and analyze the data manually using spread sheet (Microsoft Excel) to calculate the natural radioactivity concentrations in the samples. The detector is located inside a cylindrical lead shield of 5cm thickness with internal diameter of 24cm and height of 60cm, the lead shield is lined with various layers of copper, cadmium, and Plexiglas, each 3mm thick. A counting time of 36,000 seconds (10hrs) was used to acquire spectra data for each sample. The activity concentrations of the uranium-series were determined using γ-ray emission of ^{214}Pb At 351.9 keV (35.8%) and ^{214}Bi at 609.3 keV (44.8%) for ^{226}Ra, and for the ^{232}Th-series, and the emission of ^{228}Ac at 911 keV (30.1%) were used. The ^{40}K activity concentration was determined directly from its emission line at 1460.8 keV (10.7%). The γ-rays emission of 137Cs was determined at 661.66 keV.

Calculation of Activity Concentration

The specific activity concentrations (A) of ^{226}Ra, ^{232}Th ^{40}K and ^{137}Cs in BqL^{-1} for the water samples were determined using the following expression.

$$A = \frac{N}{P_E \varepsilon T_C M} \quad (1)$$

N. Net counts of radionuclide in the sample
P_E- Gamma ray emission probability (gamma yield)
ε - Total counting efficiency of the detector system
T_C - Sample counting time
M- Volume (L)

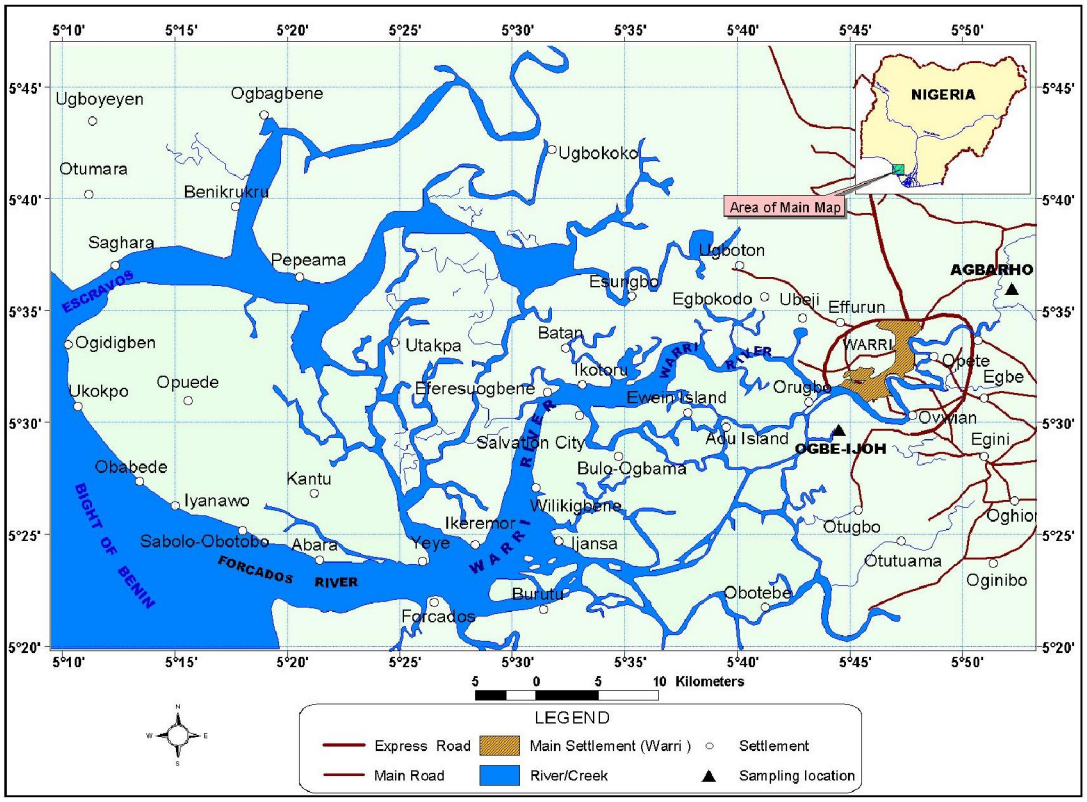

Figure 1. Map of the study area

Calibration of Gamma Spectrometry System

Prior to the measurements, the detector and measuring assembly were calibrated for energy and efficiency to enable both qualitative and quantitative analysis of the samples to be performed. The energy and efficiency calibrations were performed using mixed radionuclide calibration standard uniformly distributed in the form of solid water, serial number NW 146 with approximate volume1000mL and density 1.0gcm^{-3} in a 1.0 L Marinelli beaker. The standard was supplied by Deutscher Kalibrierdienst (DKD-3), QSA Global GmBH, Germany and contain radionuclides with known energies (^{241}Am 59.54 keV), ^{109}Cd(88.03 keV),^{57}Co(122.06 keV), ^{139}Ce(165.85 keV), ^{203}Hg(279.20 keV), ^{113}Sn(391.69 keV), ^{85}Sr(514.0 keV), ^{137}Cs(661.66 keV), ^{60}Co(1173.2 keV and 1332.5 KeV) and ^{88}Y(898.04 KeV and 1836.1 KeV) and activities in a 1000ml Marinelli beaker was used.

Minimum Detectable Activity

The minimum detectable activity (MDA) of the γ-ray measurements were calculated according to the formula:

$$\text{MDA} = \frac{\sigma\sqrt{B}}{\varepsilon PTW} \quad (2)$$

Where σ is the statistical coverage factor equal to 1.645 confidence level 95%, B is the background counts for the region of interest of a certain radionuclide, T is the counting time in seconds, P is the gamma yield for any particular element, W is the weight of empty Marinelli beaker and ε is the efficiency of the detector. The minimum detectable activity (MDA) derived from background measurements was approximately 0.11Bq L^{-1} for ^{226}Ra, 0.10 Bq L^{-1} for ^{232}Th and

0.15 Bq L^{-1} for ^{40}K. Concentration values below these detection limits have been taken in this work to be below the minimum detection limit (MDL).

Radiation Hazard Assessment

The following parameters were used for the evaluation of the potential radiological hazard associated with the radio nuclides in the surface and the ground water samples. They are: radium equivalent activity, gamma representative index, the external and internal hazard indices.

Radium Equivalent Activity

The activity concentrations of ^{226}Ra ^{232}Th and ^{40}K in the water samples can be compared by using the equivalent activity ($^{226}Ra_{eq}$), which is a popular radiological index used to evaluate the actual radioactivity in a material by a single quantity. Radium equivalent activity Based on the estimation that ^{370} BqL^{-1} of ^{226}Ra, $^{259}BqL^{-1}$ of ^{232}Th and $^{4810}BqL^{-1}$ of ^{40}K produce the same gamma-ray close rate in the samples (Beretka and Mathew, 1985; Darko *et al*., 2011). The radium equivalent activity can be expressed as follows:

$$Ra_{eq} = A_{Ra} + 1.43\ A_{th} + 0.077A_K \quad (3)$$

Where: A_{Ra} $A_{Th,}$ A_k are the activity concentrations (BqL^{-1}) of ^{226}Ra, ^{232}Th, ^{40}K respectively. The Ra_{eq} was calculated for both the surface and water sample from the coastal communities studied.

External and Internal Hazard Indices

Assessment of the gamma-radiation levels associated with natural radionuclides in water samples was calculated base on the external and internal hazard indices. The external hazard index H_{ext}, is used to estimate the level of radiological risk of the samples to the immediate environment.

The value of H_{ext} must be less than one (Hamlat *et al*., 2001, Heaton and lambley, 1995; Darko *et al*., 2011). The value of Hext was calculated as

$$H_{ext} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \quad (4)$$

The internal exposure to radon and its daughter products were quantified by the internal hazard index ($H_{int.}$). To account for this, the maximum permissible concentration of ^{226}Ra must be reduced to half of the original activity (Darko *et al*, 2011; El-Afifi *et al*., 2004; El-Afifi and Awward, 2005).

$$H_{int.} = \frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \leq 1$$

Where A_{Ra}, $A_{Th,}$ A_k, have been earlier defined. The internal hazard index should be less than one in order to have a low risk of radiological hazards on the respiratory organs from radon and its daughters.

Gamma radiation representative index (I_γ)

It is used to estimate the γ- radiation hazard associated with the natural radionuclide in specific investigated samples. The representative index has been defined by (NEA-OECD, 1979) as $I_\gamma = A_{Ra}/150 + A_{Th}/100 + C_k/150 \leq 1$ values of $I_\gamma \leq 1$ corresponds to annual effective dose of less than or equal to 1mSv, while $1_\gamma \leq 0.5$ correspounds to annual effective dose less or equal to 0.3mSv (Turham *et al*., 2008) Avwiri *et al*. (2012).

RESULTS AND DISCUSSION

The result of the activity concentration obtained in this work are presented in Tables 1 and 2 for the surface (river) and ground (borehole) water samples respectively. In table 1, the activity concentration for ^{226}Ra ranged between 0.88 $\pm$ 0.13 – 5.60 $\pm$ 0.80 BqL^{-1} with mean value of 3.15 $\pm$ 0.29 BqL^{-1}. ^{232}Th activity concentration range from 0.90 $\pm$ 0.14 – 4.4 $\pm$ 0.05 BqL^{-1} with average of 3.05 $\pm$ 0.39 BqL^{-1} while ^{40}K activity

Table 1. The activity concentration of radionuclides in surface (river) water samples

Sample ID	Coordinates	Lithology	^{226}Ra BqL^{-1}	^{232}Th BqL^{-1}	^{40}K BqL^{-1}
S1	N05.44037° E005.82111°	Limestone, Sand stone	5.6 ± 0.8	4.4± 0.1	8.5 ± 0.9
S2	N05.44729° E005.87140°	Limestone, Sand stone	2.6 ± 0.2	0.9± 02	4.81 ± 0.2
S3	N05.44125° E005.83321°	Sand stone	3.3 ± 0.1	3.3± 0.6	14.1 ± 1.4
S4	N05.57049° E005.68766°	Sand stone	3.4± 0.2	3.2 ± 0.7	17.5 ± 1.7
S5	N05.53742° E005.69172°	Sand stone	0.9± 0.4	3.5 ± 0.5	16.5 ± 1.1
	Mean		3.2 ±0.3	3.1 ± 0.4	12.3 ±1.1

Table 2. The activity concentration of radionuclides in ground (bore hole) water samples

Sample ID	Coordinates	Lithology	^{226}Ra BqL^{-1}	^{232}Th BqL^{-1}	^{40}K BqL^{-1}
T1	N05.44037° E005.82111°	Limestone Sand stone	4.8 ± 0.2	3.2±0.67	6.4 ± 0.9
T2	N05.45094° E005.86865°	Limestone Sand stone	0.7± 0.2	0.30±0.18	2.6 ± 0.9
T3	N05.4438° E005.83281°	Sand stone	0.8±0.21	0.38±0.11	4.1 ± 0.4
T4	N05.57219° E005.68904°	Sand stone	3.4± 0.2	0.4±0.10	5.4 ± 0.5
T5	N05.53754° E005.69212°	Sand stone	0.5 ± 0.1	0.20±0.2	5.9 ± 0.4
		Mean	2.1 ± 0.2	0.9 ± 0.2	4.9 ±0.6

Table 3. Radiation Hazard Indices

Water sample ID	Radium equivalent activity (R_{aeq})	Gamma representative index (I_γ)	Gamma Index External (H_{ext})	Gamma index Internal (H_{int})
Surface				
S1	12.552	0.068	0.209	0.224
S2	4.299	0.009	0.110	0.118
S3	9.094	0.011	0.315	0.324
S4	0.294	0.031	0.385	0.394
S5	7.152	0.008	0.359	0.361
Bore hole				
T1	9.918	0.087	0.157	0.172
T2	1.290	0.030	0.057	0.059
T3	1.529	0.064	0.089	0.091
T4	4.477	0.066	0.123	0.132
T5	1.047	0.052	0.124	0.126

Table 4. ^{226}Ra activity concentration range in drinking water around the world

Country	Concentration range in Bq L^{-1}	Source
United states of America	0.4 – 1.8	UNSCEAR, 2000
France	7.0 – 700	UNSCEAR, 2000
Finland	10.0 – 4900	UNSCEAR, 2000
Germany	1.0 – 1800	UNSCEAR, 2000
Italy	0.2 – 1200	UNSCEAR, 2000
Poland	1.7 – 4.5	UNSCEAR, 2000
Spain	< 20 – 1200	UNSCEAR, 2000
Turkey (Istanbul)	11 – 36	Karahan *et al.,* 2000
Turkey (Eastern Black sea)	3 – 45	Cevik *et al.,* 2006
Egypt	48.8 – 112.9	Ahmed, 2004
Nigeria (South west)	2220 – 15500	Ajayi and Adesida, 2009
Nigeria (Coastal Communities Delta State)	450 – 4830	Present study

concentration range from 4.80 ± 0.2 – 17.48 ± 1.7 BqL^{-1} with mean value of 12.27 ± 1.05 BqL^{-1}. ^{40}K has the highest mean activity concentration in the surface water samples. Therefore, will have the highest contributions to the environmental and humans radiation dose (if the water is use for drinking). The natural radionuclides activities concentration obtained for surface water sample in this study were generally lower in value when compared with the result of (Umar *et al*., 2012) they obtained the following values in their (elsewhere in Nigeria) study for ^{40}K mean of 53.6 ± 5.9 BqL^{-1}, ^{226}Ra mean of 19.7 ± 1.5 $BqL^{-1}$for ^{232}Th mean of 31.2 ± 1.8 BqL^{-1}. The radium activity concentration published in Nigeria and elsewhere around the world is shown in Table 4. The radium activity concentrations in this work are within the range published by (Ajayi and Adesida, 2009). The calculated value for radium equivalent activity and the external and internal radiation indices are shown in table 3. The values for the surface water samples ranged from 4.3 BqL^{-1} to 12.66 BqL^{-1}. These values for the samples are below the recommended limit of 370 BqL^{-1}. The internal (H_{int}) and external (H_{ext}) hazard indices for surface water samples are less than unity, this implies very low radiological hazard to the environment and humans. The activity concentration obtained for the bore hole water samples, commonly used for drinking and domestic purposes in Table 2. Shows that the activity concentration for ^{226}Ra ranged between 0.5 ± 0.1 BqL^{-1} and 4.8 ± 0.2 BqL^{-1} with mean value 2.1 ± 0.2 BqL^{-1}, ^{232}Th activity concentration values ranged between 0.10 ± 0.01 BqL^{-1} – 3.2 ± 0.6 BqL^{-1} with mean value of 0.9 ± 0.2 BqL^{-1} and ^{40}K values ranged between 2.6 ± 0.1 BqL^{-1}, and 6.4 ± 0.1 BqL^{-1}, the mean value is 4.9 ± 0.6 BqL^{-1}. The mean concentration for ^{40}K was a higher than ^{226}Ra and ^{232}Th in this sample. However, mean activity concentration in the borehole water samples were lower than the mean activity concentration for the surface water. This may arise due to pollution from human activities in contact with the rivers in the communities. The activity concentration obtained in this study was lower than the values obtained for drinking water in Nigeria by (Umar *et al*., 2012). The ^{226}Ra concentrations are within the range published by (Ajayi and Adesida 2009) for drinking water in Nigeria in Table 4. The radiation hazard assessment of the borehole water samples in Table 3, the radium equivalent values are lower than the recommended limit of 370 BqL^{-1}. The internal, external and gamma indices values were less than unity, which shows that the borehole water samples has very minimal radiological hazard for human drinking and the environment.

Conclusion

The activity concentration of ^{226}Ra, ^{232}Th and ^{40}K in surface water (River) and ground water (borehole) Samples collected from five selected communities in coastal area of Delta State, Nigeria have been determined. The mean activity concentration of ^{226}Ra, ^{232}Th and ^{40}K in surface water samples are:3.2± 0.3 BqL^{-1}, 3.11±0.4, BqL^{-1} and 12.3± 1.1 BqL^{-1} respectively. For the ground water samples mean activity concentration for ^{226}Ra, ^{232}Th and ^{40}K are: 2.1±0.2 BqL^{-1}, 0.9±0.2 BqL^{-1} and 4.9± 0.6 BqL^{-1}, respectively. The radiological hazard assessment for the samples showed that the water samples constitute insignificant radiological hazard to humans and the environment.

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