Annual Effective Dose, Radium Equivalent Activity and Harzard Indices Assessment in Rainwater from Selected Areas in Akwa Ibom State, Nigeria

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Abstract: This study has been carried out mainly for the assessment of naturally occurring radionuclides i.e $^{40}\mathrm{K},\,^{238}\mathrm{U}$ and $^{232}\mathrm{Th}$ in rain water samples collected from five LGAs in Akwa Ibom State, Nigeria. The activity concentrations of the naturally occurring radionuclides $^{40}\mathrm{K},\,^{238}\mathrm{U}$ and $^{232}\mathrm{Th}$ in the rainwater samples were measured by the means of a gamma-ray spectrometry using a sodium Iodide Thallium doped NaI (TI) detector. The average activity concentration obtained for $^{40}\mathrm{K}$ in all the location was 27.8 ± 2.50 Bq.L $^{-1}$ with a range of $9.4 \pm 0.88 - 52.3 \pm 4.26$ Bq.L $^{-1}$, while for $^{238}\mathrm{U}$, the average activity concentration was 5.2 ± 0.88 Bq.L $^{-1}$ with a range of $1.2 \pm 0.21 - 15.00 \pm 2.92$ Bq.L $^{-1}$, for $^{232}\mathrm{Th}$, the average activity concentration was 6.7 ± 0.69 Bq.L $^{-1}$ with a range of $0.1 \pm 0.00 - 14.2 \pm 1.41$ Bq.L $^{-1}$. The total annual effective dose due to the intake of $^{40}\mathrm{K}$, $^{238}\mathrm{U}$ and $^{232}\mathrm{Th}$ by all the locations ranged from 0.08 ± 0.01 mSv.y $^{-1} - 1.53 \pm 0.17$ mSv.y $^{-1}$ with an average of 0.76 ± 0.08 mSv.y $^{-1}$ did not show any significant health impact since it is below the recommended public exposure limit of 1mSv.y $^{-1}$. Also the calculated values of Radium Equivalent Activity, external and internal hazard index in all the locations were found to be lesser than the recommended values.

Keywords: Natural radionuclide, dose, gamma spectroscopy, radiation, Akwa Ibom, activity concentration.

I. INTRODUCTION

Radionuclides are present in the air that humans breathe and in food and drinking water [1, 2] consumed by man and in the ground from which human settlements are built [3]. Enhanced levels of uranium, thorium and their daughter products might be present in water in area that is rich in natural radioactivity or through human activities. The dumping of large amount of waste materials in site without adequate soil protection measures results in soil as well as, surface and ground water pollution [4] Contaminants from human activities pass into air, soil and water, and, hence into fish crops and other animals. The input of radionuclides such as ⁴⁰K to the environment is derived from terrestrial soil and atmospheric diffusion [5]. Considering the high radio toxicity of ²²⁶Ra and ²²⁸Ra, their presence in water and the associated health risks require particular attention.

A radionuclide is an atom with an unstable nucleus which, to become more stable, emits energy in the form of rays or high speed particles which are known as ionizing radiation [6]. Radiation damage to tissue and/or organ depends on the dose

of radiation received, or the absorbed dose which is expressed in a unit called the gray [Gy] [7]. The potential damage from an absorbed dose depends on the type of radiation and the sensitivity of different tissues and organs. Radionuclides when ingested or inhaled enter the human body and are distributed among body organs according to the metabolism of the element involved. The organs normally exhibit varying sensitivities to the radiation and thus, varying dose and risk result from their consumption and inhalation [8]. Beyond certain thresholds, radiation can impair the functioning of tissues and/or organs and can produce acute effects such as skin redness, hair loss, radiation burns, or acute radiation syndrome [7]. These effects are more severe at higher instance; the dose threshold for acute radiation syndrome is about 1000 mSv. If the dose is low or delivered over a long period of time (low dose rate), there is greater likelihood for damaged cells to successfully repair themselves. However, long term effects may still occur if the cell damage is repaired but incorporates errors, transforming an irradiated cell that still retains its capacity for cell division. This transformation may lead to cancer after years or even decades have passed. Effects of this type will not always occur, but their likelihood is proportional to the radiation dose. This risk is higher for children and adolescents, as they are significantly more sensitive to radiation exposure than adults. Epidemiological studies on populations exposed to radiation (for example atomic bomb survivors or radiotherapy patients) showed a significantly increase of cancer risk at doses about 100 mSv [7]. Prenatal exposure to ionizing radiation may induce brain damage in foetuses following an acute dose exceeding 100 mSv between 8-15 weeks of pregnancy and 200 mSv between 16-25 weeks of pregnancy. Before the 8th week or after the 25th week of pregnancy human studies have not shown radiation risk to fetal brain development. Epidemiological studies indicate that cancer risk after fetal exposure to radiation is similar to the risk after exposure in early childhood.

II. MATERIALS AND METHODS

A. Study Area

Akwa Ibom is a state in Nigeria. It is located in the Coastal Southern part of the country, lying between Latitudes 4°32°N

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and 5°33°N, and Longitudes 7°25°E and 8°25°E. It consists of 31 Local government areas (L.G.A). It covers an area of about 7,081Km² (2,734Sqmi). As at 2005, it estimated population was about 4,805,470. The major cities in the state are Uyo, Eket, Ikot Ekpene, Oron, Abak, Ikot Abasi, Ikono, Etinan, Esit, Eket and Uruan. The major ethnic groups of the state are Ibibio, Annang, Oron, Eket and Obolo.

For this study, five L.G.A of Akwa Ibom State were considered. The Local Government Areas include Uyo, Ikot Ekpene, Essien Udim, Obot Akara and Ikono. The choice for Uyo and Ikot Ekpene L.G.A for this study is based on the fact that this L.G.As has larger population when compare to others and also a lot of economic activities takes place in these areas.



Fig 1.Study areas in Akwa Ibom state.

B. Sample collection and treatment

The rainwater samples were collected directly from the sky i.e free falling rainwater and the container for collecting them was placed on top of a support which was a distance of 1.5m from the ground in other to avoid unwanted particles from entering the samples collected with the aid of a funnel. The rainwater samples were collected from five Local Government Areas in Akwa Ibom States. The L.G.As includes Uyo, Ikot-Ekpene, Essien Udim, Ikono and Obot Akara. In Uyo L.G.A, a sample of rainwater was collected each from ten(10) different localities which includes Eniong Offot, Obio-Offot, Plaza, Mbribit-Itam, Abiakpo Ikot Essien, Ewet Housing Estate, Itiam Ikot Ebia, Mbereba Obio, Uniuyo permanent site and Ikot Okubo, making a total of ten(10) rainwater samples from Uyo L.G.A. From Ikot Ekpene LGA, samples of rainwater were also collected from seven (7) different localities. In Essien Udim and Obot Akara LGAs, samples of rainwater were collected from five different localities making a total of ten (10) samples from those LGAs. Also three samples of rainwater were collected from three different localities in Ikono LGA. Hence the total number of samples collected from the five (5) LGAs, is thirty (30). At the point of collection, each sample was given a sample code. Also at the

specific location were the sample was collected, the Global positioning system (GPS) was used in taking the coordinate of each sample.

Each sample consists of one liter of rainwater and was acidified at the rate of 10 mL of 11 m H₂SO₄ per liter of rainwater to obtain a PH less than 2 in other to prevent the absorption of the radionuclides into the wall of the container and then sealed in a properly cleaned container for at least one month so as to attain a state of secular radioactive equilibrium before analysis.

C. Measurement of Activity Concentration

The method employed for the measurement of the activity concentration in the samples was the gamma ray spectroscopy. The detector used for the radioactivity measurements is a lead-shielded 76 x 76 mm Sodium Iodide Thallium NaI (TI) doped detector crystal (Model No.802 series, Canberra Inc.) coupled to a Canberra series multichannel Analyzer (MCA) (Model N0.1104) through a preamplifier. It has a resolution full width at half maximum (FWHM) of about 8% at energy of 0.662 Mev. The choice of radionuclides to be detected was predicted based on the fact that the NaI (TI) detector has a modest resolution. The samples were placed symmetrically on top of the detector and measured for a period of 10 hours. The net area under the corresponding peaks in the energy spectrum was computed by subtracting counts due to Compton scattering of higher peaks. From the net area, the activity concentrations in the samples were obtained using [9, 10].

$$C(Bq.L^{-1}) = KC_n \tag{1}$$

Where
$$K = \frac{1}{\varepsilon P_{\gamma} V_{S}}$$
 (2)

C is the activity concentration of the radionuclides in the samples in $Bq.L^{-1}$, C_n is the count rate under the corresponding peak, ϵ is the detector efficiency at the specific Y-ray energy, P_Y is the absolute transition probability of the specific Y-ray energy, and V_s the volume of the sample in litres.

III. RESULTS AND DISCUSSIONS

A. Radionuclide Activity concentrations

The values for the activity concentrations of the radionuclide in the rainwater samples for all the locations are shown in table 1. The activity concentration for $^{238}\mathrm{U}$ in all the locations ranges from 1.2 \pm 0.21 Bq.L⁻¹ – 15.0 \pm 2.92 Bq.L⁻¹ with an average of 5.2 \pm 0.88 Bq.L⁻¹ and the highest activity concentration for $^{238}\mathrm{U}$ in all the locations was in Mbrit-Itam (15.0 \pm 2.92 Bq.L⁻¹) in Uyo LGA and the lowest was in Ikot Enwang (1.2 \pm 0.21 Bq.L⁻¹) in Ikot Ekpene LGA. Also, the activity concentration for $^{232}\mathrm{Th}$ in all the locations ranges from 0.1 \pm 0.00 – 14.2 \pm 1.41 Bq.L⁻¹ with an average of 6.7 \pm 0.69 Bq.L⁻¹. The highest activity concentration for $^{232}\mathrm{Th}$ in all the locations was in Mbribit-Itam (14.2 \pm 1.41 Bq.L⁻¹) in Uyo

LGA and the lowest was in Iton Odoro (0.1 \pm 0.00 Bq.L $^{\text{-}1})$ in Ikono LGA.

Figure 2 is a bar chart representation of the mean activity concentration of 40 K, 238 U and 232 Th in rainwater samples. It shows that Ikot Ekpene and Obot Akara LGAs have the highest activity concentrations of 40 K while Ikono has the lowest. The largest contribution to the overall activity concentration in the rainwater samples from all the locations came mainly from 40 K In fact; this cannot be a surprise because 40 K is a naturally occurring radionuclide which abounds in the earth crust and in human body [11]. The activity concentration due to 232 Th is relatively low in all the samples investigated when compared to that due to 238 U. This is because 238 U is more mobile than 232 Th [11].

TABLE I Mean Activity concentrations of 40 K, 238 U and 232 Th in rain water samples from study areas

	Activity concentrations in Bq.L ⁻¹				
LGA	⁴⁰ K	²³⁸ U	²³² Th		
UYO	28.4 <u>+</u> 2.68	7.5 <u>+</u> 1.47	8.0 <u>+</u> 0.84		
IKOT EKPENE	33.9 <u>+</u> 2.94	33.9 <u>+</u> 3.01	9.3 <u>+</u> 0.96		
ESSIEN EDIM	24.0 <u>+</u> 2.15	5.8 <u>+</u> 0.88	4.3 <u>+</u> 0.44		
OBOT AKARA	33.9 <u>+</u> 3.01	6.0 <u>+</u> 0.96	8.9 <u>+</u> 0.91		
IKONO	18.9 <u>+</u> 1.69	2.6 ± 0.46	2.9 <u>+</u> 0.3		

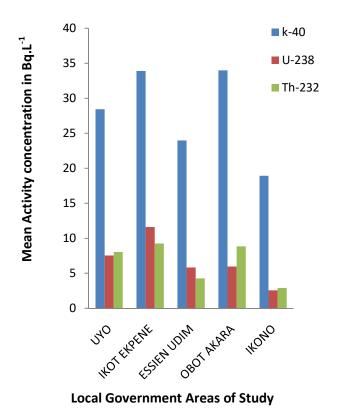


Figure 2: Distribution of the mean activity concentration in the study areas

Radium Equivalent

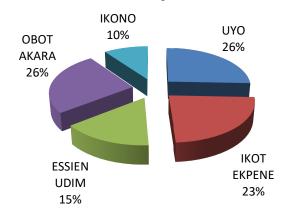


Figure 3: Percentage Distribution of Radium Equivalent In the study areas

Internal Harzard Index

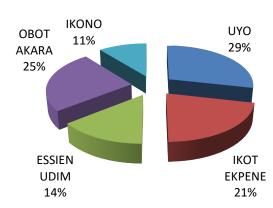


Figure 4: Percentage distribution of internal harzard indices in the study areas.

Internal Harzard Index

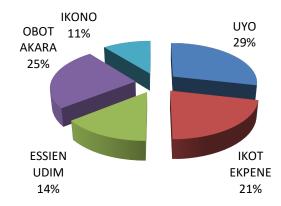


Figure 5: Percentage Distribution of Internal Harzard Indices in the study

Total Annual Effective Dose

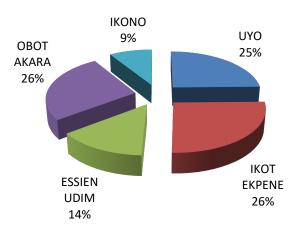


Figure 6: Percentage distribution of Total Annual Effective Dose in the study areas.

B. Annual Effective Dose (AED) and Total Annual Effective Dose

When analyzing the annual effective dose and the total annual effective dose to the human population from natural sources, the dose received by ingestion of long – lived natural radionuclide must be considered [14]. Effective doses resulting from intake of ⁴⁰K, ²³⁸U and ²³²Th may be determined directly from the rainwater since it can be ingested directly by man. Assuming the volume of the daily intake of a drinking water for adult male to be 1L.d⁻¹ [15], the annual effective dose was calculated with the intake of individual radionuclide and ingestion doses co-efficient (Sv. Bq⁻¹) reported by the International Commission on Radiological Protection[14]. The equation for calculating the annual effective dose (AED) per person for a given radionuclide is given below;

$$AED = I_i \times 365 \times D_i \tag{3}$$

While the total Annual Effective Dose (AED) due to all the radionuclides is given by

$$Total AED = \sum I_i \times 365 \times D_i \tag{4}$$

Where I_i is the daily intake of radionuclide i $(Bq.d^{-1})$ D_i is the ingestion dose co-efficient $(Sv.Bq^{-1})$. According to the ICRP 1994, the ingestion dose co-efficient for ^{40}K , ^{238}U , and ^{232}Th in public exposure are $6.2 \times 10^{-9} \ Sv.Bq^{-1}$, $4.5 \times 10^{-8} \ Sv.Bq^{-1}$ and $2.3 \times 10^{-7} \ Sv.Bq^{-1}$ respectively. From figure 2, Obot Akara LGA has the highest total AED while Ikono LGA has the lowest

C. Radium Equivalent Activity (Raea)

The distribution of ²³⁸U, ²³²Th and ⁴⁰K in rainwater is not uniform. Uniformity with respect to exposure to radiation has been defined in terms of radium equivalent (Ra_{ea}) in Bq.L⁻¹ to

compare the specific activity of samples containing different amount of $^{40}\mathrm{K}$, $^{238}\mathrm{U}$ and $^{232}\mathrm{Th}$. It is a weighted sum of activities of $^{40}\mathrm{K}$, $^{238}\mathrm{U}$ and $^{232}\mathrm{Th}$; and it is based on the assumption that 370 Bq.L⁻¹ of $^{238}\mathrm{U}$, 259 Bq.L⁻¹ of $^{232}\mathrm{Th}$ and 4810 Bq.L⁻¹ of $^{40}\mathrm{K}$ produce the same gamma radiation dose rate[16]. To minimize radiation hazards, samples whose Ra_{eq} are greater than 370 Bq.L⁻¹ should not be ingested into the body. The radium equivalent activity Ra_{eq} for each sample was calculated by using the formula below [8].

$$Ra_{eq} = C_U + 1.43C_{Th} + 0.77C_K$$
 (5)

Where C_U , C_{Th} and C_K are the activity concentrations of ^{238}U , ^{232}Th and ^{40}K in $Bq.L^{-1}$ in the rainwater samples respectively. Using equation 5 above, the results show that the estimated Ra_{eq} for all the samples in all the locations ranged from 3.88 $Bq.L^{-1}$ to $38.92\ Bq.L^{-1}$ with an average of $18.06\ Bq.L^{-1}$. These values are smaller than the suggested maximal admissible value of $370\ Bq.L^{-1}$ [17] therefore the samples will not present any significant radiological hazard when ingested.

D. External and Internal Hazard Indices

The External Hazard Index (H_{ex}) and Internal Hazard Index (H_{in}) values was calculated using the equations below [20]. These are hazard indicators that predict the external and internal detriment of natural radiation from ^{40}K , ^{238}U and ^{232}Th .

$$H_{ex} = 0.0027C_U + 0.00386C_{Th} + 0.000208C_K$$
 (6)

$$H_{in} = 0.0054C_{IJ} + 0.00386C_{Th} + 0.000208C_{K}$$
 (7)

Where C_U , C_{Th} and C_K are the activity concentrations of ^{238}U , ^{232}Th and ^{40}K in Bq.L $^{-1}$ respectively.

From table 2, whose values were obtained by using equation 7 above, the external hazard index for the rainwater samples from all the locations ranged from 0.01 to 0.11 with an average of 0.05. Since these values are lower than unity which is the recommended value, therefore, according to the radiation protection 112 report [21] the rain water from these locations is safe without posing any significant radiological threat to the locations. Also, from table 2, whose values were obtained by using equation 8 above, the internal hazard index for the rainwater samples for all the locations ranged from 0.01 to 0.15 with an average of 0.06 which is also less than the recommended value of 1[21].

TABLE II: Estimated mean Ra_{eq} in $Bq.L^{-1}$, H_{ex} , H_{in} and the Total AED in $mSv.y^{-1}$ in the study areas.

LGA	Ra _{eq}	H _{ex}	H_{in}	Total AED
UYO	21.20	0.05	0.08	0.86 <u>+</u> 0.10
IKOT EKPENE	19.39	0.05	0.06	0.90 <u>+</u> 0.09
ESSIEN EDIM	12.64	0.04	0.04	0.49 <u>+</u> 0.05
OBOT AKARA	21.25	0.06	0.07	0.92 <u>+</u> 0.09
IKONO	8.17	0.03	0.03	0.31 <u>+</u> 0.03

IV. CONCLUSION

This work is aimed at assessing the concentration of naturally occurring radionuclide in rain water samples from some selected Areas in Akwa Ibom State, Nigeria. The total annual effective dose, radium equivalent and the radiological indices received by all the locations did not show any significant health impact since it is below the ICRP recommended public exposure limit of 1mSv.y⁻¹ [22].

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