An Assessment of the Radiological Cancer Mortality and Morbidity Risk of Uranium in Rainwater from Selected Areas in Akwa Ibom State, Nigeria

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Abstract
The activity concentrations of uranium in rainwater from selected areas in Akwa Ibom State, Nigeria were measured by the means of a gamma-ray spectrometry using a sodium Iodide Thallium doped NaI (TI) detector. The results obtained were used to calculate human radiological risk over lifetime consumption by the population in the area. The mean activity concentration for Uranium in all the rainwater samples from all the areas was 5.2 ± 0.88 Bq. L^{-1} with a range of 1.2 ± 0.21 to 15.00 ± 2.92 Bq. L^{-1}. The mass concentration was found to range from 46.74 ± 8.46 to 602.86 ± 117.67 µg. L^{-1}. The radiological risks for cancer mortality risk ranged from 4.3 × 10^{-5} to 5.6 × 10^{-4}, while that of the morbidity risk ranged from 6.5 × 10^{-5} to 8.5 × 10^{-4} these values were found to be low. However, the chemical toxicity was found to vary from 1.28 to 16.51 µg.kg^{-1}.day^{-1} while the hazard quotient ranges from 2.13 to 27.53 which are higher than the recommended acceptable safe level by various international organizations. Therefore, it is recommended that regular monitoring of rainwater in these areas should be carried out and also, adequate measures such as providing clean and properly treated water to people living in those areas should be taken.

Keywords: *Activity Concentration, Chemical toxicity, Morbidity risk, Mortality risk, Uranium.*

1 Introduction

In 1896 Becquerel found that a uranium compound affected a photographic plate wrapped in light-proof paper. He called the phenomenon radioactivity (Stabin, 2007). The observation of this phenomenon was confirmed in 1898 by Marie Curie and her husband Pierre Curie. They discovered that a uranium ore, pitchblende, was a more intense source of radioactivity than uranium. They isolated the first concentrated radioactive substances or elements called polonium and radium. Radioactivity is the spontaneous decay of the nucleus of the atom of an unstable element to emit alpha, beta particles or gamma rays or a combination of all of them and energy (Best, L., Rodriguez, G. and Velker, V. 2013). The radioactive decay process proceeds at a well-defined rate characterized by the radionuclides considered. This attribute may be exploited to date materials, both geological and biological in nature, and to determine the kinetics of environmental processes such as water mixing and sediment deposition. Alternatively, artificial

and natural radionuclides with enhanced concentration due to anthropogenic influences may be used as tracers for water masses (Shultis and Faw, 2007). Some radionuclides deserve special consideration due to the threat they may pose as environmental pollutants when they undergo radioactivity, such radionuclides include ^{40}K, ^{238}U and ^{232}Th which are present since the formation of the universe often referred to as naturally occurring radionuclides and materials wherein, they are present. Water is an essential commodity without which there is no life. Radionuclides are present in the air that humans breathe and in food and drinking water (Olomo, 1990) consumed by man and in the ground from which human settlements are built (Olomo, J.B., Tchokossa, P. and Aborisade, C.A. 2003). Enhanced levels of uranium and it 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 contaminants from human activities pass into air, soil and water, and, hence into fish crops and other animals. The input of radionuclides such as ^{40}K to the environment is derived from terrestrial soil and atmospheric diffusion. Considering the high radio toxicity of ^{226}Ra and ^{228}Ra, their presence in water and the associated health risks require particular attention. 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 (UNSCEAR, 1988).

2 Material and Methods

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 10mL of 11m H_2S0_4 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. The rain water samples were analyzed by the means of a gamma-ray spectrometry using a Sodium Iodide Thallium NaI (TI) doped detector. This analysis was done at the National Institute of Radiation Protection and Research (NIRPR), University of Ibadan, Ibadan.

2.1 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 the equations below (Jibiri *et al*., 1999; Awudugba and Tchokossa, 2008).

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

$$\text{Where } K = \frac{1}{\varepsilon P_\gamma V_s} \quad - \quad - \quad - \quad - \quad - \quad - \quad (2)$$

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 ϒ-ray energy, P_γ is the absolute transition probability of the specific ϒ-ray energy, and V_s the volume of the sample in litres.

3 Results and Discussions

3.1 Activity Concentration and Mass Concentration of Rainwater Samples

From table 1 below the activity concentration for ^{238}U in all the locations ranges from 1.2 ± 0.21 $Bq.L^{-1}$ to 15.0 ± 2.92 $Bq.L^{-1}$ with a mean value of 5.2 ± 0.88 $Bq.L^{-1}$ and the highest activity concentration for ^{238}U in all the locations was in Mbrit-Itam (U4) (15.0 ± 2.92 $Bq.L^{-1}$) in Uyo LGA and the lowest was in Ikot Enwang (IE 16) (1.2 ± 0.21 $Bq.L^{-1}$) in Ikot Ekpene LGA. Figures 1 and 2 below also shows the distribution of the mean activity concentration and mass concentration respectively of the rainwater samples in all the study areas.

The values for the activity concentrations of the uranium were converted to the uranium mass concentration ($\mu g.l^{-1}$) using the following conversion factors:

$$1\ Bq.L^{-1} = 27.0\ pCi.L^{-1}, \quad 1\ \mu g.L^{-1} = \frac{pCi.L^{-1}}{0.67} \quad - \quad - \quad - \quad (3)$$

The mass concentration values are also shown in Table 1 below. It ranges from 46.74 ± 8.46 to 602.86 ± 117.67 µg. L^{-1} for all the samples. Various health and environmental protection agencies have recommended a safe limit of uranium in drinking water for human beings. The World Health Organization (WHO, 2003) has recommended 15 µg. L^{-1} of uranium in water as a safe limit for drinking purposes. The United States Environment Protection Agency (USEPA, 1994, 2003) has regulated uranium in public water supplies and has set the value of 30 µg. L^{-1} as the safe limit. Health Canada (1999) sets the standard as 20 µg. L^{-1}. These levels were set to represent the concentration that would not result in any significant risk to health over a lifetime's drinking of water. Comparing uranium concentrations in the various samples obtained in this study with the recommended values of 15 µg. L^{-1} (WHO, 2008) and 30 $\mu g.L^{-1}$ (USEPA, 2003), it could be seen that the mass concentration of all the samples exceeded the safe limits. These high

values may be attributed to the fact that the area is known to have high outdoor gamma radiations.

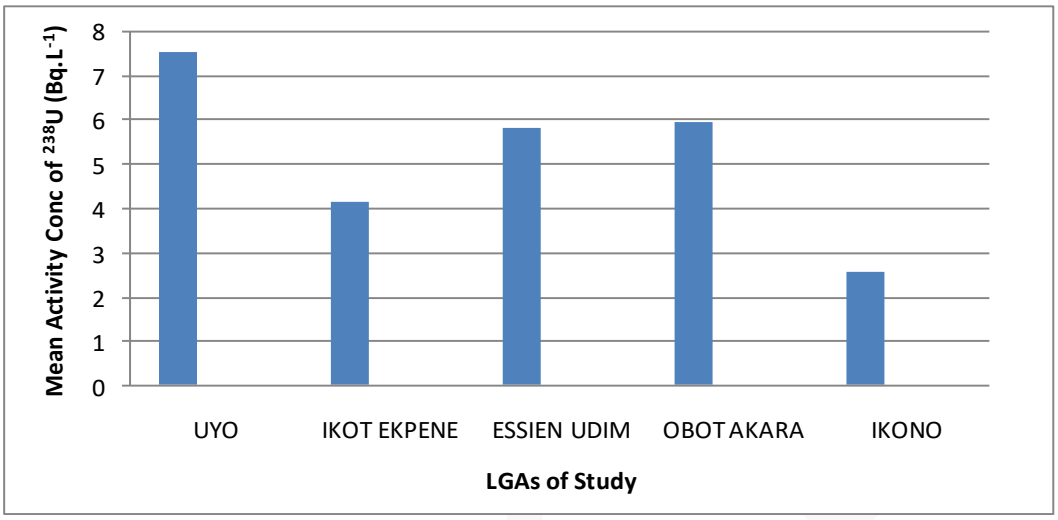


Figure 1: Showing the mean activity concentration of ^{238}U (Bq.L^{-1}) in the rainwater samples from all the study areas

Table 1: Activity concentrations and mass concentration of ^{238}U in rain water samples from all locations

LGA	Samples code	^{238}U (Bq.L^{-1})	^{238}U (pCi.L^{-1})	^{238}U (µg.L^{-1})
UYO	Uy 1	9.5 ± 2.01	255.69 ± 54.27	381.63 ± 81.00
	Uy 2	4.8 ± 0.76	130.95 ± 20.52	195.44 ± 30.63
	Uy 3	BDL	BDL	BDL
	Uy 4	15.0 ± 2.92	403.92 ± 78.84	602.86 ± 117.67
	Uy 5	8.8 ± 1.83	238.14 ± 49.41	355.43 ± 73.74
	Uy 6	11.6 ± 2.01	312.39 ± 54.27	466.25 ± 81.00
	Uy 7	6.2 ± 1.21	167.94 ± 32.67	250.65 ± 48.76
	Uy 8	7.5 ± 1.35	203.04 ± 36.45	303.04 ± 54.40
	Uy 9	1.7 ± 0.32	44.82 ± 8.64	66.89 ± 12.89
	Uy 10	10.3 ± 2.29	279.18 ± 61.83	416.68 ± 92.28
	Mean	7.5 ± 1.47	203.58 ± 39.69	303.85 ± 59.24
IKOT EKPENE	IE 11	3.0 ± 0.45	80.19 ± 12.15	119.68 ± 18.13
	IE 12	3.8 ± 0.78	103.41 ± 21.06	154.34 ± 31.43
	IE 13	4.1 ± 0.74	109.35 ± 19.98	163.21 ± 29.82
	IE 14	BDL	BDL	BDL
	IE 15	2.4 ± 0.46	64.53 ± 12.42	96.31 ± 18.54
	IE 16	1.2 ± 0.21	31.32 ± 5.67	46.74 ± 8.46

	IE 17	10.5 ± 1.64	283.23 ± 44.28	422.73 ± 66.09
	Mean	4.2 ± 0.61	112.05 ± 16.47	167.23 ± 24.58
	EU 18	5.9 ± 0.82	158.22 ± 22.14	236.15 ± 33.04
	EU 19	2.0 ± 0.33	52.65 ± 8.91	78.58 ± 13.30
ESSIEN	EU 20	3.6 ± 0.54	97.74 ± 14.58	145.88 ± 21.76
UDIM	EU 21	11.9 ± 1.82	322.11 ± 49.14	480.76 ± 73.34
	EU 22	BDL	BDL	BDL
	Mean	5.8 ± 0.88	157.68 ± 23.76	235.34 ± 35.46
	OA 23	4.3 ± 0.79	115.29 ± 21.33	172.07 ± 31.83
	OA 24	10.3 ± 1.68	279.18 ± 45.36	416.68 ± 67.70
OBOT	OA 25	4.9 ± 0.65	130.95 ± 17.55	195.44 ± 26.19
AKARA	OA 26	3.1 ± 0.63	83.97 ± 17.01	125.33 ± 25.39
	OA 27	7.3 ± 1.05	197.10 ± 28.35	294.18 ± 42.31
	Mean	6.0 ± 0.96	161.19 ± 25.92	240.58 ± 38.68
	IK 28	2.2 ± 0.40	60.48 ± 10.80	90.27 ± 16.12
IKONO	IK 29	2.8 ± 0.47	74.25 ± 12.69	110.82 ± 18.94
	IK 30	2.7 ± 0.51	72.36 ± 13.77	108.00 ± 20.55
	Mean	2.6 ± 0.46	69.12 ±12.42	103.16 ± 18.54

BDL= BELOW DETECTION LIMIT

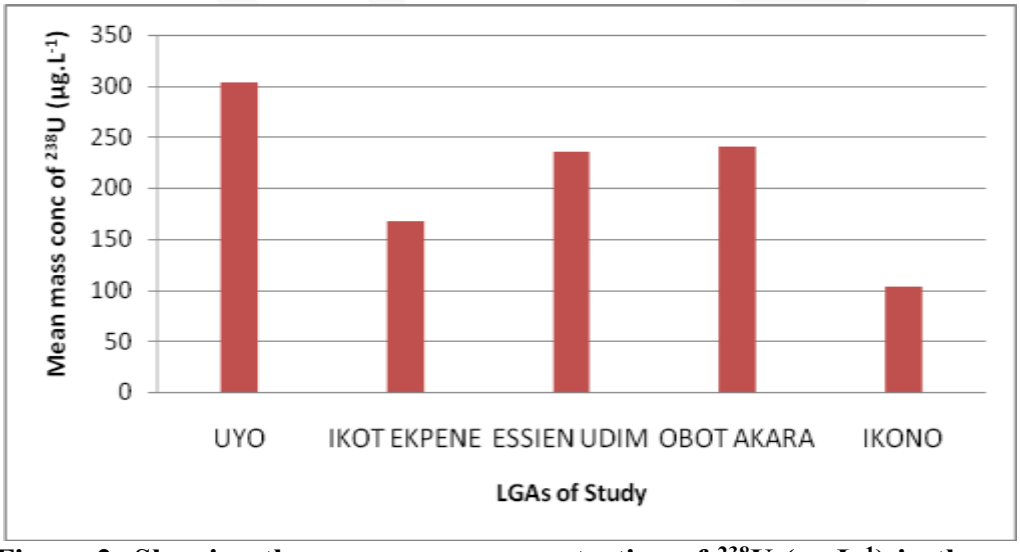


Figure 2: Showing the mean mass concentration of ^{238}U (µg.L^{-1}) in the rainwater samples from all the study areas

3.2 Cancer Mortality and Morbidity risk assessment

The lifetime cancer risks, R, associated with intake of a given radionuclide were estimated from the product of the applicable risk coefficient r and the per capita activity intake I expressed in equation (3).

$$R = r \times I \quad - \quad - \quad - \quad - \quad - \quad (3)$$

According to WHO (2008), the average life expectancy at birth in Nigeria is 45.5 y and, an annual consumption of water for an individual is about 730 litres. This brings the lifetime intake of water to 33,215 litres. The cancer risk coefficients of uranium of 1.13×10^{-9} and 1.73×10^{-9} Bq^{-1} for mortality and morbidity respectively were obtained from the literature (EPA, 1999; UNSCEAR, 2000). Using equation 3 above and these coefficients the cancer mortality and morbidity risks of uranium over lifetime consumption of water were calculated. The results obtained are presented in Table 2 below. From the table 2 below, the cancer mortality risk ranges from 4.3×10^{-5} to 5.6×10^{-4} while for morbidity risk, it ranges between 6.5×10^{-5} and 8.5×10^{-4}. The cancer risk obtained for all the samples is low when compared to the acceptable level for the radiological risk which is 10^{-3} (Ye-shin et al., 2004).

Table 2: The estimated life time cancer mortality and morbidity risk of uranium in the rainwater samples from the study areas.

LGA	Samples code	cancer mortality risk	cancer morbidity risk
UYO	Uy 1	3.5×10^{-4}	5.4×10^{-4}
	Uy 2	1.8×10^{-4}	2.7×10^{-4}
	Uy 3	BDL	BDL
	Uy 4	5.6×10^{-4}	8.5×10^{-4}
	Uy 5	3.3×10^{-4}	5.0×10^{-4}
	Uy 6	4.3×10^{-4}	6.5×10^{-4}
	Uy 7	2.3×10^{-4}	3.5×10^{-4}
	Uy 8	2.8×10^{-4}	4.3×10^{-4}
	Uy 9	6.2×10^{-4}	9.4×10^{-5}
	Uy 10	3.9×10^{-4}	5.9×10^{-4}
IKOT EKPENE	IE 11	1.1×10^{-4}	1.7×10^{-4}
	IE 12	1.4×10^{-4}	2.2×10^{-4}
	IE 13	1.5×10^{-4}	2.3×10^{-4}
	IE 14	BDL	BDL
	IE 15	9.0×10^{-5}	1.3×10^{-4}
	IE 16	4.3×10^{-5}	6.5×10^{-5}
	IE 17	3.9×10^{-4}	5.9×10^{-4}
ESSIEN UDIM	EU 18	2.2×10^{-4}	3.3×10^{-4}
	EU 19	7.3×10^{-5}	1.1×10^{-4}
	EU 20	1.3×10^{-4}	2.0×10^{-4}
	EU 21	4.4×10^{-4}	6.8×10^{-4}
	EU 22	BDL	BDL
	OA 23	1.6×10^{-4}	2.4×10^{-4}
	OA 24	3.9×10^{-4}	5.8×10^{-4}

OBOT	OA 25	1.8×10^{-4}	2.7×10^{-4}
AKARA	OA 26	1.1×10^{-4}	1.7×10^{-4}
	OA 27	2.7×10^{-4}	4.1×10^{-4}
	IK 28	8.4×10^{-5}	1.3×10^{-4}
IKONO	IK 29	1.0×10^{-4}	1.5×10^{-4}
	IK 30	1.0×10^{-4}	1.5×10^{-4}

BDL= BELOW DETECTION LIMIT

3.3 Chemical toxicity risk

The chemical toxicity risk was evaluated using the lifetime average daily dose of uranium through drinking water intake, and compared it with the reference dose (RFD) of 0.6 $\mu g.kg^{-1}.day^{-1}$ (Ye-shin *et al*., 2004) used as a standard criteria for uranium in several foreign organizations and thereby produce a hazard quotient (Equation 4).

$$\text{Hazard quotient} = \frac{LAD}{RFD} \quad - \quad - \quad - \quad - \quad - \quad - \quad - \quad (4)$$

$$\text{Ingestion LADD of drinking water} = \frac{EPC \times IR \times EF}{AT \times BW} \quad - \quad - \quad - \quad - \quad (5)$$

Where LADD, lifetime average daily dose ($\mu g.kg^{-1}.day^{-1}$); EPC is the exposure point concentration ($\mu g.L^{-1}$); IR is the water ingestion rate ($l.day^{-1}$); EF is the exposure frequency (days.year1); ED is the total exposure duration (years); AT is the average time (days) and BW is the body weight (kg). Using therefore, IR = 2 $l.day^{-1}$; EF = 350 days, ED = 45.5 y, AT = 16,607.5 (obtained from 45.5 × 365) and BW = 70 kg (for a standard man) the chemical toxicity risk of uranium over a lifetime consumption was estimated. Figure 3 below shows the distribution of LADDs and hazard quotient values of rainwater samples in all the study areas. The result is presented in Table 3 below. From table 3 below, the exposure dose ranged from 1.28 to 16.51 $\mu g.kg^{-1}.day^{-1}$ while the hazard quotient ranged from 2.13 to 27.53 .By comparing the lifetime average daily dose (LADD) obtained in this study and the reference dose (RFD) (0.6 $\mu g.kg^{-1}.day^{-1}$) which is the acceptable level, the chemical toxicity risk due to uranium in the rainwater samples were all above the RFD. This shows that there are health risks associated with uranium in the rainwater samples which are mainly due to chemical toxicity risk.

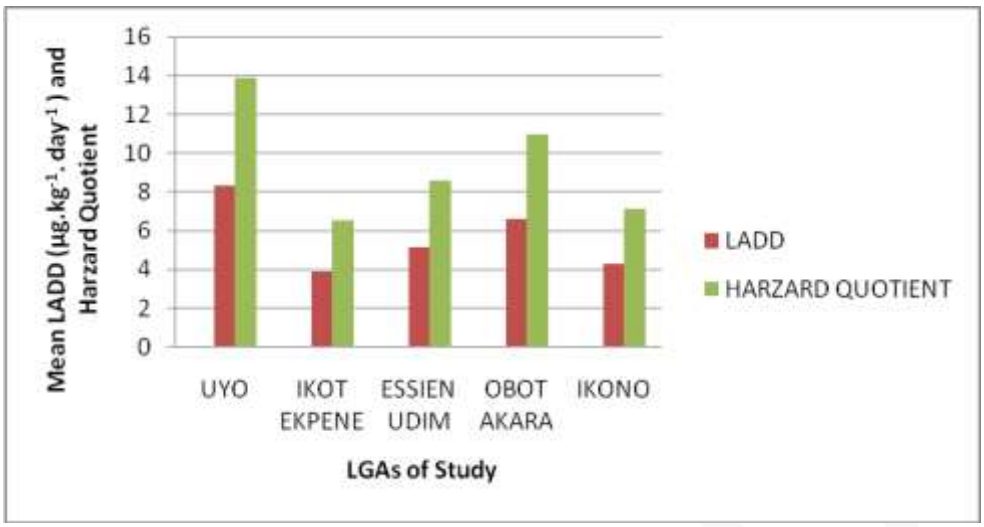


Figure 3: Showing the Mean LADD (µg.kg^{-1}. day^{-1}) and Hazard Quotient of rainwater samples in all the study areas

Table 3: Estimated lifetime average daily dose (LADD) of uranium in the rainwater samples.

LGA	Samples code	LADD (µg.kg^{-1}.day^{-1})	Hazard Quotient
UYO	Uy 1	10.45	17.42
	Uy 2	5.35	8.92
	Uy 3	BDL	BDL
	Uy 4	16.51	27.53
	Uy 5	9.74	16.23
	Uy 6	12.77	21.28
	Uy 7	6.87	11.44
	Uy 8	8.30	13.84
	Uy 9	1.83	3.05
	Uy 10	11.41	19.02
	Mean	8.32	13.87
IKOT EKPENE	IE 11	3.28	5.46
	IE 12	4.23	7.04
	IE 13	4.47	7.45
	IE 14	BDL	BDL
	IE 15	2.64	4.40
	IE 16	1.28	2.13
	IE 17	11.58	19.30
	Mean	4.58	7.63
	EU 18	6.47	10.78

	EU 19	2.15	3.59
ESSIEN	EU 20	4.00	6.66
UDIM	EU 21	13.17	21.95
	EU 22	BDL	BDL
	Mean	6.45	10.74
	OA 23	4.71	7.85
	OA 24	11.41	19.02
OBOT	OA 25	5.35	8.92
AKARA	OA 26	3.43	5.72
	OA 27	8.06	13.43
	Mean	6.59	10.98
	IK 28	2.47	4.12
IKONO	IK 29	3.03	5.06
	IK 30	7.40	12.33
	Mean	4.30	7.17

BDL= BELOW DETECTION LIMIT

Conclusion

The activity concentrations of uranium in rainwater from selected areas in Akwa Ibom State, Nigeria were measured by the means of a gamma-ray spectrometry using a sodium Iodide Thallium doped NaI (TI) detector. The activity concentration for ^{238}U in all the locations ranges from 1.2 $\pm$ 0.21 $Bq.L^{-1}$ to 15.0 $\pm$ 2.92 $Bq.L^{-1}$ with a mean value of 5.2 $\pm$ 0.88 $Bq.L^{-1}$ and the highest activity concentration for ^{238}U in all the locations was in Mbrit-Itam (U4) (15.0 $\pm$ 2.92 $Bq.L^{-1}$) in Uyo LGA and the lowest was in Ikot Enwang (IE 16) (1.2 $\pm$ 0.21 $Bq.L^{-1}$) in Ikot Ekpene LGA. The mass concentration values ranges from 46.74 ± 8.46 to 602.86 ± 117.67 $\mu g.L^{-1}$ for all the samples. Also, the cancer mortality risk ranges from 4.3×10^{-5} to 5.6×10^{-4} while for morbidity risk, it ranges from 6.5×10^{-5} to 8.5×10^{-4}. The cancer risk obtained for all the samples is low when compared to the acceptable level for the radiological risk which is 10^{-3} (Ye-shin et al., 2004). The chemical toxicity risk due to uranium in the rainwater samples were all above the reference dose (RFD) (0.6 $\mu g.kg^{-1}.day^{-1}$) which is the acceptable level. This shows that there are health risks associated with uranium in the rainwater samples which are mainly due to chemical toxicity risk.

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