An Evaluation of the Chemical and Radiological effect of Uranium in Rainwater from Selected Areas in Akwa Ibom State, Nigeria

Ehiwe Isaac Osadebamwen[1], Aniesua Akpan Essiett[2], Edu Inam[3]
[1,2]*Department of Physics, University of Uyo, Akwa Ibom State, Nigeria*
[3]*Department of Chemistry, University of Uyo, Akwa Ibom State, Nigeria*

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 gotten were used to calculate the radiological risk of human 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 obtained ranges from 46.74 ± 8.46 to 602.86 ± 117.67 $\mu g.L^{-1}$. 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. The chemical toxicity was found to vary from 1.28 to 16.51 $\mu g.kg^{-1}.day^{-1}$ while the hazard quotient ranges from 2.13 to 27.53 and these values are above 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, Akwa Ibom, Chemical toxicity, Hazard Quotient, rainwater, Uranium.**

I. INTRODUCTION

In 1896 Becquerel found that a uranium compound affected a photographic plate wrapped in light-proof paper. He called the phenomenon radioactivit[y9]. 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[2]. 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[8]. 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[7] consumed by man and in the ground from which human settlements are built[6]. 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[10].

II. 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 doped detector. This analysis was done at the National Institute of Radiation Protection and Research (NIRPR), University of Ibadan, Ibadan.

2.1 Activity Concentration Measurement

The method used for the activity concentration measurement in the samples was the gamma ray spectroscopy. The detector used for the measurements of the radioactivity is a lead-shielded 76mm x 76 mm NaI (TI) doped detector crystal which is coupled to a Canberra series multichannel Analyzer (Model N0.1104) through a pre-amplifier. It has a resolution full width at half maximum of about eight percent 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 on top of the detector in a symmetrical manner and then measured for a period of 10 hours and 30mins. From the net area, the activity concentrations in the samples were obtained using the equations below[1,5].

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

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 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.

III. RESULTS AND DISCUSSIONS

Figures 1 shows the distribution of the mean activity concentration of the rainwater samples in all the study areas. Figure 2 which is a contour map for ^{238}U shows how ^{238}U is distributed in all the study area.

The activity concentrations values of the uranium were converted to its mass concentration (µg.l^{-1}) using the following conversion factors:

1 $Bq.L^{-1}$ = 27.0 $pCi.L^{-1}$,

$$1\ \mu g.L^{-1} = \frac{pCi.L^{-1}}{0.67} \qquad (3)$$

The mass concentration values are shown in Table 1. The values ranges from 46.74 ± 8.46 to 602.86 ± 117.67 µg.L^{-1} for the samples. A good number of health and environmental protection agencies have recommended a particular safe limit of uranium in drinking water for humans. The World Health Organization (WHO)[14] recommended 5 µg.L^{-1} of uranium in water as a safe limit for drinking purposes. The United States Environment Protection [12,13] as part of its funtion regulates uranium in public water supplies and as a result set 30 µg.L^{-1} as the safe limit.

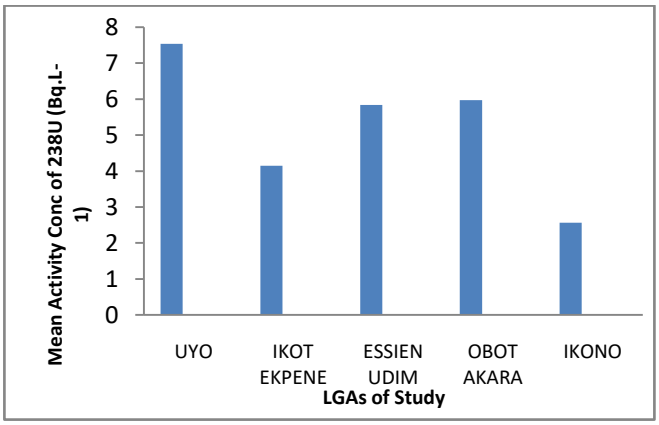

Figure-1: Showing the mean activity concentration of ^{238}U (Bq.L^{-1}) in the rainwater samples from all the study area

Table 1: Mean Activity concentrations and mean mass concentration of ^{238}U in rain water samples from the study areas.

LGA	238U (Bq.L-1)	238U (pCi.L-1)	238U (µg.L-1)
Uyo	7.5 + 1.47	203.58 + 39.69	303.85 + 59.24
kot Ekpene	4.2 + 0.61	112.05 + 16.47	167.23 + 24.58
Essien Udim	5.8 + 0.88	157.68 + 23.76	235.34 + 35.46
Obot Akara	6.0 + 0.96	161.19 + 25.92	240.58 + 38.68
Ikono	2.6 + 0.46	69.12 +12.42	103.16 + 18.54

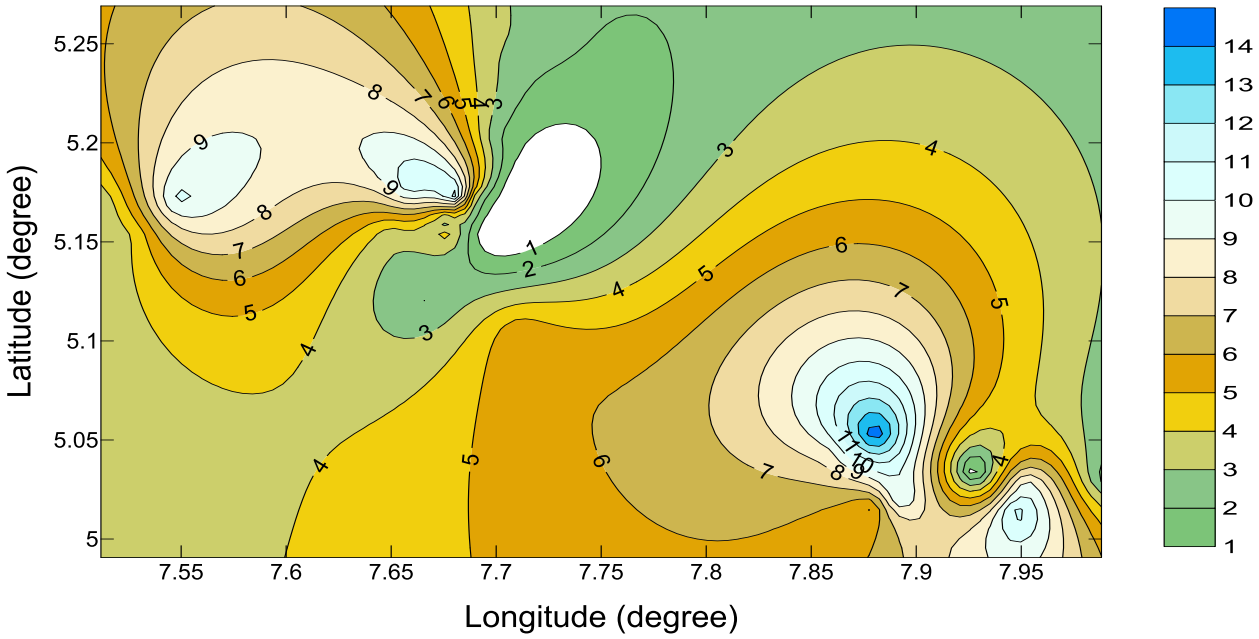

Figure-2: Contour diagram for the activity concentration of ^{238}U in Bq.L^{-1} for the collected samples**.**

3.1 Cancer Mortality and Morbidity Health risk assessment

The lifetime cancer health risks, R, due to the intake of a given radionuclide was estimated by multiplying the risk coefficient applicable, r and the activity intake I per capita shown in equation (4).

$$R = r \times I \qquad (4)$$

According to WHO [15], the average life expectancy at birth in Nigeria is 45.5 y and, the consumption of water annually for an individual is about 7.30 x 10^2 litres. This brings the lifetime intake of water to 3.3215 x 10^4 litres. The coefficients cancer risk of uranium for mortality and morbidity respectively used for the estimation were 1.130 × 10^{-9} Bq^{-1} and 1.730 × 10^{-9} Bq^{-1} as obtained from the literature[3,11]. Using equation 4 and these coefficients the cancer mortality and morbidity risks of uranium over lifetime consumption of water were calculated. results obtained are presented in Table 2. The cancer risk obtained for the samples is found to be low when compared to the acceptable level for the radiological risk which is 0.001 [16]

Table 2: The mean life time cancer mortality and morbidity health risk due to uranium in the rainwater samples from the Study areas.

LGA	mortality risk	morbidity risk
UYO	4.3 x 10-4	6.5 x 10-4
IKOT EKPENE	4.3 x 10-5	6.5 x 10-5
ESSIEN UDIM	4.4 x 10-4	6.8 x 10-4
OBOT AKARA	1.8 x 10-4	2.7 x 10-4
IKONO	1.0 x 10-4	1.5 x 10-4

3.2 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 µg.kg^{-1}.day^{-1} [16] used as a standard criteria for uranium in several foreign organizations and thereby produce a hazard quotient (Equation 5).

$$\text{Hazard quotient} = \frac{LAD}{RFD} \qquad (5)$$

$$\text{Ingestion LADD} = \frac{EPC \times IR \times EF}{AT \times BW} \qquad (6)$$

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

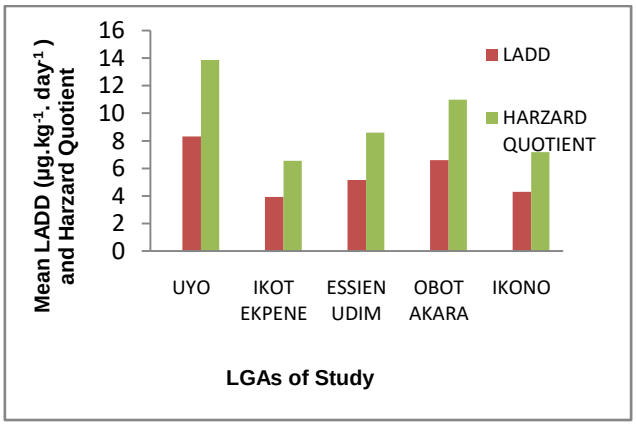

Figure-3: Showing the Mean LADD (µg.kg^{-1}. day^{-1}) and Hazard Quotient of rainwater samples in all the study areas.

Table-3: Estimated mean LADD due to uranium in the rainwater samples from all study areas

LGA	LADD (µg.kg-1.day-1)	Hazard Quotient
Uyo	8.32	13.87
Ikot Ekpene	4.58	7.63
Essien Udim	6.45	10.74
Obot Akara	6.59	10.98
Ikono	4.30	7.17

IV. 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 (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. The mass concentration values ranges from 46.74 ± 8.46 to 602.86 ± 117.67 µg.L^{-1} for all the samples. The chemical toxicity risk due to uranium in the rainwater samples were all above the reference dose (RFD) (0.6 µg.kg^{-1}.day^{-1}) which is the acceptable level. This shows rainwater samples which are mainly due to chemical toxicity risk. that there are health risks associated with uranium in the

REFERENCES

[1] Awudugba, A.O. and Tchokossa, P. (2008). Assessment of radionuclide concentration in water supply from bore-holes in Ogbomoso land, Western Nigeria: ind and Buil.Environ., 17(2):183-186.

[2] Best, L., Rodriguez, G. and Velker, V. (2013). Radiation Oncology. Primer and review. Demos Medical Publishing. ISBN 978-1-62070-004-4.

[3] Environmental Protection Agency (EPA) (1999). Cancer risk coefficients for Environmental exposure to radionuclides. United State Environmental Protection Agency. Federal Guidance Report No -13 (EPA. 402 R-99-001).

[4] Health Canada (1999). Summary of guidelines for Canadian drinking water quality: Prepared by federal provincial subcommittee on drinking water of the federal provincial-territorial committee on environmental and occupational health.

[5] Jibiri, N.N., Mabawonku, O.A., Oridate, A.A. and Ujiagbedion, C.A.(1999). Natural radionuclide Concentration levels in soil and water around a cement factory at Ewekoro, Ogun, Nigeria: Nig. J.phy; 11:12-16.

[6] Olomo, J.B., Tchokossa, P. and Aborisade, C.A. (2003). "Study of Radiation Protection guidelines in the use of Building Materials for Urban Dwellings in South- West Nigeria,"Nigeria journal of Physics, Vol. 15, No. 1, pp. 7-13.

[7] Olomo, J.B. (1990). "The Natural Radioactivity in Some Nigerians Foodstuffs," Nuclear Instruments methods in Physics Research A, Vol. 299, No. 1-3, pp. 666-669. doi: 10-1016/0168-9002(90)90866-5.

[8] Shultis, J. K. and Faw, R. E. (2007). Fundamentals of Nuclear Science and Engineering (2nd edition). CRC Pres. P.175. ISBN 978-1-4398-9408-8.

[9] Stabin, M. G. (2007). Radiation Protection and Dosimetry: An introduction to Health Physics. Springer. doi: 10-1007/978-0-387-49983-3. ISBN 978-0-387-49982-6.

[10] United Nation Scientific Committee on the Effects of Atomic Radiation (UNSCEAR), (1988). "sources, effects and risks of Ionizing Radiation", New York.

[11] United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) (2000). Sources, effects and risks of ionizing radiation. Report to the General Assembly. ISBN 92-1-142238-8, New York

[12] United States Environmental Protection Agency (2003). Current Drinking Water Standards, Ground water and drinking water protection agency, pp. 1–12.

[13] United State Environmental Protection Agency (USEPA) (1994). Comparison of Health Risks Arising From the Radiological and Chemical Toxicity of Uranium, Appendix F of Radiation Site Cleanup Regulations: Technical Support Document For The Development Of Radionuclide Cleanup Levels For Soil, Review Draft, EPA 402-R-96- 011 A.

[14] World Health Organization (WHO) (2003). Guidelines for drinking water quality Health criteria and other supporting information, Geneva, 2nd edition., 2: 367–370.

[15] World Health Organization (WHO) (2008). Meeting the MDG drinking water and sanitation target: the urban and rural challenge of the decade. WHO Library Cataloguing-in-Publication Data.

[16] Ye-shin K., Hoa-sung P., Jin-yong K., Sun-ku P., Byong-wook C., Ig-hwan S and Dong-Chun S (2004). Health risk assessment for uranium in Korean groundwater. J. Environ. Radioactivity, pp. 77-85