

Investigation of Radionuclide Contents and Radiological Implications of Groundwater in Mining Areas of Plateau State, North Central of Nigeria

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Abstract

The investigation of the Radionuclide Concentrations in Water from Mining Areas of Plateau State and their Radiological Implications was carried out. Twenty five samples were drawn randomly in two litre of plastic containers and acidified with 0.2ml of concentrated nitric acid for preservation. The samples were analysed using ICP-MS technique to determine trace to ultra-trace concentrations of radioactive elements in the samples. The results showed that the concentration of ^{238}U ranged from 7.41×10^{-4} to 2.09 Bq/kg with an average value of $1.1 \times 10^{-1} \text{ Bq/kg}$, ^{232}Th ranged from 2.05×10^{-4} to $9.89 \times 10^{-4} \text{ Bq/kg}$ with a mean value of $2.51 \times 10^{-4} \text{ Bq/kg}$, and ^{40}K ranged from 3.34×10^2 to $7.67 \times 10^3 \text{ Bq/kg}$ with a mean value of $1.256 \times 10^3 \text{ Bq/kg}$. The results of the evaluation of Ra_{eq} , AEDE, AGDE and ELCR were calculated, it was below the world average of 370 Bq/kg , 0.1 mSv/yr , $300 \text{ } \mu\text{Sv/yr}$ and $0.29 \times 10^{-3} \text{ mSv/yr}$ in some locations and exceed the allowable limit in some of the areas respectively. The results were found to be comparable to reported data internationally for some locations and above the permissible limit in some areas. Hence, the investigation of groundwater can be considered to have some radiological hazard indices of causing cancer and toxicity of the kidney over a long period of exposure.

Key words: Investigation, groundwater, radiological, Radionuclide, ICP-MS technique.

1. Introduction

We are surrounded with radiation in every facet of our lives being in the natural radioactive area. Naturally occurring radioactive materials are present in its crust, the floors and walls of homes, schools, or offices and in the food we eat and drinks [1]. Researchers are continually exploring ways of measuring the radiation levels and quantify the risks and doses affecting people, animals, plants and all kinds of life as a result of the geology of the area or the inappropriate mining by artisans of all age brackets.

Industrial activities such as extraction and processing of minerals may cause the incorporation of radionuclides into the hydrosphere through surface or ground water [2]. Naturally occurring radionuclides of thorium and uranium are the significant contributors of ingestion dose and are present in the biotic system of plants, animals, soil, water, air and thus in food [3]. Water contains a number of

both alpha (such as ^{238}U , ^{226}Ra and ^{210}Po) and beta emitters (such as ^{40}K , ^{228}Ra and ^{210}Pb). Natural radioisotopes as ^{40}K and the nuclides from the ^{238}U and ^{232}Th series are the greatest source of internal and external exposure in human beings. Water is one widely used natural resources related directly to the survival of all living organisms and is not completely free of radionuclide which is an atom with an unstable nucleus which, to become more stable, emits energy in the form of rays or high speed particles. The ionizing radiation effects of the radionuclide when inhaled or ingested can create ions by displacing electrons in the body. This gives the relevance of the investigation of the radiation levels and the radiological implications. The investigation of activity concentration of natural radionuclides in groundwater is mandatory for regular surveillance of radioactivity level in groundwater sources.

The study seeks to explore the activity levels of natural radionuclides (^{238}U , ^{232}Th , ^{40}K) in ground-

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water (boreholes and wells) from chosen regions of Plateau State North Central Nigeria, covered by Naraguta topographical sheet 168 where comprehensive tin mining and radiological risk assessment operations were conducted and processed.

1.1 Geology of the Study Area.

The geology of the Jos Plateau State of Nigeria (Naraguta sheet 168) is made-up of the Precambrian Basement migmatite-gneiss-quartzite complex which underlies about half of the entire State and in some places has been intruded by Precambrian to the late Paleozoic Pan-African granite (Older Granite), diorite, charnockite etc. Intrusive into these Basement Complex rocks are the Jurassic androgenic alkali Younger Granites. In association to the Younger Granites are volcanic rocks such as basalts and rhyolites that overly or cross-cut this formation as well as the Basement rocks. It is thought that these volcanic rocks were created during the early Cenozoic (Tertiary) "Older Basalts" and the "Newer Basalts" [3]

2. Materials and Methods

The raw data collection was carried out during the dry season when dilution is lowest and free from other sources that might find their way into the groundwater. Their depths to static water level vary approximately from 5-20 metres. Water (groundwater) was collected from mining areas

of Plateau state and its environs covering Naraguta Topographical sheet 168. The area is bounded between longitude $8^{\circ}31'1.00''E$ and latitude $9^{\circ}34'00.00''N$ to $9^{\circ}55'00.00''N$ as shown in Figure 1.

2.1 Sample Collection and Preparation.

In order to measure the radionuclide in ground waters, 25 water samples were collected randomly from different locations. The choice of sampling locations was based on population density and accessibility.

The sample container was rinsed 2-3 times with deionized water/water samples being collected to minimize contamination from original content of the sample container. The amount of water collected was such that an air space of about 1% of container capacity was left for thermal expansion. About 0.2ml of concentrated nitric acid were added to the samples (for preservation) immediately after collection. The sample was tightly covered and taken to ACME laboratory Vancour Canada for analyses

2.2 Description of Counting Equipment

ICP-MS (inductively coupled plasma-mass-spectrometry) is a technique to determine low-concentrations (range: ppb = parts per billion = $\mu\text{g/l}$) and ultra-low concentrations of elements (range: ppt = parts per trillion = ng/l). Atomic elements are lead through a plasma source where they become ionized. Then, these ions are sorted on account of their mass. The ICP-

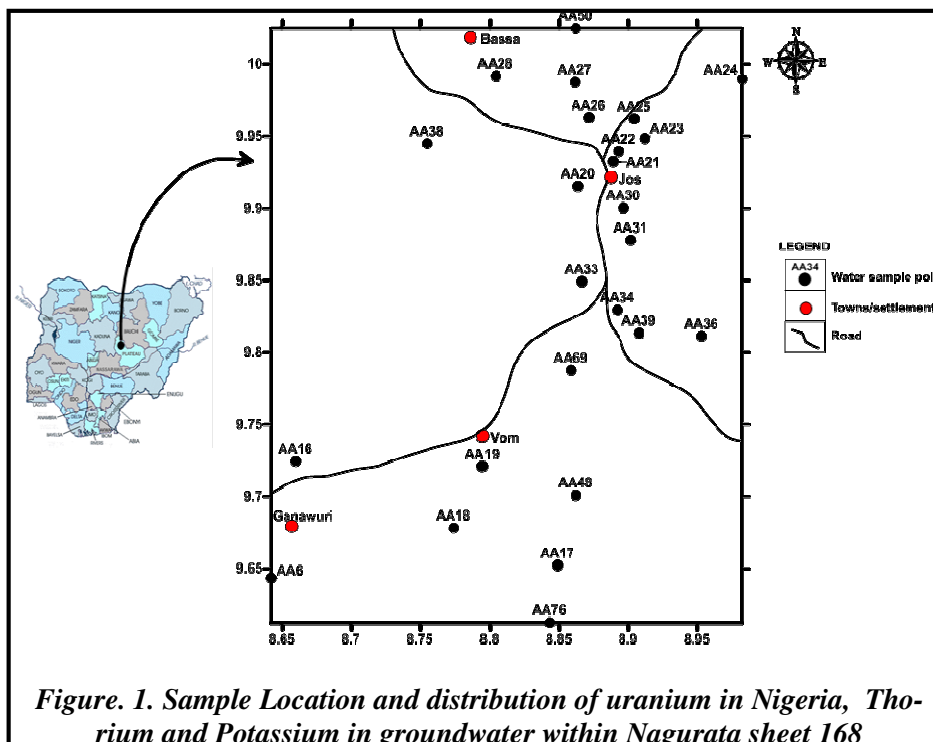


Figure. 1. Sample Location and distribution of uranium in Nigeria, Thorium and Potassium in groundwater within Nagurata sheet 168

MS has an extremely low detection limits, a large linear range and Possibilities to detect isotope composition of elements. The ICP –MS sampling processing analyses the sample which is introduced as a liquid using a nebulizer and spray chamber. The nebulizer uses supersonic expansion of gas to turn the liquid into a fine mist, and the spray chamber then removes any droplets that are too large to be processed in the plasma. This occurs at the sample interface of the instrument. Liquid samples are typically digested and then reconstituted in an aqueous matrix to stabilize elements as an ionic solution. The matrix typically contains 2% nitric acid, and may have 0.5% hydrochloric acid added to stabilize certain elements. The final composition of any matrix is highly dependent on the nature of the analytes being measured. The results are in ppb/ppm which is converted to Bq/kg according to [4].

2.3 Radiological Risk Assembly

Standard radiation indices are used to evaluate the effects of radiation on the health of people exposed to radiation and the environment. The indices evaluated included:

2.3.1 Radium Equivalent Activity (*Raeq*)

This is used to assess the hazards associated with materials that contain ^{226}Ra , ^{232}Th and ^{40}K in Bq kg^{-1} which is, calculated on the assumption that 370 Bq kg^{-1} of ^{226}Ra or $^{259}\text{Bq kg}^{-1}$ of ^{232}Th or 4810 Bq kg^{-1} of ^{40}K produce the same gamma dose rate [6] The *Raeq* of the sample in Bq kg^{-1} was achieved using the equation [7].

$$R_{aeq} = (A_{\text{Th}} \times 1.43) + (A_{\text{K}} \times 0.0077) + (A_{\text{R}}) \quad (1)$$

where A = Activity concentrations of ^{232}Th , ^{40}K , and ^{226}R , respectively.

The radium equivalent is the most useful guideline for regulating safety standards on radiation protection for the general public [5].

2.3.2 Annual Effective Dose Equivalent (*AEDE*)

The annual effective dose is the measure of the can-

cer risk to a whole organism due to ionizing radiation delivered non-uniformly to part(s) of its body. It is not intended as a measure of deterministic or other effects of radiation although it is used to estimate inherited effects.

The equation used to calculate the dose is

$$DR = A \times IR \times ID \quad (2) \quad [8]$$

where DR= the effective equivalent Dose

A= is the activity (Bq/l)

IR= intake of water per person per year (2L/day) = 730litres

ID= is the ingestion dose equivalent factor of 3.58×10^{-7} mSv/yr

And standard AEDE is given as 0.1 mSv/yr

2.3.3 Excess Life Cancer Risks (*ELCR*)

This deals with the probability of developing cancer over a lifetime at a given exposure level, considering 70 years as the average duration of life for human being. It is given as [9]

$$ELCR = AEDE \times DL \times RF \quad (3)$$

where AEDE = Annual Effective Dose Equivalent

DL = Average Duration of life (estimated to be 70 years)

RF = the risk factor (Sv^{-1}) i.e fatal cancer risk per Sievert for stochastic effects, ICRP 60 use value of 0.05 for the public [9,10 & 11]

2.3.4 Annual Gonadal Dose Equivalent

The annual gonadal dose equivalent is a measure of the genetic significance of the yearly dose received by the populations reproductive organs [12]. Organs with rapidly dividing cells such as gonads, the active bone marrow and bone surface cells are considered as organs of interest by the United Nations Scientific Committee on the Effects of Atomic Radiation [13]. The annual gonadal dose equivalent (AGDE) due to the specific activities of ^{226}Ra , ^{232}Th , and ^{40}K was calculated using the following formula [14]

$$AGDE (\mu\text{Sv yr}^{-1}) = 3.09A_{\text{Ra}} + 4.18A_{\text{Th}} + 0.314A_{\text{K}} \quad (4)$$

Table 1: The Activity Concentrations of Uranium, Thorium, Potassium, Radium equivalent and External hazard index in Ground Water Supply in Jos Plateau State.

		METHOD	2C	2C	2C	2C	
		ANALYTE	DILUTION				
		UNIT	NONE	Bq/Kg	Bq/Kg	Bq/Kg	
		MDL	1	2.47 $\times 10^{-4}$	2.05	1.3 $\times 10^{-2}$	
S/NO	SAMPLE ID	SOURCE		²³⁸ U	²³² Th	⁴⁰ K	(Ra _{eq}) Bqkg ⁻¹
1	AA6	GW	1	0.000864	0.000205	720.57	55.49
2	AA16	GW	1	0.000741	0.000287	572.83	44.11
3	AA17	GW	1	0.002720	0.000205	1083.45	83.43
4	AA18	GW	1	0.000864	0.000205	689.47	53.09
5	AA19	GW	1	0.003460	0.000205	694.65	53.49
6	AA20	GW	1	0.013100	0.000205	7674.89	590.98
7	AA21	GW	1	0.073100	0.000205	443.23	34.20
8	AA22	GW	1	0.001230	0.000989	1682.20	129.53
9	AA23	GW	1	0.005190	0.000205	624.67	48.11
10	AA24	GW	1	0.001850	0.000205	378.43	29.14
11	AA25	GW	1	0.023000	0.000205	469.15	36.15
12	AA26	GW	1	0.018600	0.000205	388.80	29.96
13	AA27	GW	1	0.045700	0.000205	1264.89	97.44
14	AA28	GW	1	0.006300	0.000205	759.45	58.48
15	AA30	GW	1	0.005060	0.000205	1619.10	124.68
16	AA31	GW	1	0.011900	0.000205	808.70	62.28
17	AA33	GW	1	0.004820	0.000205	1461.88	112.57
18	AA34	GW	1	0.218000	0.000205	2249.85	173.46
19	AA36	GW	1	0.000741	0.000205	334.67	25.77
20	AA38	GW	1	0.001360	0.000205	435.45	33.53
21	AA39	GW	1	0.001980	0.000205	793.15	61.07
22	AA48	GW	1	0.006920	0.000205	435.45	33.54
23	AA50	GW	1	0.231000	0.000205	1866.23	143.93
24	AA69	GW	1	2.083425	0.000492	3514.74	292.72
25	AA76	GW	1	0.005800	0.000205	479.52	36.93
			Mean	0.110709	0.000251	1257.82	97.76
			maximum	2.088425	0.000989	7674.89	590.98
			minimum	0.000741	0.000205	334.67	25.77


3. Results

The results of activity concentration and risk assessment for 25 groundwater collected at different locations around the Naraguta sheet 168 are summarised in Tables 1 and 2. The three most common primordial radionuclides determined in the study areas were ^{238}U , ^{232}Th , ^{40}K

4. Discussion

Table 1: The results of activity concentration of the radionuclide in the Nagurata sheet 168 in Jos and its environs ranged from 7.41×10^{-4} to 2.09×10^{-4} Bq/kg, 2.05×10^{-4} to 9.89×10^{-4} Bq/kg and 3.34×10^2 to 7.67×10^3 Bq/kg with an average value of 1.1×10^{-1} Bq/kg, 2.51×10^{-4} Bq/kg, and 1.256×10^{-4} Bq/kg respectively which is within the allowable limits

Table 2: The Estimated Radiation Risk Parameters in Groundwater Supply in Mining Areas of Jos Plateau State.

SAM- PLE ID	^{238}U Activ- ity (Bq/kg)	Annual Effec- tive Dose Equiva- lent mSv/yr	ELCR	^{232}Th Activ- ity (Bq/kg)	Annual Effective Dose Equivalent mSv/yr	ELCR	^{40}K Activ- ity (Bq/kg)	Annual Effective Dose Equiva- lent mSv/yr	ELC R	AGDE 
AA6	0.0008 64	2.26 $\times 10^{-4}$	7.91 $\times 10^{-4}$	0.0002 05	3.36 $\times 10^{-3}$	1.88 $\times 10^{-4}$	720.57	188.31	659.1 0	226.26
AA16	0.0007 41	1.94 $\times 10^{-4}$	6.79 $\times 10^{-4}$	0.0002 87	7.50 $\times 10^{-3}$	2.63 $\times 10^{-4}$	572.83	149.70	523.9 6	179.89
AA17	0.0027 20	7.11 $\times 10^{-3}$	2.49 $\times 10^{-4}$	0.0002 05	3.36 $\times 10^{-3}$	1.88 $\times 10^{-4}$	1083.4 5	283.15	991.0 2	340.21
AA18	0.0008 64	2.26 $\times 10^{-4}$	7.91 $\times 10^{-4}$	0.0002 05	3.36 $\times 10^{-3}$	1.88 $\times 10^{-4}$	689.47	180.19	630.6 5	216.50
AA19	0.0034 60	9.04 $\times 10^{-4}$	3.16 $\times 10^{-3}$	0.0002 05	3.36 $\times 10^{-3}$	1.88 $\times 10^{-4}$	694.65	181.54	635.3 9	218.13
AA20	0.0131 00	3.42 $\times 10^{-3}$	1.20 $\times 10^{-3}$	0.0002 05	3.36 $\times 10^{-3}$	1.88 $\times 10^{-4}$	7674.8 9	2005.76	7020. 16	2409.96
AA21	0.0731 00	1.91 $\times 10^{-4}$	6.09 $\times 10^{-4}$	0.0002 05	3.36 $\times 10^{-3}$	1.88 $\times 10^{-4}$	443.23	115.88	405.4 2	139.40
AA22	0.0012 30	3.21 $\times 10^{-4}$	1.12 $\times 10^{-3}$	0.0009 89	2.59 $\times 10^{-4}$	2.59 $\times 10^{-4}$	1682.2 0	439.63	1538. 69	528.22
AA23	0.0051 90	1.36 $\times 10^{-3}$	4.76 $\times 10^{-3}$	0.0002 05	3.36 $\times 10^{-3}$	1.88 $\times 10^{-4}$	624.67	163.25	571.3 8	196.16
AA24	0.0018 50	4.83 $\times 10^{-4}$	1.69 $\times 10^{-3}$	0.0002 05	3.36 $\times 10^{-3}$	1.88 $\times 10^{-4}$	378.43	98.10	346.1 5	118.83
AA25	0.0230 00	6.01 $\times 10^{-3}$	2.10 $\times 10^{-3}$	0.0002 05	3.36 $\times 10^{-3}$	1.88 $\times 10^{-4}$	469.15	122.61	429.1 3	147.39

AA26	0.018600	4.86	1.70		3.36 $\times 10^{-3}$	1.88	388.80	101.61	355.63	122.14
				0.000205						
AA27	0.045700	1.19	4.17		3.36 $\times 10^{-3}$	1.88	1264.89	330.57	1156.9	397.32
				0.000205					8	
AA28	0.006300	1.65	5.78		3.36 $\times 10^{-3}$	1.88	759.45	198.47	694.66	238.49
				0.000205						
AA30	0.005060	1.32	4.62		3.36 $\times 10^{-3}$	1.88	1619.10	423.14	1480.9	508.41
				0.000205					7	
AA31	0.011900	3.11	1.09		3.36 $\times 10^{-3}$	1.88	808.70	211.35	739.71	253.97
				0.000205						
AA33	0.004820	1.26	4.41		3.36 $\times 10^{-3}$	1.88	1461.88	382.05	1337.1	459.05
				0.000205					7	
AA34	0.218000	5.7	1.20		3.36 $\times 10^{-3}$	1.88	2249.85	587.98	2057.9	707.13
				0.000205					2	
AA36	0.000741	1.94	6.79		3.36 $\times 10^{-3}$	1.88	334.67	87.46	306.12	105.09
				0.000205						
AA38	0.001360	3.55	1.24		3.36 $\times 10^{-3}$	1.88	435.45	113.800	398.30	136.74
				0.000205						
AA39	0.001980	5.17	1.81		3.36 $\times 10^{-3}$	1.88	793.15	207.28	725.49	249.06
				0.000205						
AA48	0.006920	1.81	6.34		3.36 $\times 10^{-3}$	1.88	435.45	113.80	398.30	136.75
				0.000205						
AA50	0.231000	6.02	2.11		3.36 $\times 10^{-3}$	1.88	1866.23	487.72	1707.0	586.71
				0.000205					2	
AA69	2.083425	5.44	1.904			4.50	3514.74	918.54	3214.9	1110.09
				0.000492	1.29 $\times 10^{-4}$				0	
AA76	0.005800	1.52	5.32			1.88	479.52	125.32	438.61	150.59
				0.000205	3.36 $\times 10^{-3}$					
Mean	0.11071	0.0288153	0.092971				1257.82	328.6884	1156.5	395.300
				0.00025	0.000314	0.000204			1	
Max	2.08843	0.5440000	1.904				7674.89	2005.76	7020.1	2409.96
				0.00099	0.000336	0.00045			6	
Min	0.00074	0.0000711	0.000249				334.67	87.46	306.12	105.09
				0.00021	0.000075	0.000188				

for some locations and some locations exceed the permissible level. Table 2: The value for radium equivalent activity for groundwater ranged from

25.77 to 590.98 Bq/kg with an average of 97.7632 Bq/kg which is lower than the permissible value of 370 Bq/kg but sample AA20 is far above the

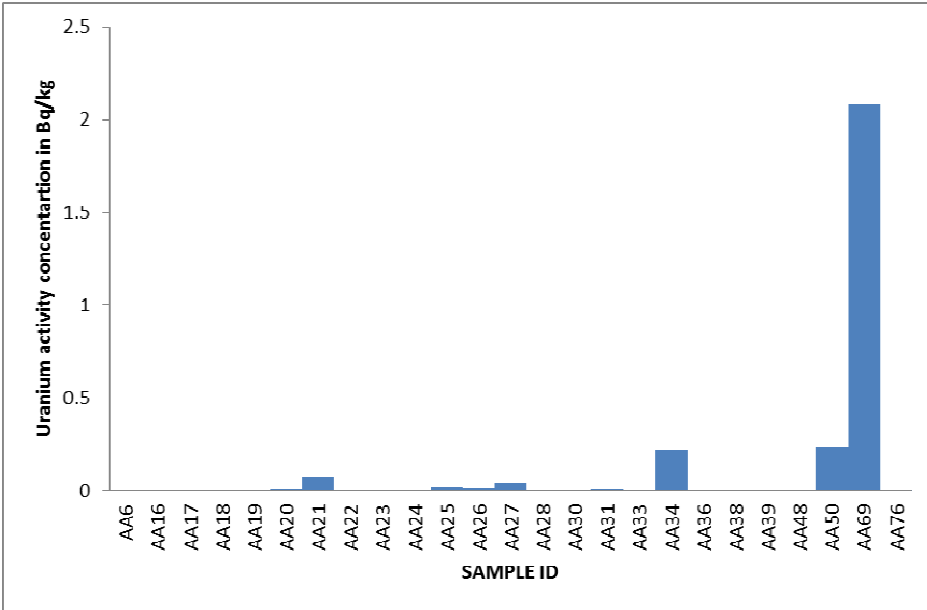


Figure 2: Graph of uranium activity concentration against sample location for ground water.

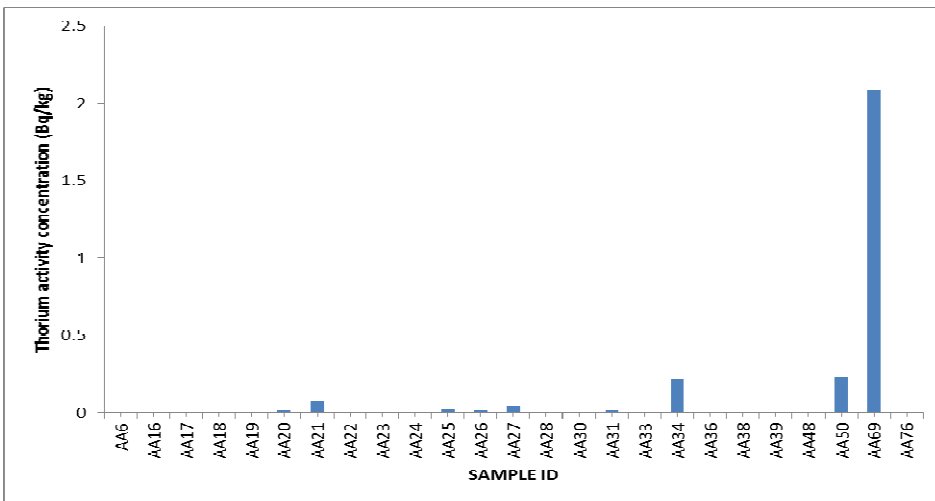


Figure 3: Graph of Thorium activity concentration against sample location

ELCR standard of 0.29 mSv/yr, [14]. Most of the samples are above the standard as recommended. The Gonads Dose Equivalent (AGDE) was calculated and the results showed 36% exceeding the limit of 300 μ Sv/y as recommended by the World average and Comparing the results with previous work of [14,15,16] on the Naragua sheet 168, there is a general elevation of radionuclides in the study area as shown in figure 2,3 and 4 respectively when compared with the dose coefficients for ingestion of radionuclides by adult members of the public as shown in Table 3. The nominal probability coefficient for radiation-induced stochastic health effects, which include fatal cancer, non-fatal cancer and severe hereditary effects for the whole population is $7.3 \times 10^{-2}/\text{Sv}$ [17]. Comparing the outcomes with RDL of 0.1mSv annual drinking water exposure provides an estimated upper-life danger that could pose an important radiological danger to residents of developing cancer and kidney damage over a lengthy period of time.

5. Conclusion

acceptable limit. For Annual Effective Dose Equivalent (AEDE) for uranium, thorium and potassium the radioactive concentration shows that uranium and thorium are below allowable limit of 0,1mSv/yr according to international standard reported, however potassium is far above the permissible level. The Excess life Cancer Risks (ELCR) value for radionuclide concentration of ^{238}U , ^{232}Th , ^{40}K values was calculated and comparing these values with the

The activity concentration of radionuclide in groundwater was determined using the ICP-MS technique, radiological risk assessment was conducted, in Jos and environs (Naraguta sheet 168). The results showed that the concentration of ^{238}U ranged from 7.41×10^{-2} to 2.09Bq/kg with an average value of 1.1×10^{-1} Bq/kg, ^{232}Th ranged from 2.05×10^{-4} to 9.89×10^{-4} Bq/kg with a mean value of 2.51×10^{-4} Bq/kg, and ^{40}K the ranged from 3.34

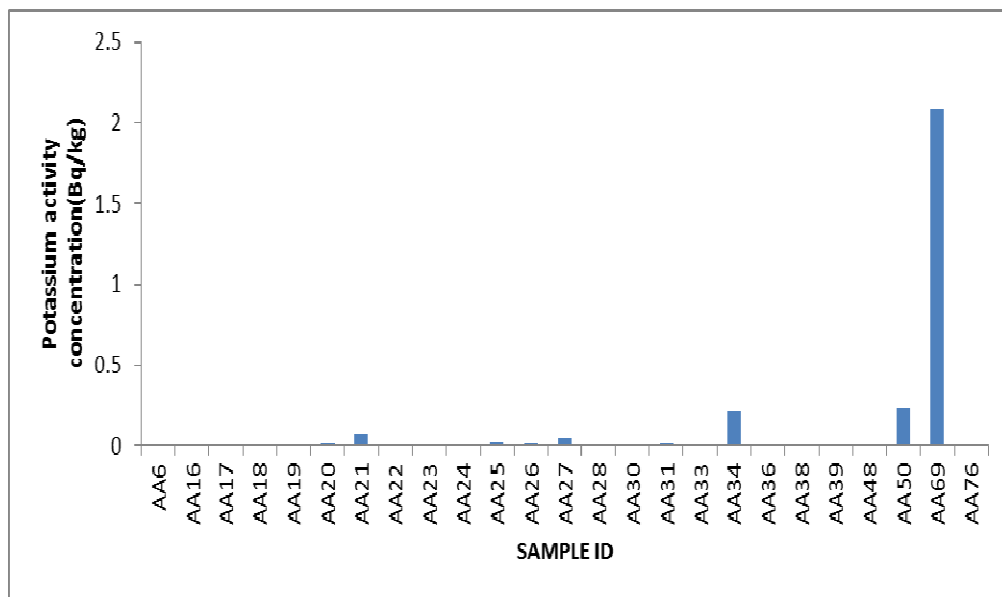


Figure 4: Graph of Potassium activity concentration against sample location for ground water.

× 10² to 7.67 × 10³ Bq/kg with a mean value of 1.256 × 10³ Bq/kg. The results of the evaluation of Ra_{eq}, AEDE, AGDE and ELCR were calculated, it was below the world average of 0.1 mSv/yr, 300 μSV/yr and 0.29 × 10⁻³ mSv/yr in some locations and exceeded the allowable limit in some areas respectively. Comparing the results with international standards, there is a general elevation of radionu-

clides in the study area which might pose a significant radiological risk to the inhabitants if the groundwater is not treated either for domestic or industrial use.

Acknowledgement

This work was carried out using the Naraguta sheet 168 and analysed at ACME laboratory Vancour Canada using ICP-MS technique.

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Table 3: Dose Coefficient for Ingestion of Radionuclides by adult members of the public.

Category	Radionuclide	Dose coefficient (mSv/Bq)
Natural uranium series	Uranium-238	4.5 × 10 ⁻⁵
	Uranium-234	4.9 × 10 ⁻⁵
	Thorium-230	2.1 × 10 ⁻⁴
	Radium-226	2.8 × 10 ⁻⁴
	Lead-210	6.9 × 10 ⁻⁴
	Polonium-210	1.2 × 10 ⁻³
Natural thorium series	Thorium-232	2.3 × 10 ⁻⁴
	Radium-228	6.9 × 10 ⁻⁴
	Thorium-228	7.2 × 10 ⁻⁵
Fission products	Caesium-134	1.9 × 10 ⁻⁵
	Caesium-137	1.3 × 10 ⁻⁵
	Strontium-90	2.8 × 10 ⁻⁵
	Iodine-131	2.2 × 10 ⁻⁵
Other radionuclides	Tritium	1.8 × 10 ⁻⁸
	Carbon-14	5.8 × 10 ⁻⁷
	Plutonium-239	2.5 × 10 ⁻⁴
	Americium-241	2.0 × 10 ⁻⁴

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