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Radionuclide Concentration of Soil and Sediment in Gold-Mining Locations of Atakumosa-West Local Government, South-West Nigeria

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Abstract: This study determined the activity concentrations of ^{40}K, ^{232}Th and ^{238}U in the soil, and sedimentusing gamma spectroscopy. This is with a view to determining the impacts of the mining operations on concentration of radioactive elements in the environment. Soil and Sediment were collected from three gold mining locations of Atakumosa-west Local Government area of Osun State for a period of 12 months. The control site selected for the study is Tonkere village, about 13Km from the study site, in Ife North-Central Local Government area of the State. Collected samples were analysed using gamma spectrometer. The NaI detector used was properly calibrated using absolute gamma ray energy for easy identification of peaks in the gamma spectrum. Results obtained from Gamma spectroscopic analysis revealed higher concentration of the three primordial radionuclidewith ^{40}K having a generally high value in the locations 1 and 2 and increasing through the wet season while having lower values in the dry season. A similar trend was observed for ^{232}Th and ^{238}U whose recorded values were also higher in the wet season than in the dry season. Values recorded in all the samples were higher than world safe limits. Notably, similar results obtained for samples from control site were lower.

Keywords: Activity, Concentration, Gold-Mining, Environment, Impact

1. Introduction

Radioactivity and radiation levels have been of great concern to scientist throughout the world. An early identification of environmental radionuclide and their contribution to total exposure have been investigated [1]. Radiation levels are being constantly monitored by scientist throughout the world, because of the hazards that they entail. Various workers have reported the occurrence of radioactive elements in environmental samples. Researchers have investigated radon levels in old abandoned metalliferous mines in South-West England. Other workers have also reported Uranium in drinking water in the U.S.A [2]. They have also published data showing significant radioactive contamination of soil around a coal-fired thermal power plant in Hungary.

Here in Nigeria, various researchers have worked on radioactivity content in various localities; notable is that of the koko radioactive waste dump site in Delta State. Well articulated and documented data on radioactivity have also been published here in Nigeria. Gbadeboand Amos [3] worked on the distribution of natural radionuclide in granitic rocks and soils of abandoned quarry sited in Abeokuta, Southwestern Nigeria. Tchokossa et al [4] reported the results of measurement of naturally occurring radionuclides in the community water supplies in two local government areas in Ile-Ife, Osun State. Similarly, Olomo et al also [5] worked on the distribution of gamma emitting natural radionuclides in soils and water surrounding nuclear research establishments in Ile-Ife, Osun State. Gbadebo and Amos [3] have assessed and published the results from radionuclides analysis from the bed rock and soil around Ewekoro cement factory located in Ogun State, south west Nigeria.

The concentration of naturally occurring radioactive materials (NORM) in most natural substances is low, higher concentrations may arise as a result of human activities. Mineral exploration and mining processes contribute to the concentration of naturally occurring radioactive materials (NORMs). Other processes such as beneficiation may increasetheir levels through movement of soils, sediments and rocks from different places on the surface or subsurface and thus, leading to exposure and concentration of these radioactive materials when the soils are dumped in the form of heaps. These mining activities and mineral exploration enhances increased radiation to man and his immediate environment. The problem associated with high exposure to radioactive elements as a result of mining activities and mineral exploration is particularly dangerous in that signs of the diseases may remain hidden for longer years after in humans after the exposure [6]. Leukemia, cataracts, hematological depressions as well as high risk of developing cancer are among the serious health issues that could results from accumulation of high doses of radioactive materials.

2. Materials and Methods

Atakumosa West Local Government area of Osun State, South-Western Nigeria lies within latitudes 7.51^0 N and 7.65^0 N and longitudes 4.61° E and 4.85° E and covers an area of 577 km² and a population of 68,643 [7]. Three mining locations each was sampled in the selected villages with Tonkere, a village about 13Km from the area was used as control site. Accessibility to the area is through a few motorable roads and several footpaths created by both farmers and miners. A sample each of Soil and Sedimentwas collected from each location. The sampling positions were marked using a Geographical Positioning System (GPS). The soil samples were collected in polythene bags, sealed properly and labeled neatly. The details of the samples e.g sample code, sampling location, latitude and longitude of the location, date of collection etc. were properly noted. The sampling locations are as described in Table 1.

Table 1. *Description of the Sample locations.*

Village	Locations	N	E
	L1	$7^0$39'43.6"	439'10.9"
Itagunmodi	L2	731'27.5"	439'10.3"
	L3	731'20.7"	439'13.8"
	L1	734'44"	441'13"
Igun	L2	734'58.7"	441'21.8"
	L3	734'51.9"	441'32.7"
	L1	731'28.1"	439'55"
Sabo	L2	731'29.4"	439'56.3"
	L3	731'46.5"	439'43.3"

Soils and Sediment samples collected were air-dried, pulverized and weighed into plastic containers which has been washed with distilled water and rinsed with dilute HCl and air-dried to prevent contamination. The difference in the weights of the empty container and the filled were noted as the weight of the pulverized sample. The labeled plastic containers were covered and properly sealed for 28 days for secular equilibrium after which each sample was counted in a NaI detector for seven (7) hours.

The activity concentration of natural radioactivity in the soil and sediments samples collected were determined using a 7.62 cm × 7.62 cm NaI (Ti) detector surrounded with adequate lead shielding that reduces the background by a factor of approximately 95%. A counting time of 25,200s was used. The activities of various radionuclide's were determined in Bq kg^{-1} from the count spectra obtained from each of the samples using the gamma ray photo peaks corresponding to energy of 1120.3 keV (214Bi), 911.21 keV (228Ac) and 1460.82 keV (40K) for 238U, 232Th and 40K, respectively. For a counting time of 25,200 s, the detection limits of the NaI (Tl) detector system were calculated as 6.77, 11.40, and 12.85 Bq kg−1 for 40K, 232Th, and 238U, respectively.

The NaI detector was calibrated using the absolute gamma ray energy for easy identification of peaks in the gamma spectrum. This was done using standard sources with specific gamma ray energies (241Am (59.5 keV), 137Cs (662.3 keV), 22Na (511.0 keV) and 60Co (1173.2 and 1332.5 keV) different from energies of radioactive elements expected from the study samples. The standards were obtained from the Isotope Products Laboratories, Burbank California, USA. The standards were counted for a long period to get well defined photo peaks, thereby ensuring the coverage of wide range of radionuclides.

3. Results and Discussion

Results obtain for the Gamma spectroscopic analysis of soil and sediment samples collected from the study area is as shown in Tables 2-4. The activity concentration of ^{40}K, ^{232}Th and ^{238}U are shown in Bq/Kg. The table revealed the activity concentration of ^{40}K in soil samples increasing in locations 1 and 2 of Igun mining village from March to June of the wet season and later decreases from July to February in the dry season with the highest concentration (1219. 1330.5) Bq/Kg (L1, L2) in June while the lowest value was recorded in February (738.5, 675.4) Bq/Kg (L1, L2). The highest concentration (1281.4 Bq/Kg), in location 3 however, was recorded in May while the lowest (726.9 Bq/Kg) came in Jan of the new season. For ^{232}Th, the highest concentration (552.6592.8, 627.4) Bq/Kg (L1, L2, L3) was recorded in April, during the wet season while concentration values decrease irregularly from May to February with the lowest value (183.3, 187.9, 202.8) Bq/Kg (L1, L2, L3) respectively. Activity concentration of ^{238}U also has higher values in the wet season increasing from March to August in locations 1, 2 and 3 (242.5, 369.7, 322.5) Bq/Kg. Notably, in control samples, all have lower concentration recorded for ^{40}K, ^{232}Th and ^{238}U, this lower value recorded in control samples shows the effect of mining operations in the study areas since no digging/mining of any sort is observed in the control site which is equally an agricultural area.

In Itagunmodi mining village, the same trend was repeated with the activity concentration of ^{40}K, ^{232}Th and ^{238}U increasing during the wet period from March to June in

location 1, 2 and 3 with the wet month of June recording the highest value (933.4, 910.3, 1110.3) Bq/Kg for ^{40}K, (711.2, 746.1, 678.9) Bq/Kg for ^{232}Th and (233.5, 279.1, 228.3) Bq/Kg for ^{238}U, and decreasing irregularly during the dry period from with lowest value in February.

(590.4, 404.1, 639.9) Bq/Kg for ^{40}K, (137.3, 146.5, 122.2) Bq/Kg for ^{232}Th and (123.8, 113.4, 107.5) Bq/Kg for ^{238}U. Also, the concentration values recorded for the radioactive elements in control samples are all lower than in the study samples. The highest value recorded for the activity concentration of ^{40}K in Sabo mining village increases steadily from March up to June and afterwards behaves irregularly until the last sampling month of February. The highest concentration were recorded in L1 and L2 (774.3, 717.4) Bq/Kg in the month of August at the tail end of the season while in the month of June, L3 recorded the highest value (719.2 Bq/Kg) of the element. For ^{232}Th and ^{238}U, the pattern was irregular throughout the study period.

Highest values (202.2, 188.4, 192.6) Bq/Kg in L1, L2 and L3 were recorded in March and April respectively for ^{232}Th and (91.3, 100.6, 110.3) Bq/Kg in L1, L2 and L3 for ^{238}U in the wet month of June. In all the months, the lowest values recorded for ^{40}K (515.3, 500.4, 481.9) Bq/Kg was in the early wet season in L1, L2 and L3, (84.8, 82.1, 81.9) Bq/Kg for ^{232}Th in L1, L2 and L3 in the late wet/early dry season. These values were all higher than values recorded in the control samples for the twelve months study period.

The mean activity concentration of ^{40}K, ^{232}Th and ^{238}U in soil samples from study and control site for the entire period is shown in Figure 1. It shows the activity concentration of ^{40}K to be highest in all the samples from all locations and the control samples exceeding the 370 Bq/Kg limit by the UNSCEAR. Lower mean concentration value in control sample is an indication of increased.

Table 2. *Activity Concentration (Bq/Kg) in Soil Samples from March to June.*

Soil	March			April			May			June		
	^{40}K	^{232}Th	^{238}U	^{40}K	^{232}Th	^{238}U	^{40}K	^{232}Th	^{238}U	^{40}K	^{232}Th	^{238}U
IgunL1	891.2	446.2	197.2	923.5	552.6	227.1	1110.5	305.7	233.5	1219.2	331.4	239.4
L2	885.1	499.1	176.2	900.7	592.8	211.5	1002.4	294.6	301.4	1330.5	310.9	344.2
L3	911.7	571.7	201.5	1001.4	627.4	277.6	1281.4	338.1	331.2	1008.7	389.2	379.4
ItagmoL1	779.5	509.5	173.6	823.5	552.9	191.3	889.6	664.6	220.4	933.4	711.2	233.5
L2	751.4	534.4	180.2	799.8	571.1	219.5	856.9	693.7	265.1	910.3	746.1	279.1
L3	800.2	498.4	156.2	886.9	500.7	199.5	903.7	618.2	200.3	1110.3	678.9	228.3
SaboL1	551.3	200.2	65.3	522.8	200.1	72.1	633.5	89.1	88.2	771.2	88.1	91.3
L2	500.4	173.1	71.3	512.7	188.4	84.6	600.7	94.6	92.5	714.9	82.1	100.6
L3	481.9	192.6	60.4	496.9	190.5	88.2	623.9	100.5	91.1	719.2	92.5	110.3
Control	304.8	91.8	28.8	302.1	88.5	28.9	310.1	94.2	29.6	300.8	91.5	30.3

Table 3. *Activity Concentration (Bq/Kg) in Soil Samples from July to October.*

Soil	July			August			September			October		
	^{40}K	^{232}Th	^{238}U	^{40}K	^{232}Th	^{238}U	^{40}K	^{232}Th	^{238}U	^{40}K	^{232}Th	^{238}U
IgunL1	1102.3	300.5	240.1	1113.3	303.5	242.5	1047.1	285.4	220.8	1080.2	294.4	204.8
L2	1008.2	308.1	366.1	1018.2	311.1	369.7	957.3	292.6	336.8	988.9	301.9	311.1
L3	1118.4	331.3	319.4	1129.5	334.6	322.5	1062.4	314.7	293.8	1096.3	324.6	271.9
ItagmoL1	881.3	225.1	229.4	890.1	227.3	231.6	837.2	213.8	211.7	863.6	220.5	194.9
L2	901.5	239.4	210.1	910.5	241.7	212.2	856.4	227.4	193.2	883.4	234.6	178.5
L3	955.1	200.4	199.5	964.6	202.4	201.4	907.3	190.3	183.5	935.9	196.3	169.5
SaboL1	766.7	89.3	89.3	774.3	90.1	90.1	728.3	84.8	82.1	751.3	87.5	75.9
L2	710.3	91.7	92.2	717.4	92.6	93.1	674.7	87.1	84.8	696.4	89.8	78.3
L3	638.2	86.3	88.3	644.5	87.1	89.1	606.2	81.9	81.2	625.4	84.5	75.5
Control	307.6	94.9	33.2	310.6	95.8	33.5	292.2	90.1	30.5	301.4	93.2	28.22

Table 4. *Activity Concentration (Bq/Kg) in Soil Samples from Nov to Feb.*

Soil	November			December			Jan			Feb		
	^{40}K	^{232}Th	^{238}U	^{40}K	^{232}Th	^{238}U	^{40}K	^{232}Th	^{238}U	^{40}K	^{232}Th	^{238}U
IgunL1	881.8	225.3	156.6	859.7	216.6	148.8	716.4	186.1	132.5	738.5	183.3	129.6
L2	806.5	231.7	237.9	786.3	221.8	226.9	655.3	191.2	201.3	675.4	187.9	197.6
L3	894.7	248.4	207.6	872.3	238.5	198.2	726.9	205.4	175.6	749.3	202.8	172.4
ItagmoL1	705.4	168.8	149.1	687.4	162.7	142.2	572.8	139.5	126.1	590.4	137.3	123.8
L2	721.2	179.5	136.5	703.1	172.3	130.2	585.9	148.4	115.5	604.1	146.5	113.4
L3	764.8	150.3	129.6	744.9	144.2	123.5	620.8	124.2	109.7	639.9	122.2	107.5
SaboL1	613.6	66.9	58.3	598.4	64.2	55.3	498.3	55.3	49.1	513.6	54.4	48.2
L2	568.4	68.7	59.9	554.5	66.3	57.1	461.6	56.8	50.7	475.9	55.9	49.7
L3	510.6	64.7	57.3	497.7	62.1	54.7	414.8	53.5	48.5	427.5	52.6	47.6
Control	246.8	71.1	21.5	239.9	68.3	20.5	199.9	58.8	18.2	206.7	57.8	17.9

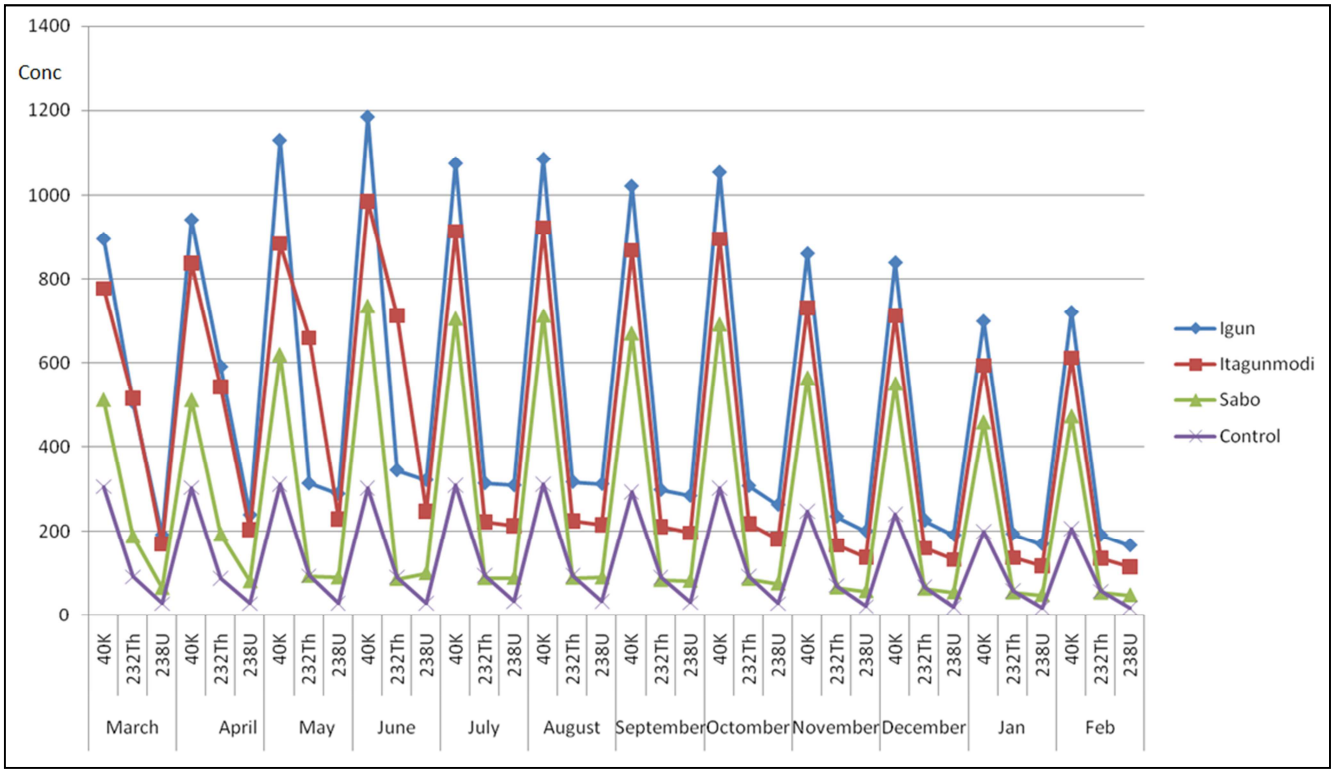

Figure 1. *Mean Activity Concentration of Radionuclide in Soil Samples and Control.*

Human activity, via mining, in the study area. High concentration of ^{40}K in soils in the area is due to the high content of feldspar, mica and clay mineral characterizing the rock and soil in the region [8]. However, in all samples and control, the 30 and 35 Bq/Kg limit was exceeded.

In sediment samples (Tables 5-7), activity concentration of ^{40}K was increasing up to October in all locations, recording the highest value (783.4, 770.8, 787.8) Bq/Kg (L1, L2, L3) at the beginning of the dry season but decreases afterwards in samples from Igun, this increasing value cannot be unconnected with the effect of run off during the wet period that tends to increase the concentration of the radionuclide which decreases during the dry season. In Itagunmodi, the highest activity concentration values (695.2, 799.9, 675.1) Bq/Kg of ^{40}K and (139.2, 137.7, 120.3) Bq/Kg of ^{238}U were recorded in L1, L2 and L3 of the village in the month of August and (252.7, 265.6, 302.6) Bq/Kg of ^{232}Th in January while the lowest values (200.9, 205.5, 280.7) Bq/Kg for ^{40}K, (278.1, 271.6, 247.1) Bq/Kg for ^{232}Th and (66.8, 76.0, 59.1) Bq/Kg for ^{238}U were recorded in the dry months of November, January and December respectively. At Sabo, the lowest activity concentration of ^{40}K was recorded in November in all locations, though the highest value came in October (589.4 Bq/Kg) for location 1 while the month of August recorded the highest for locations 2 and 3 (589.7, 669.5) Bq/Kg respectively. ^{232}Th and ^{238}U followed a similar trend, having the highest values (207.6, 198.3, 212.5) Bq/Kg and (99.2, 87.1, 101.2) Bq/Kg respectively at the little dry season experienced in August (of the study year). The lowest value (125.4, 119.8, 128.3) Bq/Kg for 232Th and (42.0, 37.2, 40.8) Bq/Kg for 238U came in February and December respectively during the dry season. Notably in all (Tables 5, 6, and 7), samples collected from control site has lower activity.

Table 5. *Activity Concentration of in Sediment Samples and Control for March to June.*

	March			**April**			**May**			**June**		
	^{40}K	**^{232}Th**	**^{238}U**	**^{40}K**	**^{232}Th**	**^{238}U**	**^{40}K**	**^{232}Th**	**^{238}U**	**^{40}K**	**^{232}Th**	**^{238}U**
IgunL1	564.7	241.8	103.2	489.1	171.8	115.3	611.5	242.8	100.3	677.1	131.5	103.8
L2	601.2	287.9	144.1	571.1	131.8	129.4	634.8	216.9	147.3	669.3	129.4	155.4
L3	551.5	288.1	139.2	584.3	117.3	135.5	701.3	139.4	152.9	739.1	119.2	121.9
ItagmoL1	499.1	200.1	94.2	522.5	138.6	103.6	517.3	240.3	126.2	511.1	176.9	107.9
L2	500.7	291.7	101.4	488.3	103.5	117.3	558.9	166.5	131.9	500.1	118.3	122.6
L3	515.7	238.1	96.1	568.4	155.9	129.6	615.7	261.4	100.9	558.3	199.5	95.3
SaboL1	441.2	154.2	55.7	411.4	149.6	60.4	481.3	196.1	78.2	480.1	155.7	67.8
L2	419.4	133.9	49.6	415.8	151.4	52.8	466.9	187.3	66.9	471.9	169.3	60.1
L3	398.4	159.2	50.4	400.2	163.7	61.8	507.2	201.3	80.1	499.4	182.9	65.9
Control	225.6	101.7	25.1	220.1	103.8	27.8	231.7	99.2	23.9	242.9	105.4	25.9

Table 6. *Activity Concentration of in Sediment Samples and Control for July to October.*

	July			August			September			October		
	^{40}K	^{232}Th	^{238}U	^{40}K	^{232}Th	^{238}U	^{40}K	^{232}Th	^{238}U	^{40}K	^{232}Th	^{238}U
IgunL1	599.4	148.4	118.4	707.3	152.8	119.5	759.4	125.8	108.8	783.4	139.4	106.5
L2	585.8	238.2	169.5	693.6	142.5	171.1	746.5	216.9	155.4	770.8	229.4	152.5
L3	503.1	198.5	155.3	711.1	202.4	156.8	762.9	278.5	142.6	787.8	190.6	139.7
ItagmoL1	588.4	207.6	137.9	695.2	111.6	139.2	653.9	187.2	126.8	674.2	199.4	124.1
L2	579.2	128.4	136.4	799.9	232.6	137.7	752.4	206.8	125.8	776.6	119.8	122.7
L3	568.5	288.1	119.2	675.1	192.9	120.3	635.7	163.5	109.4	655.3	178.3	107.2
SaboL1	501.3	205.6	98.3	607.3	207.6	99.2	571.2	195.2	90.6	589.4	201.4	88.4
L2	483.9	196.4	86.3	589.7	198.3	87.1	554.7	186.8	79.6	572.2	192.4	77.6
L3	562.9	210.4	100.2	669.5	212.5	101.2	629.7	199.8	92.4	649.2	206.1	90.1
Control	236.8	100.4	27.5	239.1	101.4	27.7	224.9	95.8	25.3	232.4	98.3	24.7

Table 7. *Activity Concentration of in Sediment Samples and Control for November to February.*

	November			December			Jan			Feb		
	^{40}K	^{232}Th	^{238}U	^{40}K	^{232}Th	^{238}U	^{40}K	^{232}Th	^{238}U	^{40}K	^{232}Th	^{238}U
IgunL1	380.2	136.3	76.9	353.2	122.8	64.3	519.6	278.1	65.1	535.5	273.5	63.9
L2	417.8	128.6	110.1	492.4	115.5	96.3	510.8	271.6	93.2	526.4	267.3	91.5
L3	422.3	198.8	100.9	441.8	186.9	75.5	522	247.1	85.4	538.1	243.1	83.8
ItagmoL1	200.9	105.7	89.6	395.2	193.4	66.8	447.4	252.7	75.8	461.2	248.6	74.4
L2	205.5	121.3	88.6	277.6	108.4	76.0	514.8	265.6	75.0	530.6	261.3	73.6
L3	280.7	166.7	77.4	314.3	151.4	59.1	434.5	302.6	65.5	447.8	297.7	64.3
SaboL1	362.5	154.2	63.8	401.4	148.0	42.0	390.8	127.4	54.1	402.8	125.4	53.1
L2	353.5	147.3	56.4	444.2	141.4	37.2	379.5	121.7	47.4	391.2	119.8	46.6
L3	364.5	157.8	65.1	506.2	151.4	40.8	430.8	130.4	55.1	444.1	128.3	54.2
Control	19.1	75.3	17.8	470.4	72.2	16.1	153.9	62.2	15.2	158.6	61.2	14.9

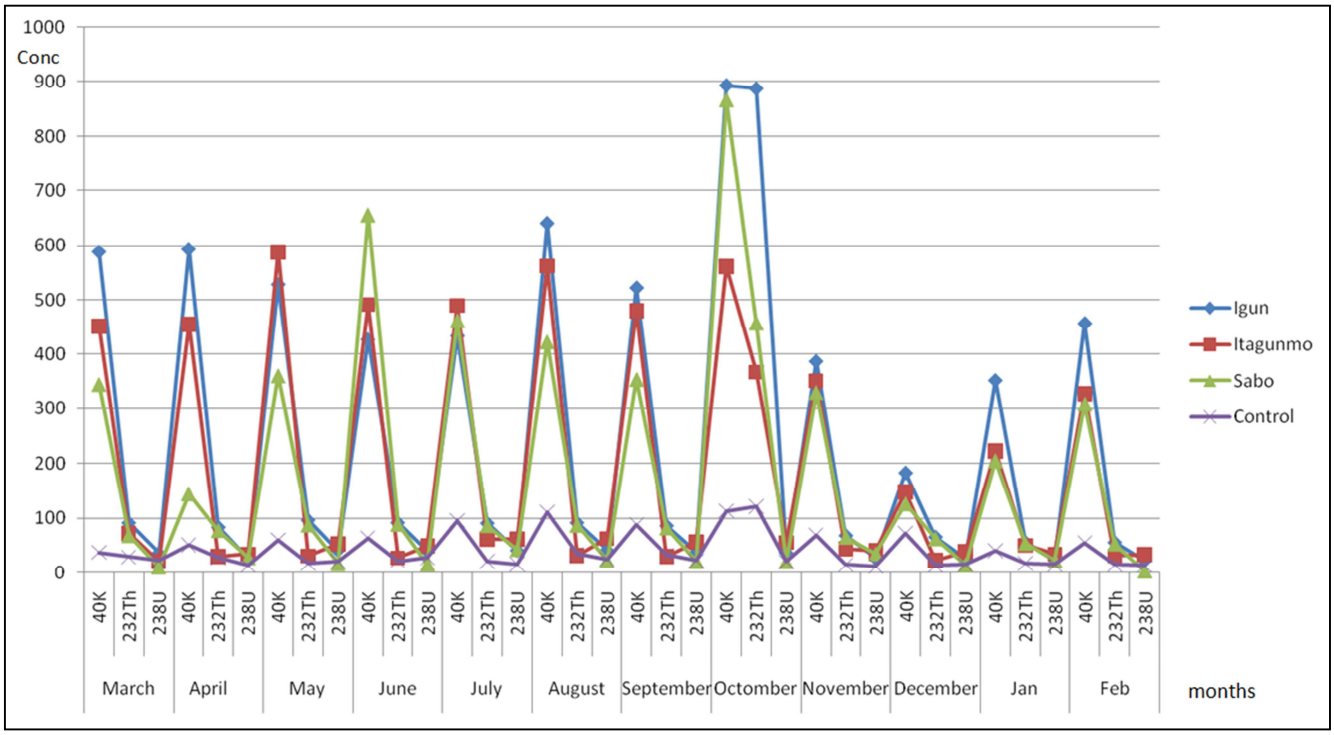

Figure 2. *Mean Activity Concentration of Radionuclide in Sediment Samples and Control.*

Concentration values for ^{40}K, ^{232}Th and ^{238}U. High values recorded in most cases during the wet and little dry season (mid-July to August) will be attributed to runoff effect observed in Nigeria between March and July of the study year. The mean activity concentration of the radioelement from the study site and control similarly showed ^{40}K as the highest concentration value in all locations, followed by ^{232}Th while the activity concentration of ^{238}U is lowers in all samples and control. In all study samples the lowest mean activity concentration of ^{40}K (461.9 Bq/Kg) recorded

in Sabo mining village, is higher than the world recommended 450 Bq/Kg for the radioelement in stream sediment samples. Similarly, all samples has mean values exceeding the 50 Bq/Kg limit for ^{232}Th and 50 Bq/Kg for ^{238}U in sediment samples, this particularly calls for concern considering the ecological importance of sediment for both benthic and aquatic organisms. Observably, the mean activity concentration recorded for control samples is higher than the world average, further study might be necessary to ascertain the source of the sediment [9] which determines, to a large extent, the load. High concentration of radionuclide in stream sediment is undesirable because of its ecological importance.

The United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) has stipulated 370, 30 and 35 Bq/Kg for ^{40}K, ^{232}Th and ^{238}U in stream sediment samples respectively [10].

4. Conclusion

Gamma spectroscopic analysis of soil and sediment samples from the study area hasrevealed higher concentration of three primordial radionuclide ^{40}K, ^{232}Th and ^{238}U generally in the wet months of March to June while having lower values in the dry months, High values recorded during the wet and little dry season (mid-July to August) has been attributed to runoff effect. Values recorded for the radionuclide in all the samples here higher than world safe limits recommended by the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) in soil and sediments. Notably, similar results obtained for samples collected from control site were however lower than recorded in study samples and were also lower than world safe limits in all the sample types. Though, concentration of naturally occurring radioactive materials (NORM) is usually very low in the environment with negligible health impacts, however, higher concentrations may arise as a result of human activities, Mining activities has been implicated as it is peculiar to the study sites, hence, the high values.

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