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Determination of Radioactivity Levels and Hazard of Water and Sediment Samples in Various Gold Mining Pits at Itagunmodi Ilesha Nigeria

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Abstract: The natural radioactivity levels in sediment and water samples of gold pits in Itagunmodi Ilesha have been determined. The activity concentrations obtained for series ^{226}Ra, ^{232}Th and non-series ^{40}K are measured in the unit of Bq kg^{-1}. The overall average concentrations of radionuclides were 28.9, 53.3 and 532.6 Bq kg^{-1} respectively in sediment samples; while the overall average of ^{226}Ra, ^{232}Th and ^{40}K in water samples were 6.0, 8.9, and 103.6 Bq l^{-1} respectively. The results obtained were compared with others radioactivity measurement in different countries sediments and water. The estimated radium equivalent activity (Ra_{eq}), the absorbed dose (D_r), external hazard index, the annual gonadal dose equivalent (AGDE), the annual effective dose equivalent (AEDE) in both sediments and water samples fall within acceptable limit. Assessment of anthropogenic pollution in sediments gives concentrations that are within acceptable safety limit. A study of elemental concentration ratios of Th/U, K/U and K/Th revealed that depletion and enrichment process of these radionuclides in sediment and water samples in the area are almost the same.

Keywords: Sediment; water; radioactivity; gold; elemental ratio; Itagunmodi Ilesha**.**

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1. Introduction

Primordial radionuclides such uranium, radium and radon have been found to have certain degree of concentrations in water, rock, soil and sediment [1], [2], [3]. The concentrations of the aforementioned radionuclides and other anthropogenic sources in sediment and water vary widely and are the source of continuous exposure of human kinds to terrestrial radioactivity in area with mineral deposit [4], 5]. These radionuclides have made all the goods, food, air [6], sediment and water partly radioactive. Therefore, man and his environment are constantly being exposed to ionizing radiation [7], [8] as a result of their dependency on ground, air, food, the universe and even elements in their own bodies [6] for survival. The demand for gold has been high since global financial crisis. Therefore, the quest for gold mining has increased. It has been established that gold bearing deposits contain harmful concentrations of lead. The crushing of rocks into sand in flour mill produces large amount of dust which is highly contaminated with lead [9]. Soluble compounds (gold salts) such as gold chlorine are toxic to kidney and liver. Potassium gold cyanide used in gold electroplating is toxic by virtue of both their cyanide and gold content [10]. These soluble compounds can enter into human bodies via water, food stuff, sediment and soil used as building material and agricultural purposes in the area where gold is deposited. Gold production is associated with contribution to hazardous pollution [11]. These have taken place worldwide both in developed, poor and developing countries which killed the marine life in long stretches of affected rivers [12], [13]. Typical of these developing counties is Nigeria, where lead of order 23000 ppm from processing of gold ore had affected people living in Bagega in Zamfara State, Nigeria [9]. This has killed about 400 children, affected about 3500 children and some adults have infertility and miscarriage [9], [14]. This was discovered in March 2010 [15]. Lead (being a progeny of 238U and 232Th) from processing of gold ore as being ascertained to enter into body through lungs and digestive tract

This study has been carried out to ascertain the concentration of radioactivity in water and sediment samples from Itagunmodi which has significant concentration of gold deposit in disseminated form. This was used for assessment public dose rate and to keep a base line data in order to ascertain if the area is contaminated as seen in Zamfara State Nigeria. More also, this study was used to determine the anthropogenic activities contribution and the pollution status of the area

2. Materials and Methods

2.1 Sediment and water sampling.

Sediment samples were taken in the bottom of five pits (out of available pits) in four locations with the help of some miners in the area. Three samples were collected from each pit at six months interval. A total of sixty sediment samples were collected in the pits that are at interval of 20 m apart. Fig 1 shows some of the samplings pits. The average for each pit was taken after exposure. The samples were properly labeled, catalogued and brought to Radiation Laboratory at the Centre for Energy Research and Development (CERD) at Obafemi Awolowo University (OAU) Ile-Ife, Nigeria for counting and processing before analysis. Samples locations are listed in Table 1. Three water samples were collected directly from each pit at six months interval using manual procedure for collecting water from deep or shallow pits. Each sample was stored in 1 litre plastic keg that has been thoroughly washed with dilute acid (0.1 M HCl) [4]. Samples locations are presented in Table 2.

2.2. Sediment samples processing and packing

At the laboratory, after drying at room temperature for many days, wood and other non-sediment matter were removed. The samples were dried in the oven at a controlled temperature of 1000C until a constant weight was obtained. The dried samples were ground, powered and pass through 2 mm sieve [16], [17].

Samples were sealed in gas-tight, radon impermeable trap-shape hermetically plastic containers of diameter 75 mm and height 37 mm [18]. The samples were sealed for a period of 4 weeks. This was done to achieve secular equilibrium between ^{222}Rn and ^{226} Ra before gamma spectroscopy.

2.3. Water samples processing and packing

Storing containers were thoroughly washed with dilute acid (0.1 M HCl) and water samples were acidified with 11 M of HCl at the rate of 10 ml per litre of sample to avoid absorption of radionuclides on the walls of the containers [4]. The samples were sealed and stored for twenty eight days for secular equilibrium to be reached among the progeny of ^{238}U and ^{232}Th.

2.4. Radiometric Analysis of sediment

Radioactivity measurements were carried out using a lead-shielded 76 mm x76 mm NaI (Tl) detector crystal coupled to a Canberra series 10 plus Multichannel Analyzer (MCA) through a preamplifier. The detector had a resolution of about 8 % at energy of 0.662 MeV. The quantification of radionuclide present in sediment samples was obtained through accurate energy and efficiency calibration. This was achieved through standard sample supplied by the IAEA, Vienna, Austria. The choice of counting time of 10 hrs was dictated by the activities of the samples and the precision required for the data [5], [19]. The activity concentration of ^{214}Bi (determined from its 1.765 MeV γ-ray peak) was chosen to provide an estimated of ^{226}Ra in the samples, while that of the daughter radionuclide ^{208}Tl (determined from its 2.615 MeV γ-ray peak) was chosen as an indicator of ^{232}Th, ^{40}K was determined by measuring the 1.460 MeV γ-ray peak.

2.5. Radiometric Analysis of water

The gamma-counting equipment was NaI (Tl) crystal detector coupled to a Canberra Multichannel Analyzer (MCA) computer system. The quantification of radionuclide present in sediment samples was obtained through accurate energy and efficiency calibration.

This was achieved through standard sample supplied by the IAEA, Vienna, Austria. This technique was described elsewhere [20]. The counting time was 10 hrs. Empty container identical to that of sample 75 mm by 37 mm was also counted for the same counting time to determine the background distribution spectrum. Certain region was set for the expected radionuclide. This belongs to the naturally occurring series-decay such as ^{238}U (^{226}Ra), ^{232}Th and non-series decay type ^{40}K. The net area count after background corrections in each photopeak was used in computation of the activity concentration of each of the radionuclides in the samples. This was described in an earlier work [21].

2.6. Lead testing using Atomic Absorption Spectrometer (AAS)

The flame photometer technique was applied for the analysis of lead and other heavy metals in the sediment samples. This is done to ascertain the concentration of lead and other trace metals in the samples. The details of the methods have been elaborated in earlier work [3], [22]. The results obtained are shown in Table 3.

3. Results and Discussion

3.1. Activity mass concentration in sediment and water.

Tables 1 and 2 show the dry weight activity concentration of the major gamma emitting radionuclides ^{226}Ra, ^{232}Th and ^{40}K in the sediment and water samples. In the sediment samples, the overall average concentration of ^{226}Ra, ^{232}Th and ^{40}K were found to be 28.9, 53. 3 and 532.6 Bq kg^{-1} (dry weight) respectively. The overall average concentrations of ^{226}Ra, ^{232}Th and ^{40}K in water samples were found to be 6.0, 8.9 and 103.6 Bq l^{-1} respectively. The overall concentrations of the aforementioned radionuclides in both sediment and water are compared with that of other authors are presented in Table 4. Figs 2 and 3 show the distribution of ^{226}Ra, ^{232}Th and ^{40}K mean activity concentrations of sediment and water samples for each location. Elemental concentration ratios such as Th/U, K/U and K/Th were determined to indicate relative depletion or enrichment of these radionuclides in sediment and water samples.

The values obtained for these radionuclides using the overall average were 1.9, 18.5 and 10.0 in sediment; while that of water were 1.5, 17.2 and 11.7 for Th/U, K/U and K/Th respectively. Closer observations of these ratios indicate that the contribution of these radionuclides to both sediment and water are almost the same. These reflect the same depletion and enrichment process in both sediment and water samples. More data should be analyzed to ascertain definite value because of the geological nature and feature in the area as seen in figs 4a and b.

3.2. Absorbed dose rate in air

Absorbed dose rate in air depends explicitly on the measured activity concentration of ^{226}Ra, ^{232}Th and ^{40}K that are relevant. Dose rate at one metre above the ground surface was calculated using the following relation;

$$D_R = A \; x \; D_F$$

Where $D_R \; (nGy \; h^{-1})$ is the dose rate, A is the activity mass concentration in Bq kg^{-1} and D_F is the dose conversion factor in units of $nGy \; h^{-1} \; per \;\; Bq \; kg^{-1}$. From UNSCEAR [23] report which is based on Monte Carlo simulation technique, these factors are 0.462, 0.604 and 0.0417 for ^{226}Ra, ^{232}Th and ^{40}K respectively. Table 1 shows the results of absorbed dose rate in sediment samples. The calculated mean absorbed dose rate varied from 30.3 to 121.5 $nGy \; h^{-1}$. The overall average of 71.1 $nGy \; h^{-1}$ is higher than 59 $nGy \; h^{-1}$ which is the population weighted average value of global natural radiation [23]. This could be attributed to sediment geological formation.

3.3. Annual effective dose

For miners, adult that live and cultivate farm products in the area, the dose rate in sediment was converted to annual effective dose by using the following relation;

$$AEDE(\mu Sv \; y^{-1}) = D_R x T x 0.2 x 0.7 x 10^{-3}$$

Where $AEDE$ is the annual effective dose, D_R is the calculated dose rate, 0.2 is the outdoor occupancy factor for adults, while 0.7 Sv Gy^{-1} is the conversion coefficient from absorbed dose in air to effective dose received by adults and T is 8760 hy^{-1}.

The results of the calculation are given in Table 1. The average annual effective dose rate varied from 37.1 to 47.9 $\mu Sv \; y^{-1}$. The overall average 87.1 $\mu Sv \; y^{-1}$ is lower than world average 460 $\mu Sv \; y^{-1}$. Due to the fact that people depend on the water in the area for some activities such as bathing, drinking and washing to mention few, it is imperative to estimate annual effective dose in water. It was estimated using the following relation;

$$AEDE = \sum_i I_i x \; 365 x D_i$$

Where I_i is the daily intakes of radionuclide i (Bq. d^{-1}), D_i is the ingestion dose coefficient given as 6.2x10^{-9}, 2.8x10^{-7} and 6.9x10^{-7} (Sv. Bq^{-1}) for ^{40}K, ^{226}Ra and ^{232}Th respectively [24].

The results obtained are presented in Table 2. The overall average estimated AEDE received by people as result of ingestion of radionuclides in water are 0.2(0.2-0.3), 0.6(0.1-0.9) and 2.3(0.9-4.0) $\mu Sv \; y^{-1}$ for ^{40}K, ^{226}Ra and ^{232}Th respectively. The doses estimated fall below 1 mSv y^{-1} recommended for public exposure.

3.4. Annual gonadal dose equivalent

Rapidly dividing cells such as bone marrow and the bone surface cells are organs of interest by UNSCEAR [25]. Therefore, The AGDE due to specific activities of primordial radionuclides for those living and cultivating both food and cash crops in the area is calculated using the following relation;

$$AGDE(\mu Sv \; y^{-1}) = 3.09A_{Ra} + 4.18A_{Th} + 0.314A_K$$

The results obtained are presented in Table 1. The mean values varied from 200. 3 to 821.9 $\mu Sv \; y^{-1}$ with an overall average of 450.0 $\mu Sv \; y^{-1}$ which is lower than 550.5 $\mu Sv \; y^{-1}$ obtained in Rize, Turkey [6] and 2398 $\mu Sv \; y^{-1}$ obtained in Eastern Desert of Egypt [26].

3.5. Radium equivalent activity and External hazard index

In Nigeria 80 % of the population lives in the house built by the sediment derived from rivers and streams. Sediment from these rivers and streams are mixed with cement to bake block and mortar for plastering and tiling.

The suitability of sediment from various pits in the area is determined by estimating radium equivalent (Ra_{eq}) and external hazard index (H_{ext}). Radium equivalent activity is calculated through the following relation;

$$Ra_{eq} = A_{Ra} + \frac{10}{7}A_{Th} + \frac{10}{130}A_K \qquad 2$$

Where A_{Ra}, A_{Th} and A_K are the activity mass concentrations of ^{226}Ra, ^{232}Th and ^{40}K respectively. The average radium equivalent calculated ranged from 64.4 to 248.4 Bq kg^{-1} with an overall average 71.1 Bq kg^{-1} as seen in Table 1. The estimated average values are all less than 370 Bq kg^{-1} [27].

The external hazard index was calculated for the investigated sediment samples using the model proposed by Krieger as follows;

$$H_{ext} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \leq 1$$

This is done to keep the annual dose below 1.5 mGy and radium equivalent activity less than 370 Bq kg^{-1}. The results obtained ranged from 0.2 to 0.7 with an overall average 0.4 as presented in Table 1. The highest value of H_{ext} which is 0.6 has been found to be < 1 for the sediment sample in the study area. 3

4. Assessment of Anthropogenic pollution in sediments

Gold ore dumps are the source of many heavy metals such as cadmium, lead, zinc, copper and mercury [10]. Therefore sediment samples which are used for different purposes are tested for the presence of these metals and the result is as shown in Table 3. The samples have elevated value of lead, chromium, zinc and iron than the concentrations in the average shale. However, lead value is lower than 23,000 ppm obtained in Zamfara, Nigeria [9] and 400 ppm safety limit in USA.

5. Conclusion

The activity concentrations of natural radionuclide in sediment and water samples from Itagunmodi gold pits and their environmental radioactivity impact have been studied in this work. The mean concentrations of ^{226}Ra, ^{232}Th and ^{40}K in the area compared suitably with literature values. Dose rates, radium equivalent activity
external hazard, annual effective dose from sediments and daily intakes of these radionuclides in water have consistent values with those reported in other countries in the world as seen in Table 4. Based on the anthropogenic assessment of sediment and radioactivity assessment in water and sediment the area is still safe as at present, but needs constant monitoring to forestall the hazard occurrence as seen in Zamfara, Nigeria.

Table 1: Average radioactivity concentrations (Bq kg^{-1}), absorbed dose rates (nGy h^{-1}), radium equivalent(Bq kg^{-1}), gonadal dose (µSv y^{-1}) and effective dose rates (µSv y^{-1}) in the sediment samples.

Location	*pit*	*^{40}K*	*^{238}U (^{226}Ra)*	*^{232}Th*	*D*	*Ra_{eq}*	*H_{ex}*	*AGDE*	*AEDE*
Oke Agunmodi	1	662.18±60	27.94±5	45.06±14	70.37	143.36	0.39	482.61	86.30
	2	308.35±44	30.13±6	19.85±10	39.33	82.23	0.21	272.90	48.23
	3	857.87±51	29.60±3	106.78±18	120.39	248.35	0.67	807.18	147.65
	4	578.03±54	26.40±4	76.15±22	86.65	179.80	0.47	581.38	106.28
	5	812.86±65	25.33±4	49.15±13	78.47	196.81	0.43	538.95	96.24
Odo Agunmodi	1	956.86±55	33.23±3	98.34±18	120.63	247.53	0.67	814.19	147.94
	2	845.82±59	10.59±2	114.82±23	117.07	239.91	0.65	778.23	143.57
	3	704.40±59	30.17±5	73.23±21	91.88	189.28	0.51	621.14	112.68
	4	1096.15±86	23.98±4	96.57±27	121.52	246.48	0.68	821.95	149.03
	5	788.72±53	38.25±5	93.89±19	112.56	233.24	0.63	758.31	138.04
Odo Ode	1	422.03±49	20.47±4	38.72±13	52.61	108.34	0.30	357.62	64.52
	2	350.64±51	18.49±3	20.45±13	36.58	74.73	0.20	252.72	44.86
	3	698.42±49	35.88±4	91.27±18	105.91	220.17	0.59	711.68	129.89
	4	413.71±34	34.85±2	10.84±7	39.93	82.21	0.22	282.90	48.97
	5	127.98±39	40.26±3	18.10±9	34.70	75.99	0.21	240.25	42.56
Imoo	1	265.42±62	45.52±5	12.74±9	39.34	84.18	0.23	277.25	48.25
	2	173.45±59	48.51±4	21.41±12	42.38	92.48	0.25	293.85	51.97
	3	115.23±32	10.29±3	31.65±18	30.32	64.42	0.17	200.28	37.18
	4	230.91±46	23.45±4	18.52±11	32.24	67.71	0.18	222.38	39.54
	5	243.81±68	43.95±4	28.56±15	48.21	103.56	0.28	331.74	59.12
Overall Average		532.64±54	28.86±4	53.31±16	71.05	149.04	0.39	450.38	87.14

Table 2. :Daily intakes of Primordial radionuclides in (Bq l1) and annual effective dose (µSv y-1)

Location	Pit	^{40}K	$^{238}U(^{226}Ra)$	^{232}Th	^{40}K	$^{238}U(^{226}Ra)$	^{232}Th
		Daily intake per person			Annual effective dose		
Oke Agunmodi	1	78.58±22	2.95±1	7.41±3	0.2±0.0	0.3±0.1	1.9±0.8
	2	105.77±31	8.64±2	5.71±2	0.2±0.1	0.8±0.2	1.4±0.5
	3	106.42±26	1.37±0	9.32±4	0.2±0.1	0.1±0.0	2.3±1.0
	4	81.03±25	6.16±2	8.71±4	0.2±0.1	0.6±0.2	2.2±1.0
	5	102.25±37	6.57±2	9.48±4	0.2±0.1	0.7±0.2	2.4±1.0
Odo Agunmodi	1	95.47±24	6.43±2	10.07±3	0.2±0.1	0.7±0.2	2.5±0.7
	2	88.63±25	6.21±3	10.60±4	0.2±0.1	0.7±0.3	2.7±1.0
	3	115.29±38	8.49±3	7.70±3	0.3±0.1	0.9±0.3	1.9±0.8
	4	76.18±17	6.77±2	8.88±3	0.2±0.1	0.7±0.2	2.2±0.8
	5	113.19±30	7.52±3	3.85±1	0.3±0.1	0.8±0.3	0.9±0.3
Odo Ode	1	91.50±21	5.94±2	4.97±3	0.2±0.1	0.6±0.2	1.3±0.8
	2	110.39±36	7.19±3	12.28±4	0.2±0.1	0.7±0.3	3.1±1.1
	3	130.39±40	4.24±1	6.45±3	0.3±0.1	0.4±0.1	1.6±0.8
	4	134.54±35	4.78±2	9.21±3	0.3±0.1	0.5±0.2	2.3±0.8
	5	103.54±26	8.61±3	15.94±5	0.2±0.1	0.9±0.3	4.0±1.3
Imoo	1	99.67±25	3.05±1	9.14±4	0.2±0.1	0.3±0.1	2.3±1.0
	2	115.99±46	5.36±2	10.72±4	0.3±0.1	0.5±0.2	2.7±1.0

	3	81.68±29	8.15±2	10.66±3	0.2±0.1	0.8±0.2	2.7±0.8
	4	143.51±46	6.38±1	7.24±2	0.3±0.1	0.7±0.1	1.8±0.6
	5	98.67±29	5.50±2	9.24±3	0.2±0.1	0.6±0.2	2.3±0.8
Overall Average		103.64±30	6.02±2	8.88±3	0.2±0.1	0.6±0.2	2.3±0.8

Table 3: Concentration of heavy metals in sediment samples in μg g^{-1} compared with average shale

Metals	Range	Mean	Average Shale [28]
Mn	132-220	173	850
Co	10-32	21	19
Cr	2-22	5	90
Zn	99-226	163	95
Ni	26-75	43	68
Cd	0.25-0.33	0.2	0.3
Pb	80-125	108	20
Cu	21-53	34	45
Fe	22-58	39	4.72

Table 4: Comparison of range activity concentrations of sediments and water with other areas of the world

Country	**^{226}Ra (Bq kg^{-1})**	**^{232}Th (Bq kg^{-1})**	**^{40}K (Bq kg^{-1})**
Sediment[29], [3], [30], [31], [32], [33], [34], [35], [36], [37]			
Nigeria, Ilesha	10-49	11-115	115-1096 Present study
Nigeria, Ibadan	1.4 - 49.5	1.7- 68.8	2.6 -194.0
Nigeria, River Niger	14.9- 175.2	1.5- 65.6	4.0- 1045.0
Spain	62-293.9	12-63	-
Albania	13-23	13-40	266-675
Spain	9-14	11-16	220-460
Algeria	-	6-32	56-607
Egypt	4-48	8-50	16-487
Italy	-	31- 37	410-475
Greece	19-81	19-88	152-1593
French	9-62	16-55	120- 1026
Water [38], [38], [38] [39], [40], [41]			
Nigeria, stream	1-9	4-12	76-144 present study
Nigeria, well	3-14	0.3-4	92-109
Nigeria, tap	11-14	2-3	71-109
Nigeria, surface	6-8	3-4	48-90
China, ground	0.001-0.93	-	-
Denmark, well	0.55	-	-
Spain, surface	0.0282	-	0.132

Fig 1. Some of the sites visited at Itagunmodi Ilesha.

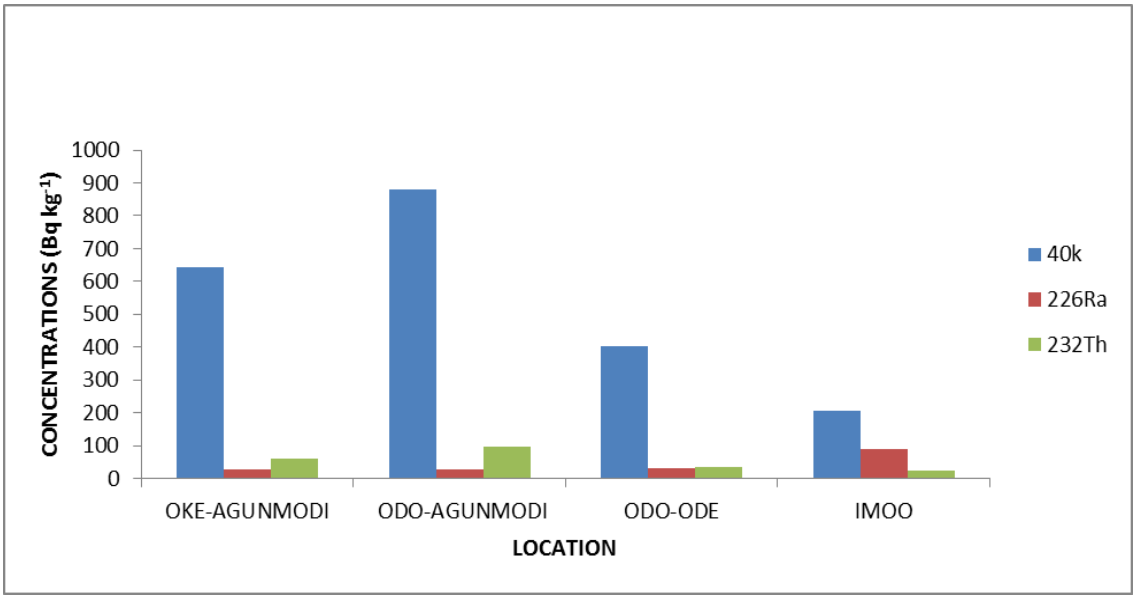

Fig 2: Distribution of Radionuclides in sediment samples.

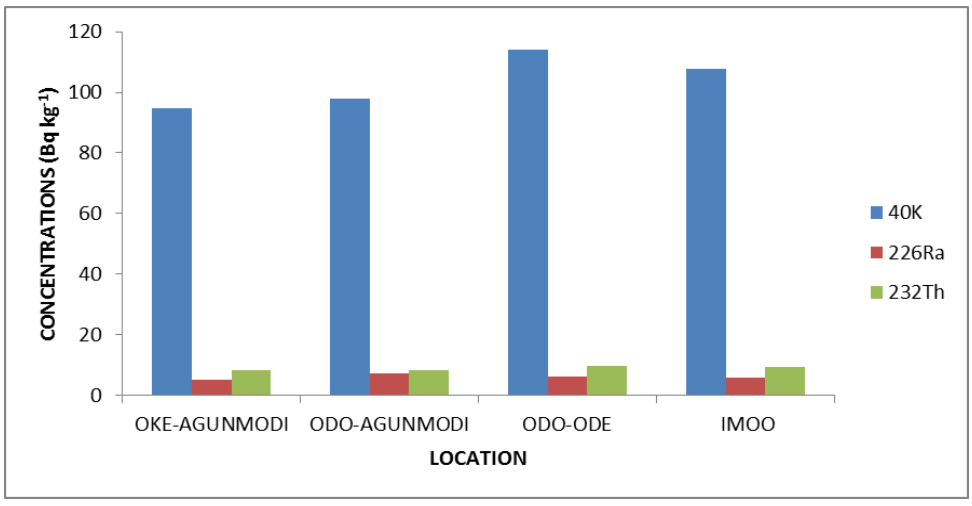

Fig 3: Distribution of Radionuclides in water samples.

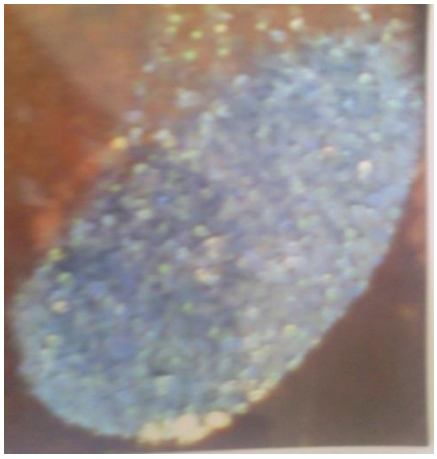

Fig 4a: Residue of gold and iron.

Fig 4b: Stone found in some of the pits

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