

Determination of natural radioactivity levels and resultant radiological hazard from soil samples around gold mining area in Ijesa-land south-western Nigeria.

ABSTRACT

Activity concentration levels and some radiological parameters owing to natural radioactivity in the soil samples randomly obtained in gold tailing sites in Ijesa-land Southwestern Nigeria were analyzed. Analyzed results from 30 soil samples using multi-channelled NaI gamma detector show that the activity concentration of ^{233}U ; ranged from 17.42 ± 0.54 to $49.89 \pm 2.42 \text{ BqKg}^{-1}$; ^{232}Th ranged from 22.10 ± 4.70 to $76.55 \pm 7.44 \text{ BqKg}^{-1}$ and ^{40}K ; ranged from 307.00 ± 23.90 to $693.58 \pm 51.50 \text{ BqKg}^{-1}$ in the samples were measured. The mean absorbed dose rate calculated was marginally higher than the world average of 59 nGyh^{-1} . The average annual effective dose rate (outdoor) for all samples ranged from $0.08 + 0.11 \text{ mSv.y}^{-1}$ with a mean of 0.07 which stays far below the permitted worldwide limit of 1 mSv.y^{-1} by International Commission on Radiation Protection (ICRP 2010). The calculated hazard indices for external and internal remained to be lower than unity signifying that mineworkers and individuals active in such area are not exposed radiation hazard.

Keywords: *Gamma, Spectrometry, Ijesha-land, Radionuclides, Radium, activity.*

1. INTRODUCTION

Ever since the formation of our planet, naturally occurring radioactive materials (norms) has remained a vital constituent of our environment [1]. Main sources of radiation to the animals or human existences naturally occur in the imaginable environment of rocks, soils and vegetation [2]. Radionuclide's like ^{40}K , members of ^{238}U , and ^{232}Th decay chain which are long-lived; are component of the earth's crust altogether are known as primordial radionuclide's [3]. According

to[4], radioactivity is everywhere and the distribution of these radionuclides depends on the main material distribution. Radiations from U, Th series and K are the main causes of the external gamma irradiation [4]. Also, exposure from natural radiation is of two major sources; terrestrial and cosmological radionuclides which leads to external and internal radiation exposure to living creature[5]. Industrial and human activities like mining and exploration of oil may lead to a situation in which increases of radionuclides can become significantly sufficient to trigger radiological hazards[1,6]. Regular contact to radioactive sources increases the radiation hazard as a result of exposure to man from terrestrial sources. [1]. Radiation from gamma has adequate strength to eject electrons from their outermost shell in the materials they enter. The cumulative effect of this radiation after a long time, can result to injury of specific organ[7]. Increased radiation doses can damage the functioning of tissues or organs and completely alter the genetic make-up of an individual, cause cancer and even death[4]. Vaporizers, contaminated soil, water and food are ways in which radionuclides get to human. Continuing interaction with radionuclides or radioactive materials without precautions have been informed to trigger human well-being problem including hemorrhage, getting old early and decrease with short lifecycle, cancer of the blood, anemia, cancer risks and other cardiac problems [9]. Radiological hazards consequences from minor amounts of ionizing radiation remain the consideration of established debate in radioactivity shield [10]. Exploration of oil gas or gold mining has been found to increase the activities concentration of mainly radium (^{226}Ra), potassium (^{40}K), thorium (^{232}Th) and their daughters [11,12]. Also, investigations have shown that tailings from gold mines contains higher concentration levels of radionuclides than the normal soil [13] This is a sign that, gold tailing or wastes are potential threat in our environment like what is happening in Nigeria particularly Ijesa-land. This could lead to significant cause of exposure to the naturally occurring

radionuclides to the people living in the locality in the gold mining areas. In view of this, there is necessity for comprehensive environmental and radiological care and safety education for the people.

Nigeria is a country rich in gold. Deposits of gold are found in south and north western of the country. In the south-western Nigeria, Ijesa-land in Osun state have been found to have large deposits of gold. The average gold deposits in Nigeria from 2000 to 2018 was estimated to be 21370kg and it increases to all-time high in the first quarter of 2018 to 21400kg

Government's exploitational tendency, lack of employment and poverty ridden society have made large number of people to engage in local/crude mining occupation. These occupational artisanal miners are from socially and economically marginalized communities (pure earth, lens).

In the last three four decades, Ijesa-land in Osun-state, Nigeria have been having mining operations by licensed companies. However, in the last ten years there has been an upsurge in the activities of artisanal miners. Gold mining operation started in Ijese-land at Ilesa-west local government in 1950. Official mining operation were suspended in mid-1990s, illegal artisanal mining operations are still active in almost all the local governments rich in gold till date [14].

In this research work, the natural activity concentration, radium equivalent activity, absorbed dose rate, annual effective dose, external hazard index and internal hazard index were measured.

Finally, alpha index was also assessed.

2. METHODOLOGY

2.1 Area of Study

Ijesha-land is located at latitude 8.92°N and longitude 3.42°E , situated at the middle of forest zone of the Yoruba nation southwest of Nigeria.[26] Effonridge separate the Ijesha from the Ekitistoward their east and to the west are Osogbo, Ado Ekiti and Ede. Ijesha land is currently made up of six local government areas in Nigeria. They are Atakumosa West, Atakumosa East, Oriade, Obokun Ilesha west and Ilesha East. Figure 1 below shows the map of Ijesha-land in the present-day Nigeria.

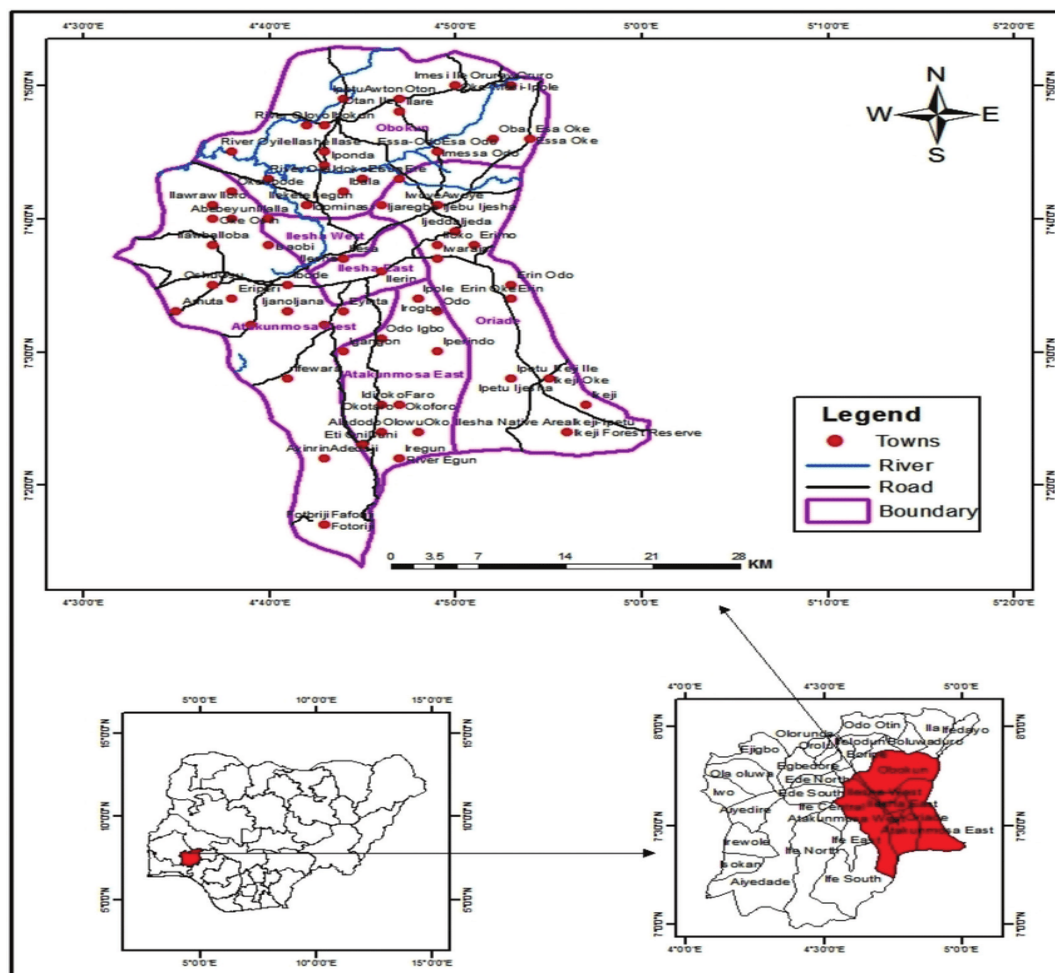


Figure 1: Showing the map of Ijesha land in Osun State Southwest of Nigeria

2.2 Sample collection, preparation and analysis

A total number of thirty (30) samples of soil tailings of 500g were taken randomly from where artisanal mining operation were predominant across the entire Ijesa land. The procedures involved in preparing the samples includes; careful gathering of soil sample from the mining sites, air drying at regular room temperature at the laboratory/workshop followed by watchfully drying at a temperature of 50⁰C in a kiln to speed the drying procedure without loss of radioactivity from soil samples [15], breaking down of soil samples by mortar and pound to a level that can be sieved by 200m sieve. Thereafter, 200g of the soil samples were packaged in a well labelled plastic containers [16]. and left for 28 days in order to create the minimum secular equilibrium among the progenies of ²³⁸U and ²³²Th decay series. The well labelled samples were later taking to Environmental Lab of Department of Engineering Physics, Obafemi Awolowo University, Ile-Ife for activity concentration measurement.

2.3 Determination of Some Radiological Parameter

2.3.1.) **Radium Equivalent Activity (Rad_{eq})**. Evaluation of gamma radiation exposure as a result of interactions of diverse mixture of soil samples comes with associated risk. It offers a specific guide which describes the contribution of gamma from various mixture of ²²⁶Ra, ²³²Th and ⁴⁰K in the soil samples. The conditions for safe level/dose to general public are that the effective dose calculated should not be more than 1.5Gy/year Proposed by UNSCEAR, 2000.

Rad_{eq} soil be calculated through the relation (1) above by [17,].

$$\text{Rad}_{\text{eq}} = \text{Ra} + C_{\text{Th}} \times 1.43 + C_{\text{k}} \times 0.077 \dots\dots\dots 1$$

Where C_U , C_{Th} and C_k are the mean radionuclides concentration of ^{238}U , ^{232}Th and ^{40}K in $BqKg^{-1}$ respectively. Ra_{eq} must not be greater than $(370Bq. Kg^{-1})$ which corresponds to an effective of 1mSv for general public

2.32) Absorbed Dose Rate (D) – This is done by means of the mean activity concentration of ^{238}U , ^{232}Th and ^{40}K ($Bq. Kg^{-1}$) correspondingly in the soil sample giving in equation (2).

$$D = 0.462A_u + 0.604A_{Th} + 0.0417A_k \dots\dots\dots 2$$

Where D is the absorbed dose rate in nGy.h, A_u , A_{Th} and A_k are the activity concentrations of ^{238}U , ^{232}Th and ^{40}K , respectively. The coefficients of the dose are in unit, of $nGy.h^{-1}$ per $Bq. Kg^{-1}$ obtained from [1].report.

2.33) The Annual Effective Dose Equivalent (AED): - The annual effective dose rates is measured by means of conversion coefficient of $0.7Sv. Gy^{-1}$ proposed by [1].to change from dose absorbed in air to effective dose. The Outdoor occupancy factors of 0.8 was used in the calculation.

Equation (3) presents the equation used to calculating AED equivalent in the outdoor

$$AED_{out} (mSv.y^{-1}) = D (nGy/h) \times 10^{-6} \times 8760h/y \times 0.8 \times 0.7(Sv. G/y) \dots\dots\dots 3$$

Where D is the calculated dose rate in (nGy/h)

2.34 Radiation Hazard indices:

This is an alternative method to evaluate the extent of gamma ray radiation relationship with natural radionuclides in a particular soil sample

Equation representing H_{ext} and H_{int} are shown in equation (4) and (5)

$$H_{ext} = \frac{A_U}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \dots\dots\dots 4$$

$$H_{int} = \frac{A_U}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \dots\dots\dots 5$$

Where A_u , A_{Th} and A_k represent the activity concentrations in of ^{238}U , ^{232}Th and ^{40}K in the soil sample. External and internal hazard indices must not be up to unity for the external gamma radiation hazards to be considered insignificant [5].

2.35) **Alpha index (I)** – Using soil from the mining sites may result into many radiations hazard to the miners and dwellers because of breath of radon and its decay products which are mainly alpha emitter. The calculation of this index is by using equation (7) [18].

$$I_\alpha = \frac{A_{Ra}}{200} \dots\dots\dots 6$$

3. Result and Discussion

3.1 Activity Concentration: The results gotten after the measurements of the radioactivity concentration of the soil samples randomly picked from gold mining tailings in Ijesa-land southwestern Nigeria are obtained in table 1. The measured radioactivity concentration of ^{40}K was in the range of 307.00 ± 23.90 to $698.58 \pm 51.50 Bqkg^{-1}$ with an average value of $458.11 \pm 32.15 Bqkg^{-1}$, ^{238}U , from 17.42 ± 0.54 to $28.63 \pm 1.43 Bqkg^{-1}$ and ^{232}Th from 22.10 ± 4.20 to $76.65 \pm 7.44 Bqkg^{-1}$. The entire values measured were higher than the global limit (UNSCEAR, 2010) activity concentration except that of ^{238}U that was lower by value of $6.37 Bqkg^{-1}$. This may be due to moderately richness of potassium, uranium and thorium in

the soils and also rocks that are rich in phosphate, granite and salts that contain natural radionuclides like uranium, thorium and potassium.[19].

Table 1. The measurements of the radioactivity concentration of the soil samples randomly picked from gold mining tailings in Ijesa-land southwest Nigeria

S/N	Sample ID	K-40 Bqkg ⁻¹	U-238 Bqkg ⁻¹	Th-232 Bqkg ⁻¹
1.	S1	572.92 ± 42.24	36.17 ± 1.76	66.96 ± 6.50
2.	S2	557.88 ± 41.13	49.89 ± 2.42	76.55 ± 7.44
3.	S3	602.60 ± 44.42	42.07 ± 2.04	68.69 ± 6.67
4.	S4	572.35 ± 42.19	37.38 ± 1.82	58.05 ± 5.43
5.	S5	594.02 ± 43.79	37.80 ± 1.84	55.87 ± 5.43
6.	S6	614.99 ± 45.34	34.30 ± 1.67	44.96 ± 4.47
7.	S7	498.85 ± 36.28	39.45 ± 1.92	51.28 ± 4.98
8.	S8	698.58 ± 51.50	36.52 ± 1.77	63.28 ± 6.15
9.	S9	507.27 ± 37.40	32.56 ± 1.58	46.64 ± 4.43
10.	S10	578.98 ± 42.68	32.63 ± 1.58	46.64 ± 4.43
11.	S11	431.68± 25.61	23.48 ± 0.90	32.71 ± 2.64
12.	S12	485.62 ± 28.47	27.60 ± 1.04	43.73 ± 3.54
13.	S13	419.99 ± 24.62	23.86 ± 0.90	30.49 ± 2.46
14.	S14	574.40 ± 33.69	30.70 ± 1.16	52.04 ± 4.21
15.	S15	377.31 ± 22.12	22.43 ± 0.85	29.14 ± 2.36
16.	S16	431.85 ± 33.62	22.37 ± 1.27	35.04 ± 4.31
17.	S17	422.68 ± 32.91	33.28 ± 1.88	47.46 ± 5.84
18.	S18	435.91 ± 33.94	22.60 ± 1.28	43.27 ± 5.33
19.	S19	307.00 ± 23.90	21.19 ± 1.20	29.52 ± 3.63
20.	S20	339.31 ± 26.42	21.42 ± 1.21	31.40 ± 3.86
21.	S21	409.06 ± 24.22	24.63 ± 1.92	32.25± 5.68
22.	S22	515.09 ± 28.27	32.69 ± 1.01	57.83 ± 3.79
23.	S23	361.70 ± 19.02	20.30 ± 1.22	22.10 ± 4.70
24.	S24	503.58 ± 27.64	30.00 ± 0.92	42.38 ± 2.78
25.	S25	500.70 ± 27.48	20.85 ± 0.64	38.25 ± 2.51
26.	S26	388.14 ± 23.90	20.22 ± 1.26	25.04± 4.54
27.	S27	385.47 ± 27.46	19.72 ± 1.22	26.95 ± 5.18

28.	S28	311.47 ± 21.00	20.75 ± 1.90	25.63 ± 4.21
29.	S29	486.81 ± 26.72	17.42 ± 0.54	30.26 ± 1.98
30.	S30	357.20 ± 26.62	24.72 ± 2.83	25.63 ± 4.52
Average		458.11 ± 32.15	28.63 ± 1.43	42.70 ± 2.80
Global limit [24]		400	35	30

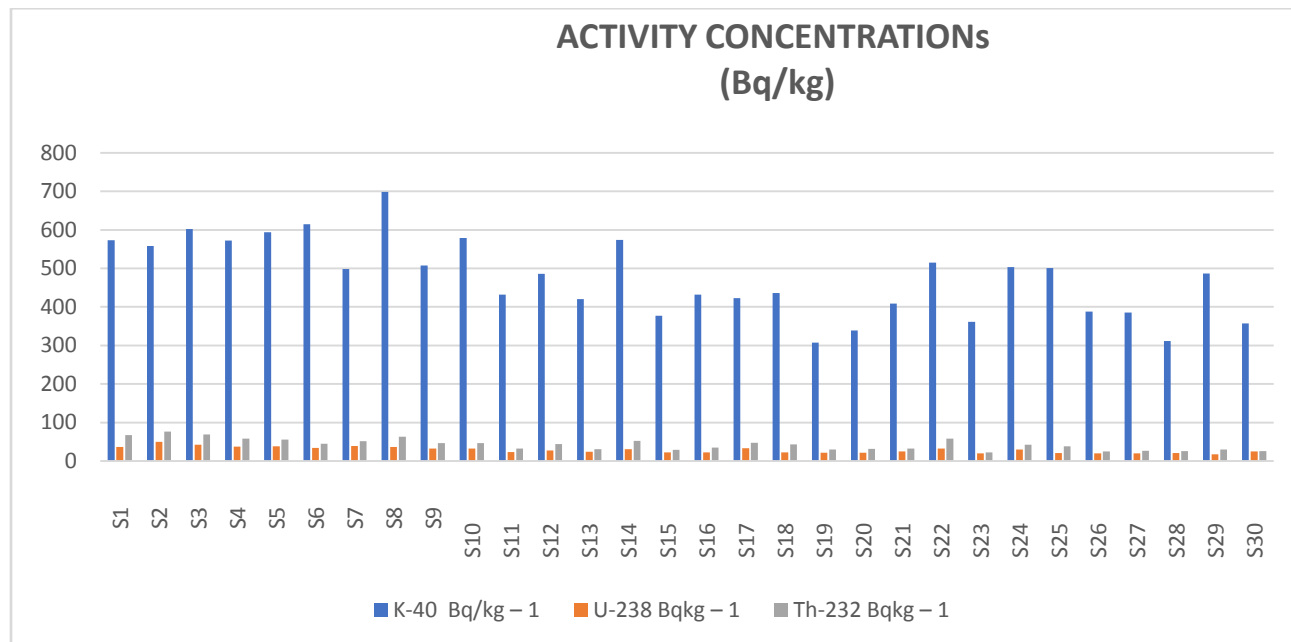


Figure 2. The graphical distribution of the measurements of the radioactivity concentration of the samples randomly picked from gold mining tailings in Ijesa- land southwestern Nigeria

Figure 2 shows the activity concentration of the radionuclides ^{40}K , ^{238}U and ^{232}Th in the soil samples. There were disparities in the measured activity concentrations of the radionuclides in all, with ^{40}K having maximum activity concentration in entire samples due to its comparative abundance and the geological location of the area [19].

In Table 3, comparison between the mean activity concentrations of this present study were made with similar works carried out in other parts of the world.

Table 2. Comparison between the mean activity concentrations of this present study were made with similar works carried out in other parts of the world

Country	$^{40}\text{K Bq kg}^{-1}$	$^{238}\text{U Bq kg}^{-1}$	$^{232}\text{Th Bq kg}^{-1}$	Ref
Iraq	876.746	28.64	14.18	[20].
Tanzania	652.36	42.59	35.48	[21].
Kenya	732.64	93.36	150.50	[22].
Egypt	586.00	79.00	44.00	[23].
Nigeria	458.11	28.63	42.70	Present Study
Worldwide	412	33	45	[24].

3.2 Radiological Hazard Assessment

Calculating radioactivity in an environmental sample is not only to evaluate the radioactivity concentrations of ^{238}U , ^{232}Th and ^{40}K but to similarly quantify the radiation dose interactions and the biological effects of such interactions with humans. Five (5) radiological parameters used to determine radiation effects on human were measured in this work. They are (i) absorbed dose rate in air, (ii) the annual effective dose (outdoor), (iii) radium equivalent (Rad_{eq}), (iv) both internal and external hazard index ($\text{H}_{\text{ext}} \& \text{H}_{\text{int}}$) and (v) alpha index (I_{α})

The aforementioned radiological parameters are presented in table 2. From Table 2, the estimated value of absorbed dose rate ranged from 37.81 to 92.55 (nGyh^{-1}) with an average of 59.75n (nGyh^{-1}) which is a bit higher than 58nGyh⁻¹ proposed by [24].

The annual effective dose rate as shown in the table 2 shows that it ranged from 0.08 to 1.1mSv.y⁻¹ by [25].

Radium equivalent calculated also ranged from 79.75Bqkg⁻¹ to 198.44Bqkg⁻¹ with an average of 124.89Bqkg⁻¹. These results indicate that the suggested radium equivalent concentration must be less than 370Bqkg⁻¹ for soil resources to be used for where people leave by organization for Economic Cooperation Development [26]. In this case it is safe dwellings.

The external and internal index as listed in table 2 show that the external hazard index varies from 0.21 to 0.42 with an average of 0.29, while internal hazard index also varies from 0.10 to 0.51 with an average of 0.36. Both have average values that are less than one (1) which is lower than the tolerable limit of 1mSvy⁻¹ [25].

Excess alpha radiation recorded also ranged from 0.11 to 0.26 with mean of 0.36 which is lower than the extreme permissible value of 1mSv.y⁻¹ which corresponds to 200Bqkg⁻¹. These results show that there stand no radiological threats for the inhabitants and mineworkers in the study areas.

Table 3. The annual effective dose rate ranged from 0.08 to 1.1mSv.y⁻¹

Sample code	Ra _{eq} (Bqkg ⁻¹)	D (n Gy.h ⁻¹)	AED _(out) (mSv.y ⁻¹)	I _α Bqkg ⁻¹	H _{in}	H _{ex}
S1	172.02	81.04	0.10	0.18	0.34	0.28
S2	198.44	92.55	0.11	0.25	0.10	0.34
S3	182.48	86.05	0.10	0.21	0.33	0.27
S4	160.45	76.78	0.09	0.19	0.49	0.40
S5	157.80	75.98	0.09	0.19	0.31	0.25
S6	141.65	68.64	0.08	0.17	0.49	0.29
S7	147.64	69.98	0.09	0.20	0.45	0.36

S8	175.92	84.22	0.10	0.18	0.38	0.32
S9	133.28	63.76	0.03	0.16	0.41	0.64
S10	144.60	69.42	0.09	0.16	0.35	0.24
S11	100.82	51.17	0.06	0.12	0.34	0.28
S12	124.13	59.41	0.07	0.14	0.42	0.34
S13	96.86	46.95	0.06	0.12	0.33	0.27
S14	145.32	69.57	0.08	0.15	0.49	0.40
S15	90.51	65.71	0.08	0.11	0.31	0.25
S16	102.70	49.72	0.06	0.11	0.35	0.29
S17	130.73	61.67	0.10	0.17	0.45	0.36
S18	114.98	54.75	0.07	0.11	0.38	0.32
S19	84.89	40.33	0.05	0.11	0.41	0.23
S20	134.96	43.01	0.05	0.11	0.35	0.24
S21	102.25	47.92	0.06	0.12	0.34	0.27
S22	151.44	71.51	0.09	0.16	0.51	0.42
S23	79.75	37.81	0.05	0.10	0.27	0.22
S24	125.85	60.46	0.07	0.15	0.43	0.35
S25	110.60	53.61	0.07	0.10	0.36	0.31
S26	85.91	40.65	0.05	0.10	0.29	0.23
S27	81.95	41.47	0.05	0.10	0.29	0.24
S28	85.38	40.02	0.05	0.10	0.29	0.23
S29	94.23	46.63	0.06	0.09	0.312	0.21
S30	88.84	41.79	0.05	0.12	0.31	0.24
Average	124.89	59.75	0.07	0.18	0.36	0.29
Global Limit	370	58	1	1	1	1
[24]						

Figure3 and 4 show the charts for absorbed dose rate and annual effective dose for all the 30 soil samples.

The charts showed that there are variations in activity concentration of soil samples from the same locality.

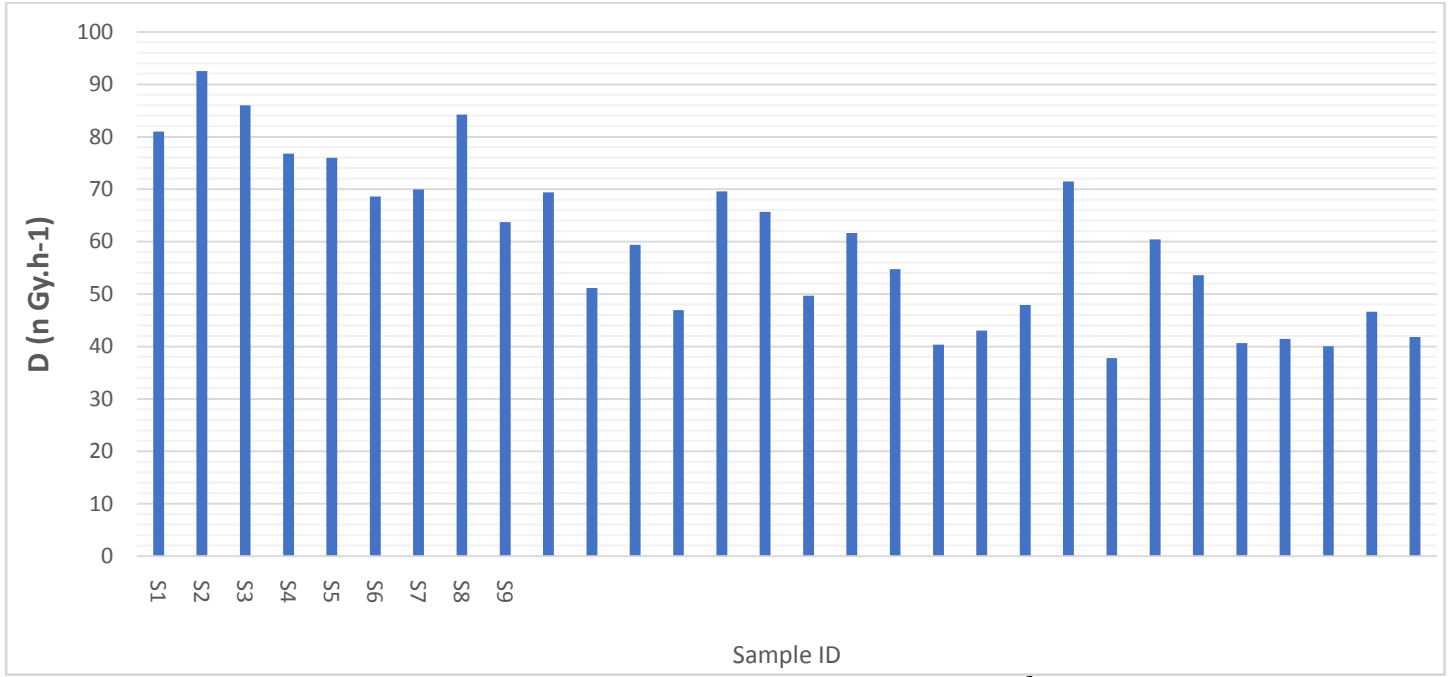


Figure 3. The graphical distribution of the absorbed dose rate of D (nGy.h⁻¹)

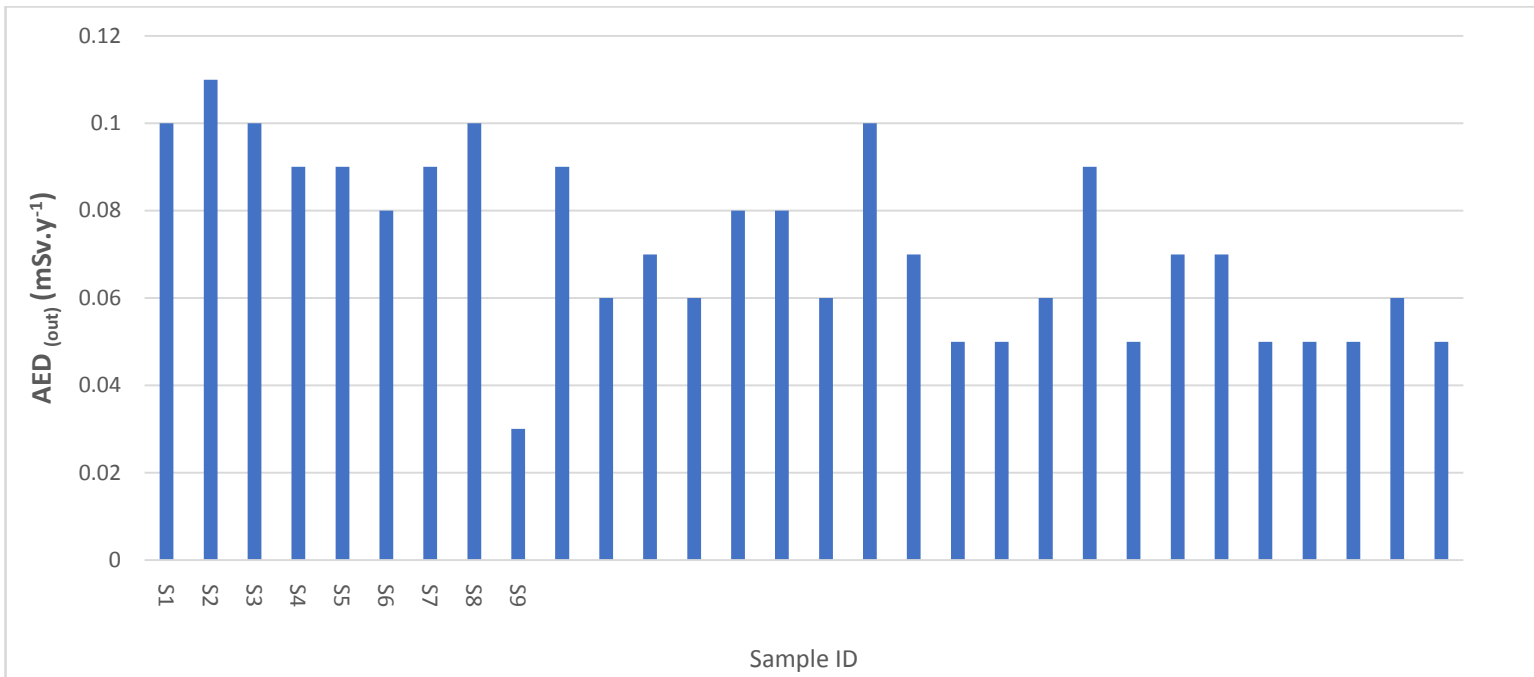


Figure 4. The graphical distribution of the annual effective dose rate ranged from 0.08 to 1.1mSv.y⁻¹

4. CONCLUSION

Radionuclides concentration of thirtysamples of soil from gold mining tailings from Ijesha-land with reference to ²³⁸U, ²³²Th and ⁴⁰K have been determined with the use of gamma spectrometry system with NaI (TI) detector. The results showed that the radioactivity concentration of natural radionuclides was not uniformly spread in the sample areas as a result of geological make-up of the of study areas, environmental factors and human activities.

The average value of absorbed dose calculated from all the soil samples are higher than the recommended value of 55nGhy⁻¹ by [25]. Average annual effective doses calculated in this study were lower than the recommended safety value of 1mSv.y⁻¹. Also, the calculated mean for external and internal hazard index for entirely sample are lower than recommended safe limit of 1. This outcome submits that the populations in such areas are not exposed to any radiological hazard.

Excess Alpha Radiation (I_{α}), ranged from 0.10Bqkg⁻¹ to 0.20Bqkg⁻¹ with a mean value of 0.18Bqkg⁻¹ which is below unity, which corresponds to 200Bqkg⁻¹. It can therefore be said that no radiological hazard is envisaged to dwellers/miners of the study areas.

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