

Radiological Safety Assessment And Physico-Chemical Characterization Of Soil Mixed With Mine Tailings Used As Building Materials From Oke-Kusa Mining Sites In Ijero, Nigeria.

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ABSTRACT: Soil samples potentially contaminated by mining by-products at Oke-kusa mines in Ijero, Nigeria were analyzed for naturally occurring radionuclides using gamma spectrometry. Also the physico-chemical determinations of parameters were based on standard methods of measurements. The highest average radioactivity levels obtained were $961 \pm 15 \text{ Bq kg}^{-1}$ (Mica site), $37 \pm 4 \text{ Bq kg}^{-1}$ (Kaolin site), $14 \pm 4 \text{ Bq kg}^{-1}$ (columbite-tantalite site) for ^{40}K , ^{226}Ra and ^{232}Th respectively. The average outdoor and indoor effective dose rates were far less than 1 mSv y^{-1} recommended limit for the member of the public. The PH of columbite-tantalite and Kaolin are acidic while that of the remaining samples are slightly alkaline. Soils pollution assessment was carried out using enrichment factor and the geoaccumulation index. The results of geoaccumulation index and enrichment factor reveal that soils of Oke-Kusa mines are uncontaminated with Mn, Fe, and Cu and Zn and these metals are depleted.

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INTRODUCTION

Natural radionuclides have been in existence in the environment since the formation of earth due to the long half-lives of radionuclides such as ^{238}U and ^{232}Th series and their decay products together with ^{40}K . These radionuclides have different sources: these include earth crust, rocks, soils, plants, water, sediments, minerals and air. The great interest expressed worldwide for the study of naturally occurring radionuclides and environmental radioactivity has led to the performance of extensive surveys in many countries of the world (UNSCEAR, 2000). The distribution of radionuclides concentrations reflects migration of uranium and thorium under surface soil condition. Thus uranium and thorium may migrate to the rivers, streams and surface soil in form of water soluble fulvatic complex compounds (Titaeva, et al, 1978) when the by-products are deposited on the surface soil. The exposure to naturally occurring radionuclide from the mining sites could be reflected in the contaminated soils samples collected from the area which are used for buildings as exposure to γ -radiation is not restricted to outdoor environment but can also occur in houses, offices and other working places (Xinwei, et al, 2006). Investigation of this can be useful for both the assessment of public dose rates and the performance of epidemiological studies, as well as to keep reference data records. Studying the naturally occurring radionuclides along with heavy metals in the in the area can provide an insight on deposition process that controls the geochemical characteristic of

soil and to help understand the influence of anthropogenic activities. In view of the fact that the associated internal exposure due to radon inhalation (Ngachin, et al, 2007) depend primarily on the geological and geographical conditions and appear at different levels in the soils of each region in the world (UNSCEAR, 2000); the objectives of the present study are:

i to determine the concentrations of natural occurring radionuclides in the surface soils mixed with mining by-products that are deposited around the mines.

ii to estimate the radiological dose that may be accrued to dwellers in buildings constructed with such soils.

iii to compare the dose rates with the existing value in the soil samples in Ijero from the areas that are far from the mines.

iv to assess the level of contamination of these soils by evaluating the geoaccumulation index and enrichment factor of heavy metals presence.

STUDY AREA

Ijero is situated in southwestern Nigeria between longitudes $4^{\circ} 30'$ and $5^{\circ} 00'$ East of the Greenwich Meridian and Latitude $7^{\circ} 15'$ and $8^{\circ} 15'$ North of the equator (UNAD, 2009). The area is entirely situated within the tropics (Fig 1). The climate is characterized by high temperature and heavy rainfall. It is drained by rivers and streams; among which are Okunrun, Ege and Olodo-Oye. The rivers and streams are used for bathing, washing and drinking. The geology of the

area is described as rock sequence that starts with the Precambrian basement. The rock sequence in this area are; pegmatites, aplites, granitic, rocks, charnockitic rocks, the quartzite series, gneisses and magnetites (Olarewaju, 1981). The solid minerals in the Oke-kusa (Mining Mountain) in Ijero are columbite-tantalite, tantalite ore (containing tantalum, iron and manganese), kaolin, mica, feldspar, flint, cassiterite and foundry sand and tin ore deposits.

METHOD AND MEASUREMENT

SAMPLE COLLECTION

A total of 100 samples were collected in all, composed of 20 soil samples from each of 5 sample sites situated on the mining mountain. The distance between each location was \approx 100-200 m depending on the topography. The samples were packed in plastics bags at the point of collection transported to the laboratory where they were divided into two parts for radioactivity and heavy metals analyses.

SAMPLE PREPARATION

At the laboratory the soil samples were air dried for 2-4 days, pulverized, crushed (Saad and Al-Azmi, 2002, Papatheodorou, et al, 2005) and made to pass through a 2 mm mesh sieved. A total of 250 g of the dried, grounded and sieved soil samples were put in plastic containers of uniform sizes. The containers were sealed with paper cello tape i.e. the containers were shielded hermetically and also shielded externally to ensure that all daughter products of uranium and thorium, in particular, radon isotope formed, do not escape. A time of 4 weeks was allowed after packing, which was sufficient time required to attain a state of secular radioactive equilibrium after their progeny (Alam, et al, 1997, Karahan and Bayulken, 2000, El-Arabi, 2005, Papaefthymiou, et al, 2007). It was ensured that the calibration standard reference samples (IAEA-375 from Austria) used for the detector efficiency calibration represented the samples to be counted. The sample has similar matrix with the soil samples counted at the spectrometer setting given from the standard recommendation (Faweya, 2010).

SAMPLE MEASUREMENT

NaI(Tl) scintillation, Bicon (serial no.3142) detector directly coupled to a pre-amplifier, a computer- controlled multichannel analyzer (MCA) was used to determine the concentrations of the primordial radionuclides (^{226}Ra , ^{232}Th and ^{40}K). The activity concentrations of natural radionuclides for all the 100 soil samples were determined for 10 h (36000 s) and the energy resolution of the detector was 8 % at 0.662 MeV of ^{137}Cs . The standard IAEA sources were used for calibration from the counting spectra.

Based on the resolution of the detector, radium content of the samples was determined from intensity of 1.765 MeV peak from ^{214}Bi ; thorium content was determined from 2.615 MeV gamma ray peak of ^{232}Th and potassium content was determined using 1.460 MeV decay of ^{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 using the expression (Noordin, 1999, Faweya and Babalola, 2010b).

$$C_c(\text{Bqkg}^{-1}) = \frac{A_{net}}{E\gamma tm} \quad 1$$

Where E in equation 1 is the detection efficiency, A_{net} is the net area under the peak, t is the counting time and γ is the gamma yield, that is the fraction of the γ -rays of the particular energy per disintegration and m is the mass of the sample.

PHYSICO-CHEMICAL MEASUREMENTS

PH Test using PH Meter

Approximately 2 g of each crushed sample was weighed out in duplicates and labeled A and B, then 10 ml of distilled water was added to A (1:1) while 20 ml of CaCl was added to B (2:1). Both were then stirred with separate stirring rods to avoid contamination and thereafter were left for 30 minutes. The PH meter was then calibrated with buffer solution of values 4 and 7. The electrode was then dipped into A and read after wish it was rinsed and wiped with tissue paper before it was then dipped into B and then read (Ademoroti, 1996). The procedure was repeated for all the samples and the average was taken (Table 3) to represent the site in consideration

Electrical Conductivity Using EC Meter

Two (2 g) gramme of each crushed sample was prepared and 10 ml of distilled water was added to each. Samples were left for more than 24 h undisturbed to allow the ions to be in solution. The meter was then standardized with distilled water. Each prepared sample was poured into the meter aperture and the reading was taken (Ademoroti, 1996). The mean EC for each site is shown in Table 3.

Phosphate

Approximately 2 g of each sample was weighed in 250 ml conical flask, and 20 ml of 0.025N HCl and 0.03N NH_4F solution was added, it was then shaken for 5 minutes on a reciprocal shaker at low medium. The sample was then filtered with a filter paper to get a supernatant solution. 3 ml of the resultant solution was then measured into a test tube using a pipette, and 3 ml of P.B (0.87N HCl and 0.87N NH_4) solution was added. 5 drops of 2.5 g of amino-2-sulphuric acid, 5 g of sodium sulphide (Na_2SO_3) and 146 g of sodium Meta bisulphide ($\text{Na}_2\text{S}_2\text{O}_5$) solution was then added as

an indicator. The sample was then left for 20 minutes. The sample was thereafter read with a calorimeter- a colour intensity reader that has been standardized in order to achieve substantial accuracy. The result was then subjected to a graphical interpolation and phosphorus was gotten. The procedure was repeated for all the samples and the average was taken to represent the site in consideration (Ademoroti, 1996, Adewuyi, et al, 2002).

Chloride.

Two (2 g) gramme of each sample was weighed out into conical flask and 5 ml distilled water was added. Samples were left for 12 h. 0.2 g of sodium bicarbonate ($\text{Na}_2(\text{CO}_3)_2$) was added to raise the PH to about 10. The resultant solution was then shaken with a reciprocal shaker for 30 minutes and filtered to get a clear solution. 2 ml of the resultant solution was then taken into conical flask and 1 ml of potassium Chromate was added as indicator. The solution was then titrated with silver nitrate to an orange colour endpoint. The chloride value was then calculated (Ademoroti, 1996, Adewuyi, et al, 2002. The average for all the sites was taken and recorded.

Heavy Metals Testing Using Atomic Absorption Spectrometer (AAS)

Laboratory apparatus were soaked in nitric acid and washed with tap and distilled water to remove any traces of cleaning agent. They were then dried and kept in a clean place (Radojevic and Bashkin, 1999). Sample was air dried and after homogenization using pestle and mortar, they are passed through a 2 mm mesh screen and stored in polyethylene bags (Sarva et al, 2007). Before the determination of heavy metals was conducted, 2 g of fine powder soil sample is digested using the high quality concentrated (70 % w/v) nitric acid, hydrogen peroxide (35 %) and hydrochloric acid (38 %) (Radojevic and Bashkin, 1999). The solution was then shaken for 1 hr using a reciprocal shaker, and then filtered using filter paper. The clear solution was then poured into sample bottles for reading in the Atomic Absorption Spectrometer. The samples bottle were then put into the AAS machine one after the other. The mean concentration of heavy metals detected for each site was recorded as seen in Table 3.

RESULTS AND DISCUSSION

RADIOACTIVITY ANALYSIS RESULTS

The three radionuclides, ^{40}K , ^{226}Ra and ^{232}Th have been detected and measured in all the samples while ^{137}Cs was not detected in any of the samples. The activity concentrations of ^{40}K , ^{226}Ra and ^{232}Th with the standard deviation (\pm SD) are shown in Table 1. Given the consideration made in the choice of peak for

analysis and the high shielded low-counting assembly used for the γ -counting, the combined uncertainties due to counting, background correction, and efficiency calibration was generally less than 25 %.

The concentration of the natural radionuclides as can be observed from Table 1 shows that the highest average concentration of ^{40}K was $961 \pm 15 \text{ Bq kg}^{-1}$. This was obtained from mica site. Kaolin recorded the highest mean concentration of ^{226}Ra of $37 \pm 4 \text{ Bq kg}^{-1}$ while the highest mean concentration of ^{232}Th of $14 \pm 4 \text{ Bq kg}^{-1}$ was obtained in columbite-tantalite site.

Since, no artificial radioactive substances were detected in any of the samples it suffices to say, therefore that the samples only exhibited natural activities. This is in agreement with the previous study which reported that the environment of Ekiti State is uncontaminated of any artificial radioactive materials (Ajayi, 2002). The overall activity concentrations of ^{226}Ra and ^{232}Th , in the area are lower than the world average crustal values of 35 and 35 Bq kg^{-1} for ^{226}Ra and ^{232}Th respectively (UNSCEAR, 2000) and higher than 400 Bq kg^{-1} for ^{40}K except flint site that is 153 Bq kg^{-1} for ^{40}K . The area can therefore be said to be radiologically safe from hazardous natural and artificial radiation sources.

These activity concentrations were only indication of levels of radionuclides present and do not relate the effect of such level on bio-system especially when these soils are used for building and agricultural purposes and to the miners excavating the mineral. The important quantity to assess when considering radiation risk to a bio-system is the absorbed dose rate. The absorbed outdoor dose rate, D_{out} (nGy h^{-1}) in air at 1m above the ground level due to the concentrations of radionuclides in the samples is calculated using a relation presented below (UNSCEAR, 2000, Jibiri, et al, 2007, Jibiri and Adewuyi, 2008, Faweya and Babalola, 2010a and 2010b)

$$D_{\text{out}} = a.C_{\text{Ra}} + b.C_{\text{Th}} + c.C_{\text{K}} + d.C_{\text{Cs}} \quad 2$$

where a is the dose rate per unit ^{226}Ra activity concentration ($4.27 \times 10^{-10} \text{ Gy.h}^{-1}$ per Bq.kg^{-1}), C_{Ra} is the concentration of ^{226}Ra in the sample (Bq.kg^{-1}), b is the dose rate per unit ^{232}Th activity concentration ($6.66 \times 10^{-10} \text{ Gy.h}^{-1}$ per Bq.kg^{-1}), C_{Th} is the concentration of ^{232}Th in the sample (Bq.kg^{-1}), c is the dose rate per unit ^{40}K activity concentration ($0.43 \times 10^{-10} \text{ Gy.h}^{-1}$ per Bq.kg^{-1}), C_{K} is the concentration of ^{40}K in the sample (Bq.kg^{-1}), d is the dose rate per unit ^{137}Cs activity concentration ($0.03 \times 10^{-10} \text{ Gy.h}^{-1}$ per Bq.kg^{-1}), and C_{Cs} is the concentration of ^{137}Cs in the sample (Bq.kg^{-1}). Since ^{137}Cs was not detected in any of the samples, the last term in Equation 2 was taken as zero.

The average outdoor dose rates for the samples are 43 ± 8 (columbite-tantalite site), 53 ± 7 (mica site), 23 ± 4 (Kaolin site), 16 ± 3 (flint site) and 42 ± 7

(feldspar site) nGy h⁻¹ as shown in Table 2. These values are lower than the worldwide average value of 60 nGy h⁻¹ for areas with normal radioactivity level.

The contribution of natural radionuclides to the absorbed dose rate in indoor air (D_{in} nGy h⁻¹) depend on the activity concentrations of ²²⁶Ra, ²³²Th and ⁴⁰K (Kurnaz, 2007, Isinkaye and Shitta 2009). This was estimated at 1m above the ground level for a room of 4 m x 5 m x 2.8 m in a typical masonry building that is built using soil samples from the mines (UNSCEAR, 1993, Markkanen, 1995, Ec, 1999, Papastefanou, et al, 2005) as follows:

$$D_{in} = 0.908C_{Ra} + 1.06C_{Th} + 0.0767A_K \quad 3$$

Where C_{Ra} , C_{Th} , and A_K are in Bq kg⁻¹. This formula assumes a wall of thickness of 20 cm and density of the structures to be 2350 kg m⁻³ (Turhan and Gunduz, 2008). The relative contributions to the absorbed dose rate in air from the activity concentrations of the three radionuclides are expressed in terms of their absorbed dose rate conversion coefficients which are 0.908 nGy h⁻¹ per Bq kg⁻¹ for ²²⁶Ra, 1.06 nGy h⁻¹ per Bq kg⁻¹ for ²³²Th and 0.0767 nGy h⁻¹ per Bq kg⁻¹ for ⁴⁰K.

The results in Table 2 revealed that the values of indoor dose rate ranged from 29 ± 5 (Flint site) to 98 ± 11 nGy h⁻¹ (Mica site). The highest mean absorbed indoor dose rate was 98 ± 11 nGy h⁻¹. Only the Mica site was above the worldwide average of 84 nGy h⁻¹ for soil as reported in UNSCEAR (2000).

The annual effective outdoor and indoor doses resulting from the outdoor and indoor dose rates value were calculated using the following relations:

$$E_{out} = D_{out}(nGy h^{-1}) \times 8760h \times 0.2 \times 0.7SvGy^{-1} \quad 4$$

$$E_{in} = D_{in}(nGy h^{-1}) \times 8760h \times 0.8 \times 0.7SvGy^{-1} \quad 5$$

Where E_{out} and E_{in} are the annual outdoor and indoor effective dose rates (m Sv y⁻¹), 8760 is the number of hours in a year, 0.7 Sv Gy⁻¹ is the quotient of the effective dose rate and absorbed dose rate in air, while 0.2 and 0.8 are outdoor and indoor occupancy factors.

The highest mean annual effective outdoor value 0.07 mSv y⁻¹ was obtained from mica site as seen in Table 2. This is higher than 0.04 mSv y⁻¹ obtained in the uncontaminated soil samples in Ijero (Ajayi, 2002) and in agreement with 0.07 mSv y⁻¹ world average (UNSCEAR, 2000). Mica site also had the highest mean annual effective indoor value of 0.48 mSv y⁻¹. This is higher than the world average of 0.41 mSv y⁻¹ (UNSCEAR, 2000). These values are less than the recommended limit of 1 mSv y⁻¹ for the members of the public (UNSCEAR, 2000, Abbadey and El-Arabi, 2006).

The magnitude of radiation exposure from natural building materials is strictly connected with the radium, thorium and potassium contents in the

building material and also on ventilation conditions; hence the Ra-equivalent concentration Ra_{eq} is a useful and instructive quantity which is an internationally accepted parameters that is applied to describe the suitability or otherwise of a building material for construction purposes. The radium equivalent in the samples was estimated using the relation given as:

$$Ra_{eq} = C_{Ra} + (C_{Th} \times 1.43) + (C_K \times 0.077) \leq 370Bqkg^{-1} \quad 6$$

The value of this parameter should be less than 370 Bq.kg⁻¹ so as to keep the annual radiation dose below 1.5 mGy y⁻¹ (UNSCEAR, 2000). The results obtained for Ra_{eq} are presented in Table 2. The mean radium equivalents obtained ranged from 33 Bq.kg⁻¹ (Flint) to 101 Bq.kg⁻¹ (Mica site). The results show that the recommended radium equivalent concentration of < 370 Bq.kg⁻¹ for building materials to be used for dwellings by OECD (Organization for Economic Cooperation Development) (Ahmad and Hussein, 1998) is applicable to the soils collected around the mine sites. The behaviour of radium equivalent activity is similar to that of radiation dose rate i.e. if the value of absorbed dose rate is high; the value of radium equivalent activity is also high and vice-versa.

The external hazard index (H_{ext}) is a criterion used for evaluation of external exposure to gamma radiation in the air. This has served as safety criterion in many countries of the world. It was proposed by Krisiuk et al (1971) and supported by Strandén (1976) and was used by Beretka and Matthew in 1985. In order to limit the external gamma radiation dose from building materials to 1.5 mGy y⁻¹ this index should be equal to or less than unity (i.e $H_{ext} \leq 1$). The maximum allowed value ($H_{ext} = 1$) corresponds to upper limit of Ra_{eq} (370 Bq.kg⁻¹) (Beretka and Matthew). This index was calculated using the relation given as follows:

$$H_{ext} = \frac{C_{Ra}}{370} + \frac{C_{Th}}{259} + \frac{C_K}{4810} \leq 1 \quad 7$$

The results obtained are shown in Table 2. The mean H_{ext} ranged from 0.09 Bq.kg⁻¹ (Flint site) to 0.27 Bq.kg⁻¹ (Mica site).

The use of soils from and around these mines for building may pose external radiation and internal hazard as a result of inhalation of radon and its decay products, which are predominantly alpha emitters by dwellers and miners. The excess alpha radiation due to radon inhalation originating from building materials is estimated using the relation below (Isinkaye and Shitta, 2009):

$$I_{\alpha} = \frac{C_{Ra}}{200} \quad 8$$

The mean value of I_{α} ranged from 0.04 (flint site) to 0.19 Bq.kg⁻¹ (kaolin site) and this is presented in Table 2. All these values for I_{α} are below the maximum permissible value of $I_{\alpha} = 1$ which corresponds to 200 Bq.kg⁻¹. It can therefore be said

that no radiological hazard is envisaged to dwellers of houses built by the soils and the miners working on those sites.

PHYSICO-CHEMICAL ANALYSIS RESULTS

Table 3 shows physico-chemical characterization of the samples. The hardness of the samples was moderate in values as shown by the presence of calcium, phosphate and chloride that were within the acceptable limit. The level of electrical conductivity, chloride and phosphate indicates an acceptable value for the soil samples. Howbeit, the PH of samples from columbite-tantalite and kaolin are acidic while that of the remaining samples are alkaline.

The assessment of soil enrichment with elements was determined by estimating the Enrichment Factors (EF) and index of Geoaccumulation (Igeo). The index of geoaccumulation (Igeo) has been used as a measure of bottom sediment contaminations since 1970s (Muller, 1969) and numerous researchers have employed it to assess the contamination of soils and sediments (Kwapulinski, et al, 1996, Miko, et al, 2000, Loska, et al, 2003, Habes and Nigem, 2006, Lokeshwari and Chandrappa, 20006, Sarva, et al, 2007, Haidarey, et al, 2010). It determines contamination by comparing current metal contents with respect to average shale value (Muller, 1969, Forstner and Muller, 1973, Forstner, et al, 1993). It is expressed as follows:

$$I_{geo} = \log_2 \left(\frac{C_n}{1.5B_n} \right) \quad 9$$

Where C_n is the measured concentration of heavy metal, B_n is the geochemical background value in average shale of element n (Turekian and Wed-epohl, 1961, Habes and Nigem, 2006) and 1.5 is the background matrix correction factor due to lithogenic effects. The index of geoaccumulation consists of seven grades; the highest grade (6) reflects 100-folds enrichment above background values. This according to Forstner (1993) is listed in Table 4. The result obtained using the mean concentrations from Table 3 are shown in Table 5. The results are compared to that in Table 4. It showed that $0.01 < I_{geo} \leq 0.40$ which indicates that soils in the mines are unpolluted with respect to Cu, Zn, Mn and Fe and safe when used for building. In this study, enrichment factor (EF) calculated to assess the level of contamination and the possible anthropogenic impact of the soils. Iron (Fe) was used to normalize heavy metals contaminants (Mucha, et al, 2003) in this study and it also served as a conservative tracer to differentiate natural components from anthropogenic components. This was estimated using the relation below (Ergin et al, 1991, Ghrefat and Nigem, 2006).

$$EF = \frac{\left(\frac{M}{Fe}\right)_{sample}}{\left(\frac{M}{Fe}\right)_{background}} \quad 10$$

Where EF is the enrichment factor, $\left(\frac{M}{Fe}\right)_{sample}$ is the ratio of metal and Fe concentration of the sample and $\left(\frac{M}{Fe}\right)_{background}$ is the ratio of metal and Fe concentration of background. Background concentrations of Cu (0.045 mg g^{-1} dry wt), Zn (0.095 mg g^{-1} dry wt) and Mn (0.850 mg g^{-1} dry wt) and Fe (46.70 mg g^{-1} dry wt) were taken from Turekian and Wed-epohl (1961) and Zhang and Liu (2002). The results are presented in Table 6. The results indicated that metals are entirely from crustal materials and natural processes. Since the values are below 1.5, these suggest that the sources of these heavy metals are not from anthropogenic.

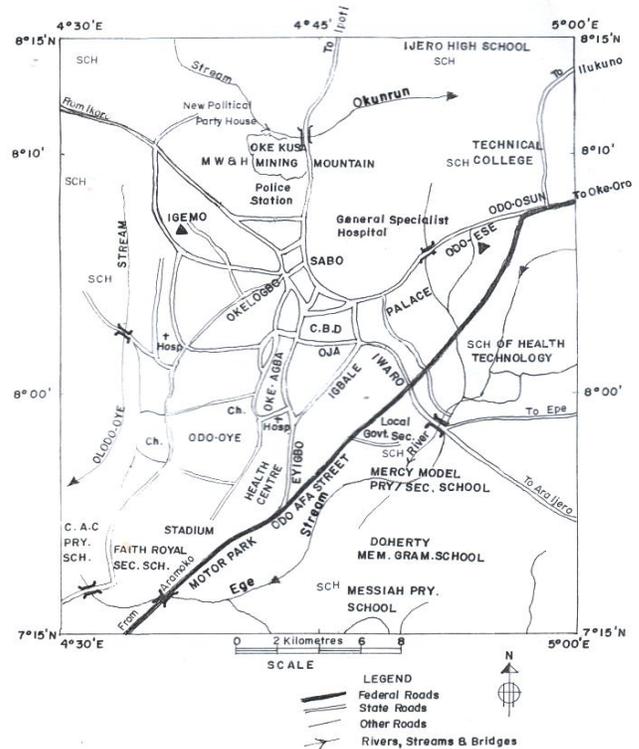


FIG. 1. 0 : THE STUDY AREA WITHIN IJERO TOWNSHIP SHOWING(OKE-KUSA)MINING MOUNTAIN, RIVERS/STREAMS & STREETS

Source: Local Government Secretariat, Ijero-Ekiti, Ekiti State/Dept of Geography & Planning Science, Cartographic Unit, University of Ado-Ekiti, (UNAD).

CONCLUSION

Since all radiological assessed parameters are within the acceptable limits, it suffices to say that Oke-Kusa mine and its environment are free from radiological burdens. All heavy metals in the samples are within acceptable limit (Eddy,et al, 2006) and do not pose a concern but the streams, the rivers and the aquatic animals in the city should be tested for consistency with the world standard.

Table 1: Activity levels of ^{40}K , ^{226}Ra and ^{232}Th in the samples.

Sample site	No of samples	Activity concentration (Bqkg^{-1})					
		^{40}K		^{226}Ra		^{232}Th	
		Range	Mean	Range	Mean	Range	Mean
Columbite-tantalite	20	325-1067	563±16	18-41	24±8	12-18	14±4
Mica	20	626-1041	961±15	8-14	11±3	7-14	11±3
Kaolin	20	250-962	432±14	9-25	37±4	3-4	4±0.3
Flint	20	98-338	153±14	3-11	7±2	6-16	10±2
Feldspar	20	431-948	770±13	9-21	13±4	5-8	7±1

Table 2: Mean radiation hazard parameters measured in the samples.

Sample site	D_{out} nGyh^{-1}	D_{in} nGyh^{-1}	E_{out} mSvy^{-1}	E_{in} mSvy^{-1}	H_{ext} Bqkg^{-1}	Ra_{eq} Bqkg^{-1}	I_{γ} Bqkg^{-1}	I_{α} Bqkg^{-1}
Columbite-tantalite	43±8	79±9	0.05	0.38	0.24	87	0.67	0.12
Mica	53±7	98±8	0.07	0.48	0.27	101	0.82	0.06
Kaolin	23±4	41±7	0.03	0.20	0.21	76	0.57	0.19
Flint	16±3	29±5	0.02	0.14	0.09	33	0.25	0.04
Feldspar	42±7	78±8	0.05	0.38	0.22	82	0.67	0.07

Table 3: The mean of physico-chemical quality parameters of the samples.

Sample site	PH H_2O	PH Cacl	conductivity μScm^{-1}	Cl mg^{-1}	P mg^{-1}	Cu	Zn (mg g^{-1})	Mn	Fe
Columbite-tantalite	6.60	5.50	0.72	10.80	15.10	0.01	0.06	0.05	8.22
Mica	8.30	8.20	0.24	28.75	32.14	0.03	0.08	5.70	43.74
Kaolin	6.30	5.70	0.72	26.80	31.10	0.01	0.10	0.09	15.10
Flint	7.70	7.70	0.44	11.81	27.52	0.04	0.05	0.12	38.20
Feldspar	7.60	7.10	0.01	12.59	16.28	0.02	0.09	0.13	39.00

Table 4: Geoaccumulation Index Classification (Forstner et al, 1993)

Soil Igeo	Class Igeo	Intensity of contamination
>5	6	Very strong
>4-5	5	Strong to very strong
>3-4	4	Strong
>2-3	3	Moderate to strong
>1-2	2	Moderate
>0-1	1	Uncontaminated to moderate
<0	0	Practically uncontaminated

Table 5: Geoaccumulation indexes of the metals in the soil samples.

Sample site	Cu	Zn	Mn	Fe
Columbite-tantalite	0.07	0.20	0.02	0.06
Mica	0.22	0.40	0.22	0.31
Kaolin	0.07	0.30	0.04	0.11
Flint	0.30	0.20	0.05	0.27
Feldspar	0.15	0.30	0.05	0.28

Table 6: Enrichment Factor (EF) of the soil samples.

Sample site	Cu	Zn	Mn	Fe
Columbite-tantalite	1.3	0.4	0.3	-
Mica	0.7	0.1	0.7	-
Kaolin	0.7	0.3	0.3	-
Flint	1.1	0.1	0.2	-
Feldspar	0.5	0.1	0.2	-

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