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# **Estimation of Natural Radionuclide Concentrations and Heavy Metal Contents due to Bitumen Exploitation at Gbeleju-Loda, Ode-Irele, Ondo State, Southwestern Nigeria**

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#### *Authors' contributions*

*This work was carried out in collaboration between all authors. Author IAO designed the study, performed the statistical analysis, wrote the protocol, and wrote the first draft of the manuscript. Author OPA managed the analyses of the study and prepared the samples for analyses. Author BSC managed the literature searches and performed heavy metal analyses. All authors read and approved the final manuscript.*

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# **ABSTRACT**

Bitumen exploitation has resulted in the destruction of organisms over a large expanse of land and water bodies. Thus, the aim of this present study is to collect soil and plant samples at bitumen exploitation affected areas of Gbeleju-Loda Ode-Irele, Ondo State, Southwestern Nigeria in order to estimate the natural radionuclide concentrations ( $^{40}$ K,  $^{238}$ U and  $^{232}$ Th) using a well calibrated NaI(TI) Scintillation Radiation Detector and heavy metal contents using an Atomic Absorption Spectroscopy (AAS) machine. The results obtained for natural radionuclide contents showed that <sup>40</sup>K were present in the range 320.46  $\pm$  45.16 to 543.23  $\pm$  14.08 Bqkg<sup>-1</sup>; <sup>238</sup>U were present in the range 24.33  $\pm$  11.41

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to 32.10  $\pm$  16.25 Bqkg<sup>-1</sup> while <sup>232</sup>Th were present in the range 12.36  $\pm$  8.12 to 20.41  $\pm$  3.50 Bqkg<sup>-1</sup> respectively in the soil samples. The results also showed that radionuclides were present in the concentrations of 145.16  $\pm$  12.40 to 280.35  $\pm$  27.32 Bqkg<sup>-1</sup> for <sup>40</sup>K, 10.62  $\pm$  4.91 to 22.24  $\pm$  3.51 Bqkg<sup>-1</sup> for <sup>238</sup>U and 6.34  $\pm$  5.28 to 9.58  $\pm$  3.07 Bqkg<sup>-1</sup> for <sup>232</sup>Th respectively in the plant samples. The results obtained for heavy metal (elements) contents strongly indicated the adverse effect of bitumen on soil and plants as the values obtained showed that the soil cannot support plant growth and this were visual in many dying plants in the environment and farmlands. The values obtained for natural radionuclide concentrations and heavy metal contents were higher than the values reported for several other locations while  $40K$  values reported were higher than the world average values.

*Keywords: Bitumen; radionuclides; heavy metals; exploitation; Ondo State; Nigeria.*

# **1. INTRODUCTION**

Bitumen is a sticky, black, and highly viscous liquid or semi-solid form of petroleum. It may be found in natural deposits or may be a refined product, and is classed as a pitch. The primary use of bitumen is in road construction, where it is used as the glue or binder mixed with aggregate particles to create concrete. Its other main uses are for bituminous waterproofing products, including production of roofing felt and for sealing flat roofs [1]. Bitumen is one of the richly deposited mineral resources in Nigeria and in some Africa countries, just like crude oil, it is found in Ondo, Lagos, Ogun, and Edo State [2]. However, the discovery of oil in the early 1960's in commercial quantity brought to a close the exploration of bitumen in Nigeria [3]. The exploitation sites of Bitumen were distributed mainly along a stretch beginning at the outcrop belt northeast of Lekki Lagoon in Ogun State and extend toward the southeast in Ondo State [2].

Bitumen exploitation and oil exploration has resulted in the destruction of organisms over a large expanse of land and water bodies [1,4]. These operations have not only caused degradation to the environment and destroyed the traditional livelihood of the region but have also caused environmental pollution which has affected weather conditions, soil fertility, waterways aquatic habitats and wildlife. Ondo state has the largest coastline among oil producing states and has the largest bitumen deposits in Nigeria and second most deposits in Africa [3]. Agriculture land degradation have resulted from shoreline erosion and landscape destruction associated with bitumen exploitation activities. Large chunk of coastal land has been made unfit for crop production thereby worsening existing economic hardship being experienced by local farmers [5].

Corollary damage has been done to the environment due to bitumen exploitation activities to the host communities, the environment has been denigrated and desecrated thereby causing peasant farmers to have suffered loss of farm steads and social displacement. Bitumen exploited lands were reported to be radioactive as such the radioactivity levels in foods grown in such soils may be enhanced through the plantroot uptake of radionuclides and that all food sources combined exposes a person the more to radioactive substances [6]. Natural radioactivity is associated with natural sources such as Uranium deposits and natural gas fields [7].

At Gbeleju Lodan area of Irele, where the residents are predominantly small scale farmers with farming and fishing as the principal economic activities which are feasible in the community. The introduction of toxic chemicals into the sea from the bitumen sediments, massive migration of various fish species from coastal areas land had taken place and the water polluted creating health hazards to the inhabitants. The environmental degradation decreases the production of the natural resources such as farming and fishing) from which the petty farmers wrest their livelihoods. There is therefore the need to address these problems especially as it affects agricultural productivity in such areas. Estimating the level of radiation in the environment is important in implementing a suitable control for the sake of radiological protection and radiation exposure [6], since its known that no mineral resource is environmentally friendly when it comes to exploitation.

#### **2. MATERIALS AND METHODS**

# **2.1 Geographical Description of the Study Area**

Gbaleju Lodan is located within Irele local government area and is part of the bitumen deposit zone within Ondo state, Southwestern Nigeria. The study area is located within the geophysical grids of latitude  $6^035'12.6''N$  and  $004^048'17.6''E$  and longitude  $6^038'15.2''N$  and  $004<sup>0</sup>50'29.8''E$ . The study area has tropical wet and dry seasons, the predominant occupation of the inhabitants is farming in both crops and fishing.

# **2.2 Samples Collection**

10 soil samples were taken at depth 10 cm within Gbeleju Lodan area of Irele local government. The samples were taken based on land form between 10 m apart. Also 10 plant leaf samples were taken along with the soil samples within the farm lands [7]. The plant indicator parts taken for analysis are leaves of commonly consumed food such as cassava, yam, plantain and maize. At the collection point, all samples were wrapped in separate black plastic bags and were well labelled with a paper masking tape [8]. Sampling points were geo-located using geographical positioning system (GPS) to ensure consistency. The soil and sand samples were collected inside polythene bags and stored in an insulated boxlike container.

# **2.3 Samples Preparation for Specific (Radionuclides) Activity**

The soil samples were well mixed after removing extraneous materials such as roots, pieces of stones and gravels. Samples were then weighted and dried into an electric oven at 110ºC for 4 days until a constant dry weight was obtained. After crushing and mixing thoroughly, soil samples were shaken in a sieve shaker and were scaled in 200g each. Plant leaf samples were cleared by fresh water for removing the surface dusts and surface contaminations. All the plant samples were then dried under the sun and humidity condition for 2 days, they were then weighed in 200g each [9].

The plant samples were charred and then finally ground into fine powder, the ash samples were cooled at room temperature. The cooled ash samples were weighed. For radionuclide contents analysis, samples were kept in radon tight containers for a minimum of 4 weeks so as to reach secular equilibrium between radon and its daughter nuclei [10].



**Fig 1. Geological map of Southern part of Ondo state showing the study area**

The radionuclide activity determination of the soil and plant samples were measured using a wellcalibrated Sodium Iodide doped with Thallium NaI(TI) with a well shielded detector coupled to a computer resident quantum MCA2100R multichannel analyzer.

The 1460 KeV gamma radiation was used to determine the concentration of  $40K$  in the samples, gamma transition energy of 1764.5 KeV was used to determine the concentration of <sup>238</sup>U while gamma transition energy of 2614 KeV was used to determine the concentration of <sup>232</sup>Th. The efficiency calibration of the detector was done using a reference standard mixed with source traceable to Analytical Quality Control Service (AQCS, USA) which has certified activities of the selected radionuclides. The activity of the radionuclide in the source that is emitting γ ray is expressed as:

$$
S = \frac{(N_p)_E}{(P_\gamma \lambda_\gamma)_E} \tag{1}
$$

Where  $(N_p)^{}_{\!\!E}$  is count per second under the photo peak due to gamma energy E,  $P_v$  is the gamma yield defined as the probability of the emission of gamma photon with energy E in each decay of the parent nuclide and  $\lambda_{\gamma}$  is the efficiency of the detector system.

The purpose of calibration is to evaluate  $\lambda_{\gamma}$  for range of energies covered by this present work.  $(N_p)^{}_{\!E}$  was obtained at each gamma energy by evaluating  $\lambda_{v}$  from the equation below:

$$
\left(\lambda_{\gamma}\right)_{E} = \frac{\left(N_{p}\right)_{E}}{s_{SD}\left(P_{\gamma}\right)_{E}}\tag{2}
$$

Also, at a constant geometry and constant γ-ray,  $\lambda_{\gamma}$  and  $P_{\gamma}$  are constant; hence the equation can be written as

$$
S = \mu(N_p) \tag{3}
$$

Where  $\mu = \frac{1}{P_Y \lambda_Y}$  which is the constant proportionality between source activity and the count per second under the photo peak.

#### **2.4 Samples Preparation for Heavy Metals Detection/Measurements**

The soil samples were air dried to remove the moisture content. After drying, the samples were crushed with a clean dry mortar and pestle, and then sieved through a 2-mm sieve to fineness. 3 g of sieved soil samples were weighed, and then digested with a mixture of 10 ml concentrated hydrochloric acid (HCL) and 3.5 ml concentrated nitric acid  $(HNO<sub>3</sub>)$ . The mixtures were left overnight without heating under the switch-on fume cupboard and heated for 2 hours at 104°C on the next day. Distilled water was added to the digested sample and then filtered with a filter paper and topped up to 100 ml volumetric flask with distilled water.

The solution was transferred into sampling bottles for analysis. The Plant samples were oven dried at 100°C for 24 hours and blended to fineness for easy digestion and then sieved through a 2 mm mesh sieve for easy digestion. 5 ml of 4:1 mixture of concentration  $HNO<sub>3</sub>:HClO4$ was added to 1 g of weighed plant with an analytical weighing balance. It was heated at a temperature of 105°C for 1 hour to dryness [11]. Then allowed to cool and made up to the mark of 50 ml volumetric flask with 1M HNO3. The solution was centrifuged using a centrifuge for 30 min then transferred into sampling bottles for analysis [12].

The concentration of these heavy metals in the soil samples and all digested plant samples were analyzed using an AAS (Atomic Absorption Spectrophotometer) - GBC Avanta PM. Ver 2.02. The equipment was calibrated before use and quality checks were also performed on the instrument by checking the absorbance after every five samples runs. To validate the procedure, the instrument was programmed and it carried out metal detection by displaying three absorbance readings and what was reported was the average. Blanks were also used for correction of background and other sources of error.

#### **3. RESULTS AND DISCUSSION**

#### **3.1 Activity Concentrations in Soil Samples from the Study Area**

The radionuclides concentration in the soil samples from the study area ranged from 320.46  $\pm$  45.16 to 543.23  $\pm$  14.08 Bqkg<sup>-1</sup> with a mean value of 421.67 $\pm$ 28.90 Bqkg<sup>-1</sup> for <sup>40</sup>K; 24.33  $\pm$ 11.41 to 32.10  $\pm$  16.25 Bqkg<sup>-1</sup> with a mean value of 26.35  $\pm$  32.58 Bqkg<sup>-1</sup> for <sup>238</sup>U and 12.36  $\pm$  8.12 to 20.41  $\pm$  3.50 Bqkg<sup>-1</sup> with a mean value of 16.09  $\pm$  21.02 Bqkg<sup>-1</sup> for <sup>232</sup>Th respectively.

All values reported for the present study were higher than the values reported for Okitipupa, a nearby town to the present study area [13]. The mean value of  $40K$  reported for the present study is higher than the world average of 410.00 Bqkg<sup>3</sup>, however the values reported for  $238$ U and  $232$ <sup>232</sup>Th were lower than the world average values of 35.00 and 28.00 respectively [6].

### **3.2 Activity Concentrations in Plant Leaves Samples from the Study Area**

The radionuclides concentration in the plant leaves samples from the study area ranged from

145.16  $\pm$  12.40 to 280.35  $\pm$  27.32 Bqkg<sup>-1</sup> with a mean value of 389.71 $\pm$ 22.48 Bqkg<sup>-1</sup> for <sup>40</sup>K; 10.62  $\pm$  4.91 to 22.24  $\pm$  3.51 Bqkg<sup>-1</sup> with a mean value of 24.78  $\pm$  31.92 Bqkg<sup>-1</sup> for <sup>238</sup>U and 6.34  $\pm$ 5.28 to 9.58  $\pm$  3.07 Bqkg<sup>-1</sup> with a mean value of 15.45  $\pm$  24.29 Bqkg<sup>-1</sup> for <sup>232</sup>Th respectively.

The activity concentration in the plant leaves were in the order Yam>Cassava>Maize>Plantain for all radionuclides. The values reported were higher than the values reported for yam, cassava, water leaf and bitter leaf for  $40$ K,  $238$ U and <sup>232</sup>Th respectively in Okitipupa [14].



<b>Samples</b>	<b>GPS location</b>	$\sqrt[40]{K}$ (BqKg <sup>-1</sup> )	$238$ U (BqKg <sup>-1</sup>	$\frac{232}{2}Th(BqKg^{-1})$
S1	$6^{0}33'43.5''N, 4^{0}50'54.3''E$	343.18±16.43	24.46±13.32	12.67±41.36
S <sub>2</sub>	$6^{0}33'47.1''N$ , $4^{0}51'18.9''E$	320.46±45.16	10.18±19.88	10.18±19.88
S <sub>3</sub>	$6^034'07.8''N$ , $4^051'56.1''E$	411.39±16.32	26.08±43.16	12.36±8.12
S4	$6^{0}34'15.6''N$ , $4^{0}52'42.3''E$	382.73±24.06	32.10±16.25	15.46±82.10
S5	$6^{0}34'34.3''N$ , $4^{0}52'19.7''E$	348.33±12.68	31.61±12.06	14.35±19.73
S6	$6^{0}33'43.5''N$ , $4^{0}50'54.3''E$	$512.33 \pm 18.46$	$31.84 \pm 24.31$	$20.41 \pm 3.50$
S7	$6^035'21.9''N$ , $4^050'17.1''E$	543.23±14.08	26.42+24.16	18.75±31.08
S8	$6^035'23.3''N$ , $4^050'36.9''E$	484.34±32.41	28.13±54.16	16.32±19.12
S9	$6^035'20.3''N$ , $4^051'06.9''E$	402.16±45.80	24.33±11.41	20.34±16.19
S <sub>10</sub>	$6^035'15.9''N$ , $4^051'12.7''E$	468.52±11.36	28.36±41.16	$20.01 \pm 4.06$
Range		$320.46 - 543.23$	$24.33 - 32.10$	$12.36 - 20.41$
Mean		421.67±28.90	26.35±32.58	16.09±21.02

**Table 2. Results of activity concentrations of 40K, 238U and 232Th in plant leaves samples**

<b>Samples</b>	<b>Plant leaves</b>	$40K$ (BqKg <sup>-1</sup> )	$738$ U (BqKg <sup>-1</sup>	$232$ Th (BqKg)
S1	Plantain	160.93±10.43	8.34±35.24	$6.34 \pm 5.28$
S <sub>2</sub>	<b>Plantain</b>	183.17±53.48	$10.62 \pm 19.88$	10.18±19.88
S <sub>3</sub>	<b>Plantain</b>	411.39±16.32	26.08±43.16	12.36±8.12
S4	Maize	382.73±24.06	$32.10\pm16.25$	15.46±82.10
S5	Maize	348.33±12.68	31.61±12.06	14.35±19.73
S6	Yam	512.33±18.46	31.84±24.31	$20.41 \pm 3.50$
S7	Yam	543.23±14.08	$26.42 \pm 24.16$	18.75±31.08
S8	Cassava	484.34±32.41	28.13±54.16	$16.32 \pm 19.12$
S9	Cassava	402.16±45.80	24.33±11.41	$20.34 \pm 16.19$
S <sub>10</sub>	Cassava	468.52±11.36	28.36±41.16	20.01±4.06
Range		$320.46 - 543.23$	$24.33 - 32.10$	$12.36 - 20.41$
Mean		389.71±22.48	24.78±31.92	$15.45 \pm 24.49$

**Table 3. Comparing activity concentration values in the present study with the world average values**



#### **3.3 Heavy Metal Contents in Soil Samples from the Study Area**

The result strongly indicated the adverse impact of bitumen on soils of the studied area. Iron (Fe) element in the soil samples were found to be adequate in most of the soil samples except sample 8 that is high above the recommended 200ppm. Potassium (K) was found to be deficient in most of the soil samples and is only marginal in some as indicated in Table 4, Carbon (C) was found to be deficient and often at marginal levels. Zinc (Zn), Magnesium (Mg) and Copper (Cu) were found to be high in the soil samples above the recommended levels. The results were similar when compared with the values reported for Lodasa area of Irele [14]. The values reported for the study area indicated that soils in Gbeleju Lodan cannot support plant growth as this were also visual in many dying plants in the environment and farm lands. The pH values of the soil were lower than pH value 7.0, indicating that the soils are acidic as it ranges between 2.93 – 6.24, thereby causing retardation and chlorosis of leaves and dehydration of plants.

From Fig. 2, the chemical elements in soil samples ranges between 193.74 and 0.22 for Mg and P respectively. Comparing the result with the WHO minimum permissible level (*see* Appendix-A2), Potassium and Carbon were deficient while Zinc, Magnesium and Copper were found to be of high concentration. Only Iron (Fe) is adequate, the pH value of 4.11 was reported indicating that the soil is acidic.

# **3.4 Heavy Metal Contents in Plant Leaves Samples from the Study Area**

The results obtained for the plant leaves of yam, cassava, plantain and yam from the study area was shown in Table 5 as most of the elements were reported to be deficient. The deficient elements were Mg, N, K, and P while Fe while Zn was at marginal levels. Cu and Zn were either deficient in some plant samples and at marginal level in others. No sample indicated adequate Cu and Zn levels. The presence of these elements in the soil samples is relative to that present in plant samples. The effect of these heavy metals contents reported on soil and plants at the study area cannot be over emphasized as its degrading to lives in the environment.

From Fig. 3, Magnesium (Mg), Copper (Cu), Iron (Fe), Potassium (K) and Zinc (Zn) are highest in the maize samples and the least in the cassava samples though they are deficient when compared with the WHO minimum permissible level (*see* Appendix-A1). In some plants, Zinc (Zn) and Iron (Fe) are reported to be at marginal level.



**Fig. 2. Mean distribution of heavy metals in soil samples from the study area**



**Fig. 3. Mean distribution of heavy metals in plant samples from the study area**



# **Table 4. Results of heavy metals analysis of soil samples from the study area**

*Mean*  $(\bar{X}) = \frac{\sum X}{N}$ ; Where: X is the elements measured and N is the number of samples taken

# **Table 5. Results of heavy metal analysis of plant leaves samples from the study area**



*Mean*  $(\bar{X}) = \frac{\sum X}{N}$ ; Where: X is the elements measured and N is the number of samples taken

Having concluded that no mineral resource is environmentally friendly when it comes to exploitation, the same is true of exploiting bitumen deposit.

The following recommendations were made:

- i. Farmers in affected areas should be relocated and resettled at areas where they can earn living through farming.
- ii. Organic fertilizers should be used to boost soil fertility in order to reduce problems arising from acidity since soils at Gbeleju Lodan are reported to be acidic due to the impact of bitumen exploitation.
- iii. Crops that can survive the acidity of the soil should be planted in that area.
- iv. Government should put in place legislation to protect the rights of the inhabitants of the mineral deposit areas.

# **4. CONCLUSION**

Agriculture land degradation, high radioactivity level has resulted from shoreline erosion and landscape destruction associated with bitumen exploitation activities in most part of the world. These often makes agriculture land unfit for crop production. This study has attempted to estimate the natural radionuclide concentrations and heavy metal contents due to bitumen exploitation on soil and plants in Gbeleju Lodan area of Ode-Irele, Ondo state, Southwestern Nigeria.

It was confirmed that bitumen exploitation has affected the environment of the study area as reported. The soil at Gbeleju Lodan cannot support plant growth as evidence and affirmed due to the presence of many stunted and dying plants in the area this is because Zinc (Zn), Magnesium (Mg) and Copper (Cu) were found to be high in the soil samples above the recommended levels and that plants from the area are slightly radioactive especially for the presence of radioactive potassium that is above the recommended world average values.

The pH value of the soil ranges between 2.93- 6.24 which is acidic. Most of the elements analyzed were deficient or at marginal level in the plants causing destruction to the plants. The pH value of the water samples taken at different locations within the study area ranges between 3.85-5.70. Therefore, the effect of bitumen deposits and its exploitation has adverse effects on soil and plants of the study area. This is degrading to the lives in the environment.

# **COMPETING INTERESTS**

Authors have declared that no competing interests exist.

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#### **APPENDIX**



#### **Table A1. Elements concentration in plants (WHO minimum permissible level)**

#### **Table A2. Elements concentration in soil (WHO minimum permissible level)**



#### **Table A3. Activity concentration values (world average)**



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