Health Risk assessment of Radionuclides Contaminated Quarry soils using Chromolaena odorata from Ratcon Quarry Oyo State South West Nigeria

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Abstract

Radionuclides are quarry pollutants produced during the crushing of rocks and have been reported to cause severe health effects in the environment. This study assessed the health risk associated with *Chromolaena odorata* in soils polluted with radionuclides from quarry site in Ibadan, Oyo state. Soil and plant samples (10 each) were collected from different sampling points making (20 samples) in the quarry site and control. The soil and plant samples were quantified for radionuclides (^{238}U, ^{232}Th, and ^{40}K) using standard procedures, Gamma-ray Spectroscopy Coupled NaI Detector was used. Health risk assessments were estimated for radionuclides vis-a-vis external hazard index, internal hazard index, excess lifetime cancer risk (ELCR). Data obtained were subjected to descriptive (Mean ± SD) and inferential (Anova) statistics. The concentrations of radionuclides detected in soils were within the WHO allowable limits, except for ^{232}Th and ^{40}K which were slightly higher than the permissible limit. However, the health risk assessment values revealed that ELCR was higher in almost all the sampling points than the recommended value of 2.9×10^{-4}, this implies that there would be a risk of cancer due to radionuclides in the location.

1.0 Introduction

A quarry is a surface mine that generates large amounts of building materials for industrial use, such as limestone (gypsum) and gravel (Majid *et al.*, 2015). Quarry takes off usually alter landscape drastically, vegetation clearing disrupts the ecosystem, while surface hydrology, groundwater levels, and flow paths are mostly diminished (Oyinloye & Olofinyo, 2017). The type of rocks extracted, the surrounding environment, the topography of the location, and the size of the quarry all affect how a quarry operates in terms of the environment (Oggeri *et al.*, 2019). The negative effects of quarrying activities on human health and crop yield are mostly inversely proportional to rural livelihood. The blasting, crushing, and emission of harmful gases usually have severe effect on human health and well-being. Doubtlessly, the most controversial environmental impact experienced by people living near quarries is those caused by blasting. Oyo State is one of the granite rich states in Nigeria, this have attracted various investors to the state, greatly contributing to the revenue generation and growth of the state. However, the negative effects of such activities have been seen on the environment, which usually results to instant/accumulative, physical, chemical, biological, and health hazards (Ndejjo *et al.*, 2015).

Radionuclides are an unstable chemical element that releases radiation as they break down and become more stable. Environmental exposures to radioactive elements are found in almost all sedimentary elements within the ecological environment and are widely distributed. These radioactive materials are called Naturally Occurring Radioactive Materials (NORM), including ^{238}U and ^{232}Th, and the single decay ^{40}K radionuclides comprising the vast bulk of NORMs.

The primary consumers of radioactive emissions in the food chain during atmospheric discharge of radioactive particles are plants. The two common route of plant contamination are Roots absorption and airborne deposits of dispersed radioactive particles. To determine the risk level consequences on public

health, an exact assessment of such radioactive particles in generally consumed dietary products is necessary. Also, a little amount of radioactivity in the soil is released to plants. Plants take radioactive elements from the soil and feed them to humans. Among the most crucial is the soil-to-plant transfer factor metrics in measuring the level of radioactive emission in plants and its effect in adding to internal dosage.

This study aims to assess the health effect associated with radionuclide *in polluted soils using Chromolaena odorata* in Ibadan, Oyo State Nigeria.

2.0 Materials and Methods

2.1 Study Area

Oluyole Local Government was established in 1976, and the Council occupies about 629 square kilometers. Based on the 2006 population Census, its population is 202,725. The Agricultural products in the area include cocoa, citrus, cassava, and maize. It is located approximately on Latitudes 7º 3' 0" to 7º 3' 21" N and Longitudes 3º 42' 0" to 4º 3' 0" E. Oluyole is within the forest savannah eco-climatic zone with an average annual rainfall of about 1225mm and an average annual maximum and minimum temperature of 34.80^{O}C and 24.30^{O}C. The rainy season commences around the end of March and ends around the first week of November.

2.2 Sampling Techniques

Using Systematic sampling techniques, top soil samples (0-15cm) and plants were collected from 10 different points (A1-A10) and (B1-B10) at 400m intervals each making (20) samples. The control sample was collected in an undeveloped location in Oluyole LGA with no history of pollution. Samples of soil were air-dried at room temperature for several days, and stones, organic debris, and pebbles were removed before sieving. The samples were crushed, homogenized, and sieved through a 2mm mesh sieve to obtain a smaller fraction, which was then ground into a fine powder and kept in a glass jar before analysis.

2.3. Measurement of Activity Concentration

The method (Alexander & Samuel, 2022) was adopted to assess the concentration of radionuclides. Using Gamma-Ray Spectroscopy, the activity concentrations of ^{238}U,^{232}Th, and ^{40}K were determined. After drying in the sun, 1 gram from each sampling point was ground to a fine powder. An electric oven was used to dry the samples for 24–48 hours at 80^{O}C to evaporate off all the moisture content of the soil. The samples were brought to secular equilibrium between long-lived parent radionuclides (^{226}Ra and ^{228}Ra) and their short-lived ones by storing them in airtight plastic containers for one month. Radionuclides activity concentration in sample soils was determined by Gamma Spectroscopy.

The measurement system was made up of a scintillation detector sealed with a photomultiplier tube coupled to a Canberra series ten plus multi-channel analyzer through a preamplifier base (MCA).

NaI (Tl) detector with the model number 3M3/3 was used. At 0.662 MeV of ^{137}Cs, the detector has a resolution of around 8%.

The Radiochemical Centre, Amersham, England, prepared a reference standard source (IAEA-444) to calibrate the system's detecting energy. The concentration of ^{40}K in the various samples was ascertained using the 1460.75 KeV gamma of ^{40}K.

The ^{238}U concentration of in the samples was determined using the gamma transition of ^{214}Bi, which had an energy of 1764.5 KeV, and the concentration of ^{232}Th using ^{208}Tl, which had an energy of 2614.7 KeV. Data was collected after each sample had been counted for eight hours. The MAESTRO software application automatically looked for the peak, assessed the peak position relative to energy, and identified the radionuclides using a nuclide library. The activity concentration in the chosen units was displayed once the background count and net peak areas had been calculated. For 17 hours, the background-filled empty container was counted, and then the values were subtracted.

2.4. Radium equivalent activity (Raeq).

There are variations in the distribution of ^{226}Ra, ^{232}Th, and ^{40}K. However, Radium equivalent activity (Raeq) in Bq/kg was used to establish uniformity in radiation exposure to compare the particular activity of materials containing various concentrations of ^{226}Ra, ^{232}Th, and ^{40}K. The UNSCEAR, 2000 was used to compute the relation:

$$Raeq = C_{Ra} + 1.43C_{Th} + 0.07C_{K} \quad \text{..... (1) (UNSCEAR, 2000)}$$

Where C_{Ra}, C_{Th}, and C_K are, respectively, the activities of ^{226}Ra, ^{232}Th, and ^{40}K expressed in Bq/kg. The same gamma dose rate was assumed for the production of 370 Bq/kg of ^{226}Ra, 259Bq/kg of ^{232}Th, and 4810Bq/kg of ^{40}K when defining Raeq activity according to Eq. (1)

2.5. Absorbed dose rate

This was calculated using the equation below:

$$D = 0.462C_{Ra} + 0.604C_{Th} + 0.042C_{K} \quad \text{.....(2)}$$

Where:

D is the dose rate in nGy/h, and C_{Ra}, C_{Th} & C_K are the specific activities in (Bq/kg) for ^{226}Ra, ^{232}Th, and ^{40}K, respectively.

2.6. External hazard index, Hex.

The external hazard index is a parameter used to evaluate the radiological risks of materials due to external exposure to gamma radiation. The recommended value of H ex must be less than one mSv/y, and its expression is given by the following relation (UNESCO,2000)

This will be determined using the equation below:

$Hex = C_{Ra}/370 + C_{Th}/259 + C_K/4810$..... (3)

Where:

C_{Ra}, C_{Th}, and C_K are the specific activities in (Bq/kg) for ^{226}Ra, ^{232}Th, and ^{40}K, respectively.

2.7. Internal hazard index (Hin)

The internal hazard index is a matrix that assess the risks of natural radionuclides in building materials due to internal exposure to radon (^{222}Rn) and its daughter progenies by inhaling alpha particles. The following formula gives its expression:

$Hin = C_{Ra}/185 + C_{Th}/259 + C_K/4810$ ..... (4)

2.8. Excess lifetime cancer risk (ELCR)

The carcinogenic effects of gamma radiation due to ingestion, inhalation, and external exposure to radioactive sources are characterized by an estimation of cancer occurrence probability during a specific lifetime. This probability is determined by calculating the excess lifetime cancer risk (ELCR). Its expression is given by the following relation (IRSN, 2007):

ELCR = AEDE × DL × RF..... (5)

In this expression, DL is the duration of life (70 years), and RF is the risk factor for cancer, considering the stochastic effects. RF is equal to 0.05 Sv − 1 for the public (IRSN, 2007).

3.0 Results and Discussion

^{238}U, ^{232}Th, and ^{40}K activity concentrations were measured by the Gamma-ray Spectrometry method for various soil samples collected at 0-15cm depth in the quarry site. Specific activity concentrations of the natural radionuclides obtained are presented in Tables 1 and 2. The activity concentration of ^{238}U in the study area ranged from 15.06 ± 1.50 Bq/kg to 30.92 ± 1.10 Bq/kg. These values showed a lower world average Uranium-238 concentration of 35 Bq/kg. While the concentration of ^{238}U in the plant ranged from 2.95 ± 0.72 Bq/kg to 11.16 ± 1.04 Bq/kg, (Table 1).

^{232}Th activity concentration ranged from 62.34 ± 1.55 Bq/kg to 82.41 ± 0.12 Bq/kg. The ^{232}Th activity concentration in this study was two-fold higher than the world average concentration of 30 Bq/kg. The ^{232}Th concentration in the plant ranged from 8.02 ± 0.16 Bq/kg to 18.32 ± 0.32 Bq/kg (Table 2). Research evidence shows that inhaling thorium dust increases the risk of lung, liver, and pancreatic cancer. Individuals exposed to thorium also have an increased risk of bone cancer because thorium may be stored in bone (USEPA, 2015).

The ^{40}K activity concentration ranged from 421.07 ± 2.24 Bq/kg to 640.83 ± 2.57 Bq/kg. The mean activity concentration was slightly higher than the world average of 400 Bq/kg. The plant activity concentration for ^{40}K ranged from 21.17 ± 1.11 Bq/kg to 142.62 ± 0.11 Bq/kg. The sample locations were high in ^{40}K, which could reflect this locality's soil fertility. Potassium-40 is a health hazard that results in cell damage due to ionizing radiation from radioactive decay, with the potential to induce cancer subsequently.

The main natural radionuclides had been identified and quantified. The results showed ^{232}Th and ^{40}K predominance in the sampling sites. The distribution of ^{238}U, ^{232}Th, and ^{40}K in soil samples are expressed in Figs. 1,2 and 3.

Table 1

Activity Concentration of ^{238}U, ^{232}Th, ^{40}K in Soil Samples.

Sample Locations	**^{238}U(Bq/kg)**	**^{232}Th (Bq/kg)**	**^{40}K(Bq/kg)**
A1	25.54 ± 1.02	62.34 ± 1.55	530.46 ± 2.50
A2	26.67 ± 1.17	75.04 ± 0.87	424.41 ± 1.40
A3	22.87 ± 1.16	63.96 ± 0.19	434.63 ± 2.18
A4	15.06 ± 1.5	57.46 ± 1.42	425.21 ± 1.58
A5	19.12 ± 2.04	70.48 ± 0.51	526.07 ± 2.78
A6	18.36 ± 1.17	82.41 ± 0.12	640.83 ± 2.57
A7	27.78 ± 1.28	81.11 ± 0.5	526.51 ± 1.57
A8	30.92 ± 1.10	77.04 ± 0.10	552.71 ± 2.53
A9	29.74 ± 1.26	65.33 ± 0.26	436.39 ± 2.37
A10	25.46 ± 1.64	71.37 ± 0.0	421.07 ± 2.24
Mean	**24.15 ± 1.33**	**70.65 ± 0.55**	**492.03 ±2.17**
World Average (UNESCEAR,2000)	35	30	400

Table 2: Activity Concentrations of ^{238}U, ^{232}Th, and ^{40}K in Plant Samples.

SAMPLING LOCATIONS	^{238}U(Bq/kg)	^{232}Th (Bq/kg)	^{40}K(Bq/kg)
A11	8.02±0.02	14.04±1.28	105.14±1.18
A12	11.16±1.04	18.32±0.32	96.12±1.53
A13	3.89±0.13	8.46±0.20	21.17±1.11
A14	6.06±1.1	7.32±0.01	110.15±0.76
A15	3.28 ±1.04	13.50±0.52	119.00±1.67
A16	4.12± 0.01	9.53±0.05	142.62±1.67
A17	5.77±1.32	14.83±0.7	153.14±0.59
A18	6.95±0.34	5.60±0.50	123.11±1.43
A19	7.54±1.12	8.02±0.16	114.42±2.32
A20	2.95±0.72	12.33±1.11	109.04±1.45
Mean	5.98±0.6	11.20±0.50	109.39±1.30
World Average (UNESCEAR,2000)	33	45	420

3.1. Radiological parameters

This present study has shown that the study area has a relatively high concentration of ^{232}Th and ^{40}K. Thus, radiological risks were calculated using radiological parameters and values obtained were compared to international standards. Tables 3 show the results.

Radium equivalent Raeq calculated using the activity concentrations of ^{238} U, ^{232}Th, and ^{40} K varied between 144.76Bq/kg and 181.04 Bq/kg. The mean values of Raeq was 159.71 Bq/kg, which was below the threshold value of 370 Bq/Kg. This indicates that there was no harmful effect due to ionizing radiation in the study area.

Tables 3 show that the minimum value of the absorbed gamma dose rate (D_R) obtained was equal to 67.45ηGy/h. In contrast, the maximum value was equal to 85.17. The mean value of D_R in this study was 74.50ηGy/h, slightly lower than the recommended maximum limit value of 84ηGy/h. The annual effective dose equivalent (AEDE) values ranged from 0.37mSv/y to 0.52mSv/y, with a mean value of 0.46mSv/y, which is less than the recommended value of 0.48mSv/y (Table 3).

The annual gonadal dose equivalent (AGDE) ranged from 420.23 μSv/y to 650.20 μSv/y, and the mean value was 529.45μSv/y, which was far lower than the recommended value of 1000μSv/y. Internal hazard risk ranged from 0.39 to 0.57 with a mean value of 0.50. External hazard risk ranged from 0.35 to 0.50 with a mean value of 0.44. These values were below standard limits of 1.

ELCR values were higher than the recommended 2.90×10^{-4} in most sampling points. The radiological risk assessment revealed that the values of excess lifetime cancer risk (ELCR) were significantly higher than the recommended value. Hence, there is a risk of cancer in the location. This agreed with Beogo *et al.* (2019), who reported the average ELCR value higher than the recommended value in the study area with more cancer cases in the locality.

Table 3: Radiation Hazard Indices of Soil Samples

Sampling Locations	Raeq (Bq/Kg)	D_R (nGy/h)	AEDE (mSv/y)	Hin	Hex	ELCR (x10-4)	AGDE
A1	152.82	71.73	0.44	0.48	0.42	3.1	506.06
A2	163.69	75.47	0.46	0.53	0.45	3.2	529.34
A3	144.76	67.45	0.42	0.45	0.40	2.9	474.50
A4	126.99	59.52	0.37	0.39	0.35	2.6	420.23
A5	156.73	73.50	0.45	0.47	0.43	3.2	518.87
A6	181.04	85.17	0.52	0.54	0.50	3.7	602.42
A7	180.62	83.94	0.52	0.57	0.50	3.7	650.20
A8	179.78	84.03	0.52	0.55	0.50	3.7	581.70
A9	153.71	71.53	0.44	0.51	0.42	3.1	502.00
A10	156.99	72.64	0.45	0.52	0.43	3.2	509.21
Mean value	159.71	74.50	0.46	0.50	0.44	3.2	529.45
Worldwide	370	84	0.48	1	1	2.90	1000

4.0 Conclusion

The Activity concentrations of radionuclides in the soil's samples showed that ^{238}U had a lower value than the world average, while ^{232}Th and ^{40}K had higher values than the world average.

From the present study, ELCR was higher in almost all the sampling points than the recommended value of 2.9 X 10 − 4. This implies that there would be a cancer risk due to radionuclides in the location before phytoremediation. This study revealed that there is risk of cancer in the study area hence, there is need for proper monitoring.

Declarations

5.0 Funding

This research work does not receive any funding.

The authors declare that Ethics, Consent to Participate, and Consent to publish this work are not applicable.

7.0 Data Availability Statement

All data generated or analyzed during this study are included in this manuscript

Author Contribution

Adetola O.O conducted the field work including sample collections, Olayinka O.O (Professor) Prepared the methodology and also proof reading of the final manuscript, Okeyode C.I (Professor) did the physics calculations and also corrected the references.

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Figures

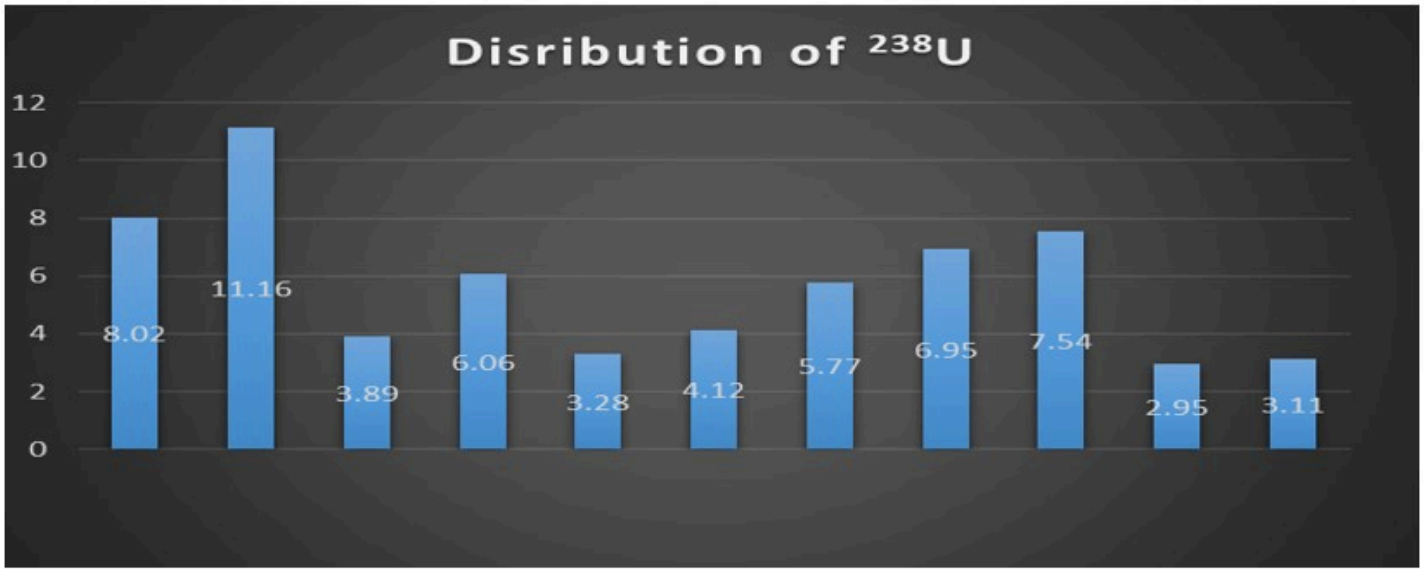

Figure 1

Distribution of ^{238}U in Soil Samples

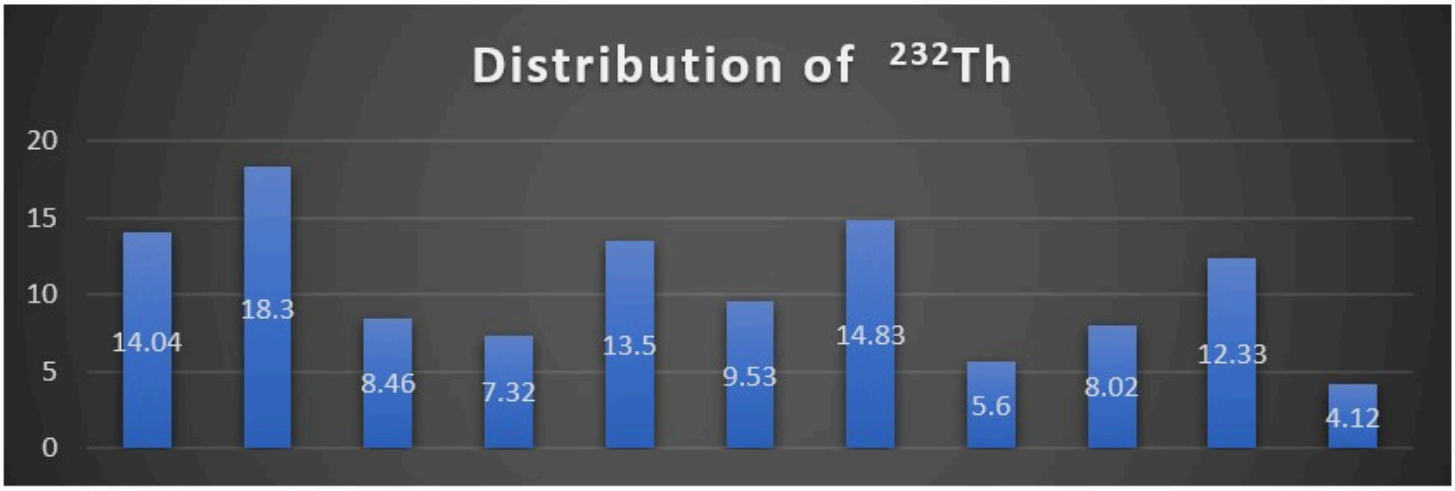

Figure 2

Distribution of ^{232}Th in Soil Samples

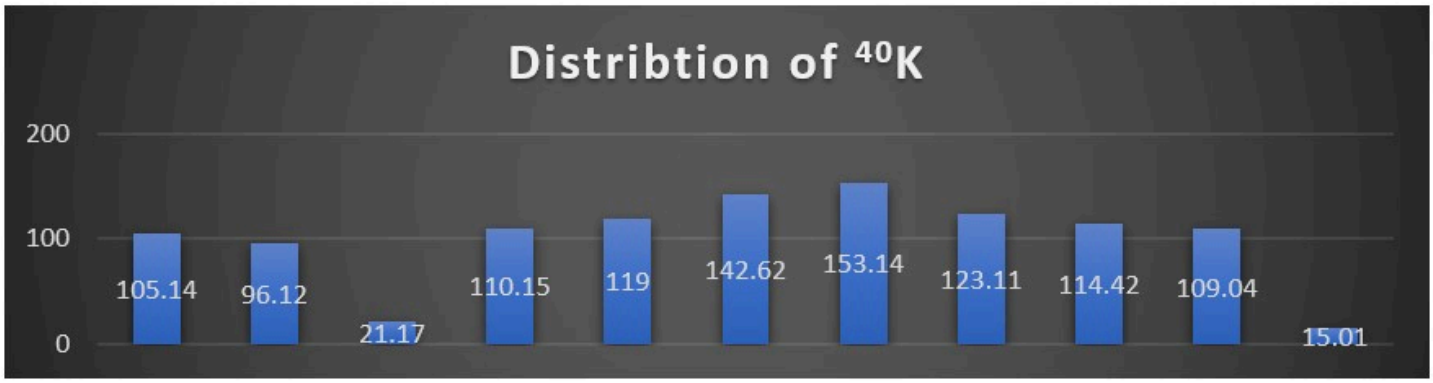

Figure 3

Distribution of ^{40} K in Soil Samples