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Assessment of Activity Concentration and Health Hazards of Radiation Exposure in Soil from Mamara Jidda and Waja Tin Mining Sites in Northern Nasarawa State, Nigeria

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Abstract

Natural background radiation exposure is an inevitable occurrence on Earth, and humans are exposed to radiation in their surroundings whether consciously or unconsciously. However, increased artisanal and illegal mining could harm the local environment and increase background radiation levels, affecting the community's overall health. This study assessed the activity concentration of primordial radionuclides (²²⁶Ra, ²³²Th, and ⁴⁰K) from soil samples obtained from Mamara Jidda and Waja tin mine sites in Nasarawa State, Nigeria, and its risk to human health. The gamma spectroscopy using a Sodium iodide-thallium activated detector was used to measure the activity concentrations of primordial radionuclides in the surface soils/sediments samples. The activity concentrations of ²²⁶Ra, ²³²Th, and ⁴⁰K for Mamara Jidda and Waja were found to be within the range of 4 - 18 Bq kg⁻¹, 41 - 105 Bq kg⁻¹, and 524 - 717 Bq kg⁻¹; 13 - 40 Bq kg⁻¹, 75 - 134 Bq kg⁻¹, and 591 - 882 Bq kg⁻¹ respectively. The average values show that ⁴⁰K and ²³²Th concentrations are above the recommended limit set by the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) in Mamara Jidda and Waja mine sites. The annual gonadal dose equivalent (AGDE), annual effective dose rate (AEDE), radium equivalent (Raeq), absorbed dose rate (D), and external hazard index (Hex) for Mamara Jidda and Waja were found to be 0.56 mSv yr⁻¹, 0.10 mSv yr⁻¹ ¹, 588.58 Bq kg⁻¹, 78.26 nGy hr⁻¹ and 0.46; 0.73 mSv yr⁻¹, 0.13 mS yr⁻¹, 742.74 Bq kg⁻¹, 103.72 nGy hr⁻¹ and 0.61 respectively. The AEDE and Hex are below the recommended safety limits. However, the high values of AGDE in both mining sites and D in Waja mine demonstrate that with continuous exposure to radiation, a significant genetic impact is eminent to the miners and inhabitants of these areas. Also, the Raeq values for both mines suggest that their soils are unsafe for use as building materials.

Keywords: Mining; natural radioactivity; activity concentration; radiological health hazard; potassium-40; radium-226; thorium-232.

I. INTRODUCTION

Human beings are subjected to radiation in their surroundings from sources such as cosmic rays, naturally occurring radionuclides in the air, plants, soil, and water, and manmade radioactivity. External exposure to gamma radiation comes from cosmic rays and naturally occurring radionuclides, while internal exposure occurs when people breathe in or consume it through food and water [1]. It is therefore evident that human health is associated with the quality of air, food, water, and the environment. Atomic radiations are limitless, and ionizing radiation exposure can result in radiation cataractogenesis, cancer induction, direct chromosomal alteration, indirect free radical generation, and bone necrosis, among other symptoms [2].

A 1999 report by the International Commission on Radiological Protection (ICRP) demonstrates that man is constantly exposed to ionizing radiation from naturally occurring radioactive materials, with the earth's crust being the source. According to the International Atomic Energy Agency (IAEA) estimates, naturally occurring radionuclides account for 85% of the doses that enter the environment, with the remaining 15% coming from nuclear and cosmic ray processes [3]. According to the World Nuclear Association (WNA), radiation protection standards assume that any radiation dose, no matter how small, presents a potential risk to human health [4, 5]. Hence, the as low as reasonably achievable (ALARA) concept, which aims to ensure that human radiation exposure is as low as practically possible, has been the norm.

The existence of naturally occurring radioactive materials (NORM) in the earth, as well as wastes and by-products from mining, could expose people to a technically elevated level of background radiation. Thus, the Nigerian government at both federal and state levels have exhibited their uneasiness towards artisanal mining activities and human health, most importantly, through the enactment of decrees, promulgating laws, and initiatives as demonstrated by the recent introduction of the previous administration's Presidential Artisanal Gold Mining Development Initiative (PAGMI) [6].

The continuous discharge of mining wastes and tailings into the environment poses a serious radiological risk to humans and the environment due to exposure to ionizing radiation from natural radioactivity, thus numerous studies have been conducted in Nigeria on the removal of large amounts of radioactively contaminated soil, solid mineral mining, and its relationship to the release of primordial radionuclides into the environment [7] – [26].

Similarly, studies conducted outside Nigeria, such as Yiangjiang, China, Orissa and Kerala, India, Ramsar, Iran and Guarapari, Coastal region of Espirito Santo, Morro Do Forro, Brazil [27, 28], have identified areas with high background radiation (HBRAs) related to the presence of monazite sand deposits, radium in soil/water, and radon in air. The effects of high-level radiation on several parameters in HBRA residents are not well documented, however, radiobiological studies conducted in HBRAs have given researchers a chance to study how long-term radiation affects people living in these locations [29].

References [30] and [31] provides further information on the radiological effects of mining activities around the world, with results compared with the recommended limit of 30, 35, and 400 Bq kg⁻¹ for ²³²Th, ²²⁶Ra, and ⁴⁰K respectively as given in [29].

Table I summarizes different studies on the radiological consequences of mining activities in Nigeria.

| | Activity Concentrations (Bq/kg) | | | | | | | | |
|-------|---------------------------------|-------------------|-----------------|---|--|---------------------|--|--|--|
| Study | ²²⁶ Ra | ²³² Th | ⁴⁰ K | In Situ Dose Measurement $(usv hr^{-1})$ | Mine Sites | Type of Sample | | | |
| [7] | N.A | 8175.2 | N.A | N.A | Jos tin Mine | Contaminated Soil | | | |
| [8] | N.A | 1680 | N.D | N.A | Jos tin Mine | Tin tailing | | | |
| [9] | 66 | 126 | 589 | N.A | Tin mining area of Bukuru and Bitsichi, Jos | Blocks | | | |
| [10] | N.A | 2.72 | 35.4 | N.A | Tin Mine in Bukuru Jos | Soil | | | |
| [11] | N.A | N.A | N.A | (5-80) | Tin Mining in Bukuru Jos | Soil | | | |
| [12] | (109-163) | (147-451) | (466-1062) | N.A | Bitsichi, Bukuru and Ropp localities Jos | Farm Soil | | | |
| [13] | N.A | (bdl-17) | (60-494) | N.A | Old tin Mine of Bitsichi, Jos | Soil and food items | | | |
| [14] | (19-30) | (27-41) | (83-129) | N.A | Old tin Mine of Bitsichi, Jos | Terrestrial food | | | |
| [15] | (109-470.6) | (122.7-2189.5) | (bdl-166.4) | N.A | Old tin Mine of Bitsichi, Jos | Crops | | | |
| [16] | N.A | (6.0-170.0) | N.A | N.A | Old tin Mine of Bitsichi, Jos | Soil | | | |
| [17] | N.A | (50-35800) | (30-670) | N.A | Old tin Mine | Tailing | | | |
| [18] | (2.0-2.7) | (0.3-1.2) | (5.7-7.3) | N.A | Igbeti Marble Mine | Marble | | | |
| [19] | (16.7-85.4) | (62.4-113.6) | (1315-1551) | N.A | Five quarry Industries in Ondo State | Granite | | | |
| [20] | - | 54.74 | 82.43 | N.A | Coal Steam, Enugu | Coal tailing | | | |
| [21] | 46.47 | 396.17 | 161.96 | N.A | Gura Topp, Jos | Soil | | | |
| [22] | 31.92 | 63.57 | 198.91 | N.A | Gababiyu Gold Mine, Minna | Soil | | | |
| [23] | (17.61-62.11) | (21.55-60.43) | (30.33-259.25) | N.A | Erena Mine Niger State | Soil | | | |
| [24] | N.A | 18.76 | 1168.13 | N.A | Bank of River Kaduna | Soil | | | |
| [25] | 2.08 | 47.23 | 382.01 | N.A | Yankandutse Gold Mine, Kaduna | Soil | | | |
| [26] | (14-62) | (12-43) | (268-721) | N.A | Kumar barite, Akiri copper, Azara barite, Ribi | Soil | | | |
| | | | | | barite, Adudu lead, Keana salt and Abuni Zinc | | | | |
| | | | | | Mine Nasarawa State | | | | |

Table I. Summary of studies on the Consequences of Radiological Mining Activities in Nigeria.

Note: N.A, N.D and bdl means Not Applicable, Not Detected and below detection limit respectively, while Readings in parenthesis are ranges and those that are not in parenthesis are mean values.

This study aims to assess the activity concentration and health hazards of radiation exposure in soil from Mamara Jidda and Waja tin mines in northern Nasarawa State, Nigeria.

A. Study Area

Mamara Jidda and Waja are local community settlements in Nasarawa North Senatorial District of Nasarawa State with coordinates 09° 01' 20.7"N, 008° 39' 55.5"E, 09° 01' 21.3"N, $008^{\circ} 39' 56.3$ "E and $08^{\circ} 55' 22.1$ "N, $008^{\circ} 38' 31.8$ "E, $08^{\circ} 55' 21.1$ "N, $008^{\circ} 38' 35.5$ "E respectively. There are currently plenty of artisanal mines in the senatorial district, with few active and accessible areas due to the security challenges in the area. Aside from farming, mining is the second largest source of income for the inhabitants of the senatorial district that is in proximity to the mines.



Fig. 1. Map of Nasarawa State Indicating Study Area [32].

II. MATERIAL AND METHODS

A. Materials

Materials used for this research include a Global Positioning System (GPS), book, pen, polyethene bags, plastic takeaway packs, soil and rock samples, candle, masking tape, matches, containers, vaseline, a rubber spoon, towel, weighing balance, and a Gamma-Ray Spectrometer (GRS).

B. Methods

1) Sample Collection

A total of twelve (12) soil samples were collected from Mamara Jidda and Waja mining sites. In each mining site, two points were randomly selected to provide an accurate sampling of the region. These samples were collected at the topsoil, 1 foot, and 2 feet below ground level respectively.*2)* Sample Preparation

The collected samples were rid of stones, pebbles, and shrubs and placed in a plastic takeaway pack, knotted tightly, and labelled to prevent contamination from other sources. The samples were dried under ambient temperature, grounded into a fine powder using mortar and pistil and sieved with a 2 mm sieve. The dimensions of 7.2 cm diameter and 6 cm height for mass analysis of bulk samples were adopted [33]. The samples were sealed with masking tapes, candle wax, and vaseline, to close the spaces between the lid and the container, to ensure that the radon does not escape. The prepared sealed samples were kept for 30 days to allow radon and its short-term offspring to attain secular radioactive equilibrium before measurements [34].

3) Radiation Measurements

Gamma spectroscopy was carried out at the newly commissioned low-background radiation laboratory at CERT, A.B.U Zaria, Nigeria. The detector system consisted of a 7.62 $cm \times 7.62 cm NaI(Tl)$ detector with an energy resolution of 72% at 661.16 keV peak of Cs-137 enclosed in a 6 cm thick lead-shield, cadmium-lined assembly with copper sheets to minimize the background radiation. A photomultiplier tube (PMT) is connected to the NaI(Tl) crystal. The setup includes an internal preamplifier and an external 1 kV source.

The requisition and analysis of the gamma spectra were done using a computer-based MCA software – Maestro for Windows by EG&G Ortec and Canberra model A65-B1 version 3.20. Calibrations of energy and efficiency were performed with IAEA standard spectrometric reference materials. The system was set at a working energy range of 0 – 300 keV. The samples were counted for 29, 000 seconds (8 hours) with an operating bias voltage of 900 V, Table II shows the spectrum energy window employed in the analysis.

Table II. Spectral energy windows used in the analysis

| | - P | 0, | |
|----------|-------------------|--------------|--------------|
| Element | Isotope | Gamma Energy | Energy (keV) |
| Analysis | used | (keV) | |
| K | ⁴⁰ K | 1460.0 | 1380 - 1500 |
| Ra | ²¹⁴ Bi | 1764.0 | 1690 - 1800 |
| Th | ²⁰⁸ T1 | 2614.5 | 2590 - 2700 |

4) Activity concentrations of ^{226}Ra , ^{232}Th , and ^{40}K in soil samples

The activity concentrations in each sample based on the peak area for each energy in the spectrum were determined using (1).

$$C = \frac{c_n}{c_{fk}} \tag{1}$$

Where *C* is the activity concentration of the radionuclide in the sample given in $Bq Kg^{-1}$, C_n is the count rate or count per second (CPS), which is given by *net count*/*live count*, and C_{fk} is the calibration factor of the detecting system.

5) Annual Gonadal Dose Equivalent (AGDE)

The annual gamma-ray doses absorbed by the rapidly dividing cells in certain sensitive organs of the body are evaluated by the AGDE for their degree of genetic implications. The gonads, active bone marrow, and bone surface cells were identified as the human organs of interest by the United Nations Scientific Committee on the Effects of Atomic Radiation [29]. Consequently, the AGDE in mSv yr⁻¹ was computed using the following relation considering the unique activity of ²²⁶Ra, ²³²Th, and ⁴⁰K [35].

$$AGDE = 3.09C_{Ra} + 4.18C_{Th} + 0.314C_k \tag{2}$$

6) Annual Effective Dose Equivalent (AEDE)

The annual effective dose equivalent (also called the outdoor annual effective dose - OAED) is estimated as the product of the gamma radiation dose, D(nGy/hr), dose

conversion factor of 0.7Sv/Gy, and occupancy factor of 20% (0.2) for outdoor exposure (duration the miners spend on the field in a year) using (3).

$$AEDE = D (nGyhr^{-1}) \times a (hrd^{-1}) \times b (dyr^{-1}) \times q \times d$$
(3)

The variables a, b, q, and d represent the number of hours in a day (24 hours per day), days in a year (365.25 days per year), outdoor occupancy factor (0.2), and conversion factor (0.7 Sv/Gy) between the absorbed dose in the air and the effective dose received by an adult.

7) Radium Equivalent (Ra_{eq})

The radium equivalent activity (Ra_{eq}) was computed using (4).

$$Ra_{eq} = C_{Ra} + 1.43C_{Th} + 0.77C_k \tag{4}$$

Where C_{Ra} , C_{Th} , and C_k are the mean activity of ²²⁶Ra, ²³²Th, and ⁴⁰K in Bq Kg⁻¹respectively.

United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) stipulates a threshold of 370 Bq/Kg for Ra_{eq} [23, 29]. This threshold activity corresponds to a gamma radiation dose of 1.5 mGy/yr.

8) Absorbed dose rate (D)

By employing conversion factors of 0.462, 0.604, and 0.0417 for 226 Ra, 232 Th, and 40 K to convert the activity concentrations into dose rates, the gamma absorbed dose rate (D) can be determined using (5) [29, 36].

$$D(nGy hr^{-1}) = 0.462C_{Ra} + 0.604C_{Th} + 0.0417C_k$$
(5)

Where C_{Ra} , C_{Th} , and C_k denote the activity concentrations of Ra, Th, and K, respectively, and $D(nGy hr^{-1})$ is the estimated absorbed dose rate in air. Values over the global average of 84 $nGy hr^{-1}$ cause harm to body tissues, organs, and cells [29].

9) External Hazard Index (H_{ex})

The risk posed by naturally occurring gamma radiation from primordial radionuclides is evaluated by the external hazard index. The primary goal is to determine whether the hazard index is smaller than unity. Equation (6) is used to compute this.

$$H_{ex} = \frac{226_{Ra}}{370} + \frac{232_{Th}}{259} + \frac{40_K}{4810} \le 1$$
(6)

III. RESULTS AND DISCUSSION

The activity concentrations of ²²⁶Ra, ²³²Th, and ⁴⁰K were calculated by converting them to conventional units using calibration factors after obtaining all the raw data.

The activity concentrations of 40 K, 232 Th, and 226 Ra for the soil samples taken at the topsoil, 1 foot, and 2 feet below ground level at Mamara Jidda and Waja mines ranged from 524.0±12.1 to 717.0±15.5 Bq/kg, 41.0±1.4 to 105.0±7.2 Bq/kg, 4.0±2.7 to 18.0±1.8 Bq/kg and 591.0±10.7 to 822.0±10.3 Bq/kg, 75.0±7.0 to 134.0±3.6 Bq/kg, 17.0±1.1 to 28.0±1.1 Bq/kg respectively as presented in Table III.

| 5105 | | | | | | | | | | |
|---------------------------------|----------------------|---------------------|----------------------|------------------|------------------|------------------|-------------------|--|-------------------|--|
| Activity Concentrations (Bq/Kg) | | | | | | | | | | |
| Mine Site | K - 40 | | | Ra – 226 | | | Th - 232 | | | |
| | Topsoil | 1 foot | 2 feet | Topsoil | 1 foot | 2 feet | Topsoil | 1 foot | 2 feet | |
| Mamara | 558±3.0 | 665±10 | 584±15.6 | 12±2.9 | 15±5.2 | 12±1.4 | 101±2.7 | 69±2.0 | 105±7.2 | |
| Jidda | 524±12.1 | 567±10.2 | 717±15.5 | 15±1.4 | 4±2.7 | 18 ± 1.8 | 52±1.9 | 98±4.2 | 41±1.4 | |
| Waja | `779±16.3 745+9.2 | 806±11.4 718+3.6 | 822±10.3 591+10.7 | 17±1.1 21+1.3 | 27±3.0 28+1.1 | 40±2.4 13+4.9 | 113±4.3 83+2.7 | 134±3.6 92+3.3 | 117±2.8 75+7.0 | |
| | 115_7.2 | 110_0.0 | 571=10.7 | 2121.5 | 202111 | 10_1.0 | 05_2.1 | <u><u><u></u></u><u></u><u></u><u></u><u></u><u></u><u></u><u></u><u></u><u></u><u></u><u></u><u></u><u></u><u></u><u></u><u></u><u></u></u> | 15=1.0 | |

Table III. Activity Concentrations of Primordial Radionuclides (²²⁶Ra, ²³²Th, and ⁴⁰K) at Mamara Jidda and Waja Mining

Table IV alongside Fig. 2 compares the average activity concentration of K-40, Ra-226, and Th-232 in Mamara Jidda and Waja mining areas of Northern Nasarawa State with the world averages. The activity concentrations of ⁴⁰K at both mining sites are higher than the world average. The activity concentration of ²²⁶Ra is lower than the world average in both the Mamara Jidda and Waja mining sites. Higher activity concentrations of ²³²Th were detected in samples from the two mines with values more than twice the world average.

Table IV. Average activity concentrations of NORMs at the two (2) mining sites.

| | | U | |
|------------|----------|---------|----------------|
| Mine Sites | K-40 | Ra-226 | Th-232 (Bq/Kg) |
| | (Bq/Kg) | (Bq/Kg) | |
| Mamara | 603±11.1 | 13±2.6 | 78±3.2 |
| Jidda | | | |
| Waja | 744±10.3 | 24±2.3 | 102 ± 4.0 |
| World | 400.00 | 35.00 | 30.00 |
| Average | | | |



Fig. 2. Average activity concentrations of K-40, Ra-226, and Th-232 in Mamara-Jidda and Waja mines in Northern Nasarawa State, Nigeria. (The dotted lines are the average world values for the primordial radionuclides).

The computed values of AGDE vary from $0.56 - 0.76 \, mSv \, yr^{-1}$. These values are higher than the recommended limits of $0.3 \, mSv \, yr^{-1}$ [29]. Thus, it may be concluded that radiation risks from these mines have a significant genetic impact.

The computed values of AEDE vary from $0.10 - 0.13 \ mSv \ yr^{-1}$. From the current study, both Mamara Jida and Waja mines have calculated AEDE lower than the recommended limits of $0.5 \ mSv \ yr^{-1}$ [29]. Thus, it may be concluded that radiation risks from these mines have no significant health effect.

The computed values of Ra_{eq} ranged from 588.85 – 742.75 Bq/kg. Hence, both mines have Ra_{eq} values higher than the safety limit of 370 Bq/kg set for this index. Therefore, the soils from these mines are unsafe to utilize as building materials.

The computed values for D vary from 78.26 – 103.72 $nGy hr^{-1}$. This implies that the Waja mining site has calculated D higher than the recommended limits of 84 $nGy hr^{-1}$. The quantity of different radionuclides in the soil determines how much of the absorbed dose rates are due to natural radionuclides [37].

The world average External Hazard Index (H_{ex}) is 1. Values higher than 1 may result in radiological risk to the inhabitants or people living in Mama-Jidda and Wajja. The computed values vary from 0.46 - 0.61. All the mines had H_{ex} values below the suggested value of 1. As a result, there is no radiological risk to the residents from external exposure.

The results of the radiation hazard indices of the mining activities in Mamara Jidda and Waja tin mining sites which include the radium equivalent, absorbed dose rate, the annual gonadal dose equivalent, annual effective dose equivalent, and eternal hazard index are presented in Table V below.

| Table | V. Mean | Values for | Radiological | Health Risk | Indicators | from Mar | nara Jidda | and Waja | mining | sites |
|-------|---------|------------|--------------|-------------|------------|----------|------------|----------|----------|-------|
| | | | U | | | | | | <u> </u> | |

| Mining Sites | Dose Rate $(nGy hr^{-1})$ | Ra _{eq} (Bq /Kg) | H _{ex} | $AEDE (mSv yr^{-1})$ | $\begin{array}{c} AGDE\\ (mSv \ yr^{-1}) \end{array}$ |
|---------------|---------------------------|------------------------------|-----------------|----------------------|---|
| Mamara Jidda | 78.26 | 588.85 | 0.46 | 0.10 | 0.56 |
| Waja | 103.72 | 742.74 | 0.61 | 0.13 | 0.73 |
| World Average | 84 | 370 | ≤ 1 | 0.5 | 0.3 |

IV. CONCLUSION

The activity concentration of primordial radionuclides and health hazards of radiation exposure of soil samples from Mamara Jidda and Waja mining sites in Northern Nasarawa State, Nigeria has been determined. The concentrations of ⁴⁰K and ²³²Th in both mines are higher than the recommended limits of 400 Bq/kg and 30 Bq/kg respectively. However, the activity concentrations of ²²²Ra for both mines are below the recommended value of 35 Bq/kg. The AGDE values vary from 0.56 – 0.76 $mSv yr^{-1}$ in both Mamara Jidda and Waja mines as against the recommended limit of $0.3 \ mSv \ vr^{-1}$ implying that continuous exposure to gamma-ray doses could genetically impact the miners and inhabitants of these areas. The Ra_{eq} values for the two mining sites range from 588.85 – 742.75 Bq/kg which is higher than the recommended safety limit of 370 Bq/kg, thus, soils from both mines are unsafe for use as building materials. The absorbed dose rate, D, for Mamara Jidda, is within the recommended safety limit. But the D for Waja mine is higher than the limit of 84 $nGy hr^{-1}$ which can cause harm to the body's tissues, organs and cells. The AEDE and H_{ex} for both mining sites are below the recommended safety limits of $0.5 \, m \, Sv \, yr^{-1}$ and <1 respectively. There is therefore no significant radiological risk to the residents from exposure associated with these indices.

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CONFLICT OF INTEREST

All authors declare that they have no conflicts of interest.

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