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**Research Article**

# Validity of Tin Mine Stream Sediments in the Construction of Residential Homes

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**Abstract:** Tin mining in the Jos Plateau, Nigeria has left the residents with a legacy of radioactively contaminated soils, polluted water supplies, scattered heaps of radioactive mine/mill residues and radioactive mine ponds/steams. The gamma radiation of sediment samples collected from the tin mine streams at Kokop, Zawan, Sabon Barki and Yelwa, which the residents used in building constructions, were measured. The samples were collected, dried under ambient temperature and sealed in plastic containers and the activity concentrations of $^{40}$K, $^{226}$Ra and $^{232}$Th measured in the laboratory using NaI (Tl) gamma-ray spectrometry technique with an accumulating time of 30,000seconds per sample. Similarly, elemental concentration levels of the samples were determined using XRF technique. The Kokop, Zawan, Sabon Barki and Yelwa measured range of activity concentration levels for $^{40}$K, $^{226}$Ra and $^{232}$Th were (324-955) Bq/kg, (307-461) Bq/kg and (851-1203) Bq/kg; (154-267) Bq/kg, (89-157) Bq/kg and (78-111) Bq/kg; (163-322) Bq/kg, (77-108) Bq/kg and (115-208) Bq/kg and (134-379) Bq/kg, (61-109) Bq/kg and (156-198) Bq/kg, respectively. The radiation hazards associated with these natural building materials were assessed using three radiological models, namely; "Radium Equivalent", "Mean Annual Effective Dose" and "Gamma Activity Concentration Index". In each case, there was an indication that Kokop sediment samples were not safe for use in the construction of residential houses and hence poses a significant source of radiation hazards to occupants of such buildings. The Kokop's mean recorded radium equivalent, mean annual effective dose and gamma activity concentration index were 2057.7±45.8 Bq/kg, 4.6 mSv/yr and 7.0, respectively. A total of 21 elements were detected in the sediment samples. Arsenic, the element of interest, was significantly high, much higher than the recommended acceptable level in soil to be used in construction of residential houses. This further justifies the unsafe use of the sediments in the construction of dwelling homes bearing in mind the activities of pica children who exhibit the hand- to-mouth habit and may probably be co-occupants as well.

**Keywords:** Activity concentration, contaminated sediment, hazards, mill tailings

## INTRODUCTION

Nigeria, in the twentieth century, was one of the world's major tin producing countries but production later faced a decline towards the end of the century. However, this did not imply that tin mining activities ceased completely in the mining areas. Besides tin, tin mining produces tin tailings, a by-product of rough concentrates of cassiterite. These tailings from tin mining industry were usually processed to extract valuable minerals such as columbite, ilmenite, monazite, zircon, xenotime and struverite. The mentioned minerals had been observed to contain high concentrations of $^{40}$K, $^{226}$Ra and $^{232}$Th, which are naturally occurring radionuclides in the environment plus the host of other heavy toxic metals (Chowdhury *et al*., 1999; Ismail *et al*., 2000, 2001; Ibeanu, 2003; Mohsen *et al*., 2005). In principle, there is a combination of both radiological and chemical toxic elements associated with tin mining processing activities (Ibeanu, 2003).

Majority of rivers and streams passing through tin mining and tailing processing environment had been observed to carry in their loads naturally occurring radionuclide's (Yusof *et al*., 2001). Most tin processing plants employed these rivers and streams for processing activities. Water used for processing (that is physical separation of valuable minerals from tin ores/tailings) was usually pumped from these water sources into either an already existing pond or artificially constructed pond and maintained as recycling close water management system. The turnout of events usually resulted in the ponds becoming the storage system to collect effluent sediments from the plants and accumulation of deposited sediments along the banks of the water bodies.

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**The question that seeks for an answer is what is the fate of these sediments especially when deposited at the banks of the water bodies?** It should be noted that these sediments were easily accessible to the residents who utilized them for different purposes, (such as making earthen huts, bricks for building residential homes, foundry sand for steel casting, road fills for road maintenance, sand for roasting groundnuts et cetera). It is a known fact that $^{226}$Ra, one of the radio nuclides detected in the sediments, is usually the reference radionuclide in assessing radiological risks (UNSCEAR, 1982).

Considering the posed potential health risk to residents and other members of the public in general, this study addresses the issue of utilization of the tin mine pond sediments for making bricks used in building residential homes through evaluation of its Naturally Occurring Radioactive Materials (NORMs) contents. The acquired data would be utilized in the assessment of the level of potential radiological risks involvement. To achieve the set goal, identification of critical groups/locations within a targeted environment was made and four rivers identified. Gamma spectrometry technique with Na(Tl) detector was employed in the analysis.

## MATERIALS AND METHODS

**In-situ measurements and sample collection:** A general survey of tin mining/mined environment was conducted in an attempt to identify critical locations/groups with threatened radioactively contaminated sediment for the study. In this regard, Yelwa, Kokop, Zawan and Sabon-Barki were selected for the study via in-situ gamma dose rate measurements taken randomly at different points. A total of 40 sediment samples (10 per location) were collected for laboratory NORMs measurements. In addition, the samples were characterized for toxic element composition and distribution.

Sediment samples were collected along the river banks. They were initially dried under ambient temperature in the laboratory and were later subjected to mild oven-drying to an approximately constant mass at a temperature of 90°C for 12 h before grinding. They were ground into fine powder to pass through a 2-mm sieve and packed to fill cylindrical plastic containers with inner dimension of 7.2 cm diameter and 6 cm height, which suited the optimal soil mass of 300-350 g for spectrometric analysis of bulk sediment samples established by Ibeanu (1999). The samples were sealed and stored for a minimum of 28 days to allow for radium equilibration with daughters. The IAEA gamma spectrometric reference materials (RGK-1, RGU-1 and RGTH-1) used in the analysis were equally prepared in a similar manner, sealed and stored as described above. The set analytical configuration/geometry was maintained throughout for both standard and samples.

Gamma ray spectrometry was employed for the activity concentration measurements. The used detector assembly consisted of a 7.62×7.62 cm NaI (Tl) detector housed in a 6 cm thick lead-shield, cadmium-lined assembly with copper sheets for the reduction of background radiation. The entire assembly was coupled to a computer-based Multichannel Analyzer (MCA) card system MAESTRO programmed used for the data acquisition and spectra analysis. The analyzer was calibrated with the IAEA supplied reference materials for the quantitative determination of $^{40}$K, $^{226}$Ra and $^{232}$Th in the sediment samples. The system was set at a working energy range of 0-3000 keV, which accommodated the energy range of interest in the study. An energy resolution of 7.2% (661.6 keV $^{137}$Cs) was obtained. Each sample was counted for 30,000seconds in the set geometry. The activity concentration of $^{226}$Ra and $^{232}$Th were determined by the γ-lines of their decay products: $^{214}$Bi (1760KeV) and $^{208}$Tl (2614KeV), respectively. The activity concentration of $^{40}$K was determined from its 1460 KeV γ-line.

**Calculation of radiological effects:** Three radiological models were employed in the assessment of the radiological effects: annual effective dose ($H_E$), radium equivalent dose ($Ra_{eq}$) and the external hazard index ($H_{ex}$) due to specified radio nuclides $^{226}$Ra, $^{232}$Th and $^{40}$K.

The absorbed gamma dose rates (nGy/h) in air at 1 m above the ground surface for the uniform distribution of radio nuclides ($^{40}$K, $^{226}$Ra and $^{232}$Th) were computed on the basis of guidelines provided by UNSCEAR (2000) report, as given in Eq. (1):

$$D = 0.042C_K + 0.462C_{Ra} + 0.604C_{Th} \quad (1)$$

To estimate the $H_E$ (mSv/y), the conversion coefficient from absorbed dose rate in air to effective dose (0.7 Sv/Gy) and outdoor occupancy factor (0.2) proposed by UNSCEAR (2000) report, were incorporated in Eq. (1) to obtain equation (2) given as:

$$H_E = D\ (nGy/h) \times 8760\ h \times 0.2 \times 0.7\ Sv/Gy \times 10^{-6} \quad (2)$$

The activity concentration distribution of $^{40}$K, $^{226}$Ra and $^{232}$Th in the sediment samples was not uniform. Uniformity with respect to exposure to gamma radiation has been defined in terms radium-equivalent activity concentration of building materials containing different concentrations of $^{40}$K, $^{226}$Ra and $^{232}$Th (Ahmed, 2005). The radium-equivalent activity concentration levels ($Ra_{eq}$) from the analyzed sediment samples were estimated using Eq. (3):

$$Ra_{eq} = 0.077C_K + C_{Ra} + 1.43_{Th} \quad (3)$$

The maximum value of $Ra_{eq}$ in the building materials must be less than 370 Bq/kg for safe

utilization (Flores *et al*., 2005). Also its suitability can be estimated as presented in Eq. (4):

$$H_{ex} = \left[\frac{C_K}{4180} + \frac{C_{Ra}}{370} + \frac{C_{Th}}{259}\right] \leq 1 \quad (4)$$

The activity index for material used in residential building is estimated using Eq. (5) proposed by Guides (2003):

$$I_\gamma = \left[\frac{C_K}{3000} + \frac{C_{Ra}}{300} + \frac{C_{Th}}{200}\right] \quad (5)$$

If the activity index ($I_\gamma$) is ≤1, the material can be used as building material, so far as the radioactivity is concerned, without restrictions. All the $C_K$, $C_{Ra}$ and $C_{Th}$ are the activity concentration levels of $^{40}K$, $^{226}Ra$ and $^{232}Th$ in Bq/kg, respectively.

## RESULTS AND DISCUSSION

Radiation exposure due to materials incorporated in the construction of residential houses is usually categorized into two parts, namely; external and internal exposures. However, the current study investigated only the degree of potential external exposures for immersion and the utilization of such buildings. The study is focused on partial/total immersion of whole body by gamma radiation. The validity of the utilization of these sediments in construction of residential houses was estimated based on the three enumerated radiological models.

Table 1 is a presentation of the mean measured activity concentration levels for the forty sediment samples ten per location and the mean dose rate (computed and in situ) of the sampling points. According to UNSCEAR (1982), the world mean activity concentration of natural radio nuclides in soil collected from an undisturbed environment for $^{40}K$, $^{226}Ra$ and $^{232}Th$ are 370.0, 25.0 and 25.0 Bq/kg, respectively. Table 2 presents computed radiological indices on which the validity of these samples for construction of residential houses is based.

According to Flores *et al*. (2005), the maximum value of $Ra_{eq}$ of any material for residential building must be less than 370 Bq/kg and this equals an annual effective dose of 1.5 mSv/yr taking potential radiological consequences of NORMs into cognizance. However, this does not provide for adequate protection of the critical population. For an adequate protection, there must be provision for constraints, which gives room for maximum radiological protection. In this regard, European Community report (EC, 1999) set this value as 0.3-1.0 mSv/yr. For proper radiological safety and adequate protection of the members of the public, this study is based on the European community report. Critical protection, none of the samples satisfied eligibility utilization criterion set by the European community. Worse still, Kokop sediment samples exhibited very high $Ra_{eq}$, which is an indication of potential radiological problem of serious concern for the particular location. It is a factor of approximately 5.4 the recommended value of ≤370 Bq/kg. This is an indication that samples collected from such locations may not be radio logically safe for utilization in residential house building constructions. There is then a need for such areas to be placed under restriction access control measures. The location should be placed under strict confinement to avoid human contact with the radioactively contaminated sediments. Its non-restriction utilization, as is the case presently is a case of serious concern and must be handled in like manner to avert probable radiological risks.

Table 2 presents calculated radiological hazard indices of the sediment samples. The acceptable annual effective dose for members of the public without constraint should be 1.0 mSv/yr for safety purposes (NBIRR, 2003; ICRP Publication 60, 1990). However under radiological constraints for an adequate protection of potential users of 0.5 mSv/y as recommended by EC (1999) report still only Kokop samples pose radiological danger as exhibited in Table 2, column 2. Kokop samples failed to meet the criteria for utilization hence unacceptable for building construction of residential homes.

Table 2, column 3, is a presentation of the calculated radium equivalents for respective locations studied. All other locations with the exception of Kokop, presented $Ra_{eq}$ values of less than 370 Bq/kg as recommended. This model once again gives an indication of non validity of Kokop samples in the building construction of residential homes.

In considering the external gamma activity concentration level indices, for materials used in bulk amounts, such as concrete, brick, etc, $H_{ex}$ and $I_\gamma$ should

Table 1: Activity concentration (Bq/kg)and mean dose rate (nGy/h)

| | | Mean Activity Concentration (Bq/kg) | | | D (nGy/h) | |
|---|---|---|---|---|---|---|
| Code | NSA | $^{40}K$ | $^{226}Ra$ | $^{232}Th$ | Computed | In-situ |
| Ylw | 10 | 248.7±17.8 | 87.8±9.80 | 176.9±10.70 | 182.9±9.5 | 201.7±11.0 |
| Zwn | 10 | 195.2±11.2 | 115.7±17.9 | 97.2±12.400 | 148.6±12.8 | 136.2±10.9 |
| Kkp | 10 | 337.8±12.8 | 407.0±9.80 | 1110.8±13.7 | 873.2±16.9 | 869.1±16.9 |
| Sbk | 10 | 238.9±21.9 | 91.1±17.90 | 155.9±19.70 | 156.3±13.8 | 152.7±8.90 |

NSA: Number of Samples Analyzed; Ylw: Yelwa; Zwn: Zawan; Kpk: Kokop; Sbk: Sabon barki

Table 2: Computed radiological hazard indices of sediment samples

| Sample code | $H_E$ (mSv/y) | $Ra_{eq}$ (Bq/kg) | $H_{ex}$ | $I_\gamma$ |
|---|---|---|---|---|
| Yelwa | 0.23±0.01 | 360.9±13.10 | 0.98±0.03 | 1.26±0.01 |
| Zawan | 0.17±0.03 | 270.0±17.30 | 0.73±0.02 | 0.94±0.02 |
| Kokop | 1.07±0.01 | 2022.5±30.1 | 5.47±0.05 | 7.02±0.02 |
| Sabon Barki | 0.19±0.02 | 332.0±22.70 | 0.92±0.01 | 1.16±0.03 |

be less than 1. In this light, therefore, Kokop sediments, as presented in Table 2, columns 4 and 5 violates the recommendation and should not be used for the building construction of residential homes.

All the studied samples exhibited fairly high concentration of arsenic bearing in mind that it is a highly toxic element. The range and mean recorded arsenic concentration levels in the samples are 116-244 (131±17), 121-203 (154±30), 132-292 (148±28) and 117-248 (126±14) ppm for Yelwa, Zawan, Kokop and Sabon Barki samples, respectively. The values in parenthesis are the means. According to Agency for Toxic Substances and Disease Registry (ATSDR, 2000a) arsenic concentration levels in soil of ≤ 20 ppm do not present a threat to humans. Samples with concentration levels higher than 20 ppm pose threats to both children and adults. Worse still when considering children with hand-to-mouth habit, it turns out an issue of great concern since African children do have access to soil, sediments and dust and play with them much often as permitted by tradition.

## CONCLUSION AND RECOMMENDATIONS

Gamma hazards and risk associated with prominent naturally occurring radio nuclides present in sediment samples collected from Yelwa, Zawan, Kokop and Sabon Barki tin ore processing streams were studied. The obtained activity concentration data showed activity enhancement of $^{226}Ra$ and $^{232}Th$ in all the samples which is more significant in Kokop samples. The radiological risk is greatly pronounced with the Kokop samples (Table 2). All the radiological models exhibited high potential radiation risk with utilization of Kokop samples. For update, further studies are recommended with reclamation of the radioactively contaminated sediments to avoid probable ingestion of the radionuclides via food chain pathways.

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