

# ASSESSMENT OF RADIOLOGICAL HAZARDS OF TIN MINING AND ORE PROCESSING IN JOS, NIGERIA



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## Abstract

On the Jos Plateau were found uncontrolled tailing heaps generated from Tin Mining Activities. To assess the associated radiological hazards, an abandoned tailing dump ground was investigated with the residents as the critical population. The mean activity concentrations of  $^{40}\text{K}$ ,  $^{226}\text{Ra}$  and  $^{232}\text{Th}$  in the 60 analyzed soil samples were  $1251.7 \pm 478.4$ ,  $3867.5 \pm 1282.7$  and  $8301.9 \pm 2862.6 \text{ Bqkg}^{-1}$ , respectively with a mean computed dose rate of  $7.2 \pm 1.6 \mu\text{Gyh}^{-1}$ . An annual mean outdoor effective dose of  $8.9 \pm 0.9 \text{ mSvy}^{-1}$  was estimated. Also the activity concentrations of  $^{40}\text{K}$ ,  $^{226}\text{Ra}$  and  $^{232}\text{Th}$  in the 60 control soil samples were  $447.0 \pm 68.0$ ,  $37.4 \pm 7.4$  and  $115.4 \pm 16.7 \text{ Bqkg}^{-1}$ , respectively with a mean dose rate of  $0.11 \pm 0.01 \mu\text{Gyh}^{-1}$ .

To account for the internal exposure, vegetables and root crops commonly grown and consumed in the area were assayed. Six varieties of vegetables and five varieties of root crops were analyzed. An internal annual mean effective dose of  $148.98 \pm 8.14 \mu\text{Svy}^{-1}$  was estimated.

The verification of dose limit compliance for members of the public demands that:

$$\frac{\text{External Dose}}{\text{Dose Limit}} + \frac{\text{Intake (ingested)}}{\text{ALI (ingestion)}} + \frac{\text{Intake (inhaled)}}{\text{ALI (inhalation)}} \leq 1$$

Based on obtained data above, there is non-compliance with the dose limit, since the first term of the compliance formula is much greater than unity. There is therefore a need for an intervention to prevent radiation over exposure of the members of the public.

The calculated cancer mortality risk for external and internal exposure scenarios for  $^{226}\text{Ra}$  and  $^{232}\text{Th}$  were  $(1.67 \pm 0.33) \times 10^{-6}$  (0.00017%) and  $(3.41 \pm 0.14) \times 10^{-6}$  (0.00034%), respectively. The  $^{226}\text{Ra}$  radionuclide contributed about 96.09% of the risk in the external scenario with only 3.09% from the  $^{232}\text{Th}$  while in the internal, the  $^{226}\text{Ra}$  contributed only 70.38%. The combined external and internal (ingestion) risk is  $(5.08 \pm 0.36) \times 10^{-6}$ /year.

## Introduction

Tin mining and ore processing have a long history in the Plateau State with Jos as capital. Babalola (1984) showed that tin ores and the associated wastes contain monazites, zircons. He measured thorium and uranium activity concentration levels of  $753960 \text{ Bqkg}^{-1}$  and  $18300 \text{ Bqkg}^{-1}$ , and  $585040 \text{ Bqkg}^{-1}$  and  $10126 \text{ Bqkg}^{-1}$  from monazite and zircon components respectively of the

tailings. It was observed that these tailings were sporadically spread over large areas around the tin worked environments and worse still, the villagers reworked these tailings, used them for building materials and farmed the lands. Ibrahim and Whicker (1988) reported on the uptake of radionuclides by plants/crops grown on radioactively contaminated soil and deposition on forage. A major concern is that the exposed natural radionuclides may gain access into man via two main critical pathways, which include external irradiation by gamma rays and internal irradiation by inhalation of radon gas from ore dust, and by ingestion of radioactively contaminated food.

In the light of the subject matter, this study was undertaken to assess the radiological danger of tin mining activities in Jos, Nigeria. It was aimed at locating an active tailing dump ground, characterizing the radioactivity distribution of the soil and commonly cultivated/consumed food items, and finally to quantify the potential radiological hazards to the members of the public.

## **Methods**

### *Preliminary Studies*

A feasibility study of three tin worked environments (Mista ali, Sabon-gida and Kuru karama) was undertaken, which assisted in the selection of a critical location for the proposed study. Kuru Karama (Jos) tailing dump ground was a choice for the study due to its relatively recorded higher activity concentrations for both soil and tailing samples.

### *Field Measurements and Sampling Procedures*

The plot was partitioned in grids of 10m by 10m spacing for detailed radiometric investigations. In-situ gamma radiation measurements were made with NaI(Tl) detector mounted at 1m above ground level. The device was regularly checked with  $^{137}\text{Cs}$  standard source. 10 readings were taken per spacing and a mean deduced. Time interval before successive readings was about 15seconds. Soil samples were collected on grid points at the rate of about 1kg per sample. A total of 60 Kuru and 60 control soil samples were collected. Each collected soil sample was emptied into a polyethylene-sampling bag, tied and labeled.

Commonly cultivated and consumed food items were sampled. These include the following vegetables: (10) lettuce (*lactuca sativa*), (12) cabbage (*brassica* var.), (10) karkashe (*radiatum sesanum*), (15) spinach (*amaranthus tricolor*), (10) green beans (*vigna unguiculenta*) and (10) garden egg (*solanum* sp) and root crops – (12) cocoyam (*colocasia esculenta*), (10) cassava (*manihot esculenta*), (10) sweet potatoes (*ipomoea batatas* var.), (05) onions (*allium cepa* var.) and (08) carrot (*daucas carota* var.).The samples were collected from the farmers from their respective farms. The initial sampling was made in March/April 1996 and repeated in Nov./Dec 1996 to account for the two seasonal variations in Nigeria. Numbers in bracket indicate number of samples analyzed.

### *Sample Preparations*

Soil samples were oven dried to approximately constant mass at a temperature of 110°C. The samples were ground to fine powder, packed to fill cylindrical plastic containers of dimension

7.2cm diameter by 6.0cm high, which accommodated about 350g mass of soil, a selected optimal sample container height that allowed for closed shield analysis. The chosen configuration and geometry was maintained throughout. The samples were triple sealed and stored for 20 days to enable the uranium and thorium get to equilibrium with their daughters. The IAEA gamma spectrometric reference materials (RGK-1, RGU-1 and RGTh-1) were packed in similar container, sealed and stored in identical manner.

### Sample Analysis

A 7.62cm by 7.62cm NaI(Tl) detector assembly enclosed in a 6cm lead shield with an inner lining of cadmium and copper sheets was employed in the analysis. The three differential spectrometry channels, which include 1460keV, 1764keV and 2614keV energy peaks were used for the determination of  $^{40}\text{K}$ ,  $^{226}\text{Ra}$  and  $^{232}\text{Th}$ , respectively. The detector assembly was coupled to a computer based multichannel analyzer (MCA) card ACCUSPEC and the MCA initially calibrated with standard isotopic sources ( $^{137}\text{Cs}$ ,  $^{60}\text{Co}$  and  $^{54}\text{Mn}$ ). The amplifier gain was set at 4.55 and the energy range of interest of the MCA set at 3000keV. An energy resolution of 7.2% of 661.6keV  $^{137}\text{Cs}$  was measured. The photopeaks of  $^{137}\text{Cs}$ ,  $^{54}\text{Mn}$  and  $^{60}\text{Co}$  were identified after 30 minutes counting time and the regions of interest marked from 570–735keV, 765–900keV, 1100–1245keV/1255–1420keV, respectively. The detector was re-calibrated using the RGK-1, RGU-1 and RGTh-1 reference materials for the quantitative determination of  $^{40}\text{K}$ ,  $^{226}\text{Ra}$  and  $^{232}\text{Th}$  in the samples. The energy windows of interest were set from 1380–1550keV for  $^{40}\text{K}$ , 1620–1820keV of  $^{214}\text{Bi}$  for  $^{226}\text{Ra}$  and 2460–2820keV of  $^{208}\text{Tl}$  for  $^{232}\text{Th}$ .

The soil samples were mounted on the surface of the detector and each counted for 6hrs in reproducible sample-detector geometry. Similarly, each food sample was counted for a minimum counting time of 10hrs. An ACCUSPEC software was used for data acquisition, processing and interpretation of gamma spectra. It provides its result in counts/sec (cps). The reference materials were equally counted and the calibration factors evaluated as shown in Table 1.

Table 1 - Energy Calibration for Quantitative Spectral Analysis

Isotope	Calibration Factor		Conversion Factor Bq·kg <sup>-1</sup> /ppm	Detection Limits•	
	x10 <sup>-3</sup> cps/ppm	x10 <sup>-4</sup> cps/Bq·kg <sup>-1</sup>		Ppm	Bq/kg
$^{40}\text{K}$	0.026	6.431	0.032	454.54	14.54
$^{226}\text{Ra}$	10.500	8.632	12.200	0.32	3.84
$^{232}\text{Th}$	3.612	8.768	4.120	2.27	9.08

- At 95% Confidence Interval (Calculated from experimental results)

### Results

The computed dose rates in air at 1m above ground level in Jos ranged from 0.6-45.2 $\mu\text{Gyh}^{-1}$  with an arithmetic mean of 7.2 $\pm$ 1.6 $\mu\text{Gyh}^{-1}$  while 0.11 $\pm$ 0.01 $\mu\text{Gyh}^{-1}$  was recorded at the control area. The measured mean activity concentration levels in Jos soil and assayed food items with the

corresponding annual mean effective doses are as shown in Table 2. The activity concentration levels in the control soils are  $447.0 \pm 68.0$  for  $^{40}\text{K}$ ,  $37.4 \pm 7.4$  for  $^{226}\text{Ra}$  and  $115.4 \pm 16.7$  Bqkg<sup>-1</sup> for  $^{232}\text{Th}$ . The  $^{232}\text{Th}/^{226}\text{Ra}$  ratios for Penang (Malaysia, Chong et al, 1985) and Jos (Nigeria) soil data are 1.74 and 2.14, respectively. This shows that the two results are comparable as tin mining environments and supports thorium enhancement. The control soil data are comparable with the world mean values for a normal environment whereas the Jos soil data are factors of about 154 and 332 the world mean values for  $^{226}\text{Ra}$  and  $^{232}\text{Th}$ , respectively. This is an indication that the area is a high background area using Burton (1998) criterion.

Table 2 - Mean Activity Concentration and Annual Mean Effective Dose

Sample Type	Number of Samples Analyzed	Mean Activity Concentration (Bqkg <sup>-1</sup> )			Mean Dose/yr
		$^{40}\text{K}$	$^{226}\text{Ra}$	$^{232}\text{Th}$ ** ( $^{228}\text{Th}$ )*	* $\mu\text{Sv}$ ** $\text{mSv}$
Soil**	60	1251 $\pm$ 478 (420-7824)	3867 $\pm$ 1282 (120-14010)	8301 $\pm$ 2862 (270-49892)	8.90 $\pm$ 0.90
Vegetable*	67 (6 varieties)	39.38 $\pm$ 3.47 (6.72-84.02)	2.17 $\pm$ 0.24 (0.01-7.77)	2.02 $\pm$ 0.37 (0.02-7.65)	75.67 $\pm$ 6.18
Root crop*	45 (5 varieties)	15.87 $\pm$ 1.12 (2.05-61.57)	1.65 $\pm$ 0.13 (0.02-7.65)	3.27 $\pm$ 0.18 (0.02-9.02)	73.31 $\pm$ 5.30
Veg. & R/crop					148.98 $\pm$ 8.14

\* fresh weight, \*\* dry weight

The mean ratio of  $^{232}\text{Th}/^{226}\text{Ra}$  in the analyzed soil samples is approximately 2.14 as against 1.92 observed in the food items. On the average, thorium absorption was preferred to radium. This is in agreement with Holtzman (1980) second statement that special situations in which thorium is concentrated in an environment can substantially increase the thorium dietary levels hence the  $^{228}\text{Th}/^{226}\text{Ra}$  would be greater than unity. It was observed that ipomoea batatas absorbed the highest  $^{226}\text{Ra}$  and  $^{228}\text{Th}$  concentration levels of  $4.32 \pm 0.17$  and  $6.35 \pm 0.05$  Bqkg<sup>-1</sup>, respectively. The preferential absorption of  $^{228}\text{Th}$  to  $^{226}\text{Ra}$  by ipomoea batatas is in good agreement with the work of Lalit and Ramachandran (1980) in Kerala (India). By the present study,  $^{228}\text{Th}/^{226}\text{Ra}$  is 1.47 as against 1.23 by the Lalit and Ramachandran. Similarly, the ratio of  $^{228}\text{Th}/^{226}\text{Ra}$  in amaranthus tricolor reported by Lalit and Ramachandran was 0.83 as against 0.71 obtained by the present study. All the sampled food items absorbed  $^{40}\text{K}$  relatively higher than the other two radionuclides. This is due to the fact that potassium is an essential element and plants are incapable of isotopic differentiation. It was observed that the activity concentration ratios of unwashed to washed for  $^{40}\text{K}$ ,  $^{226}\text{Ra}$  and  $^{228}\text{Th}$  in the vegetables were relatively of the order of 1.02–1.29, which is an indication that washing reduces the surface contamination levels. The mean intake levels of  $^{40}\text{K}$ ,  $^{226}\text{Ra}$  and  $^{228}\text{Th}$  in the vegetables and root crops are  $7.42 \pm 0.71$  (4.69-11.60),  $0.46 \pm 0.04$  (0.23-0.69) and  $0.43 \pm 0.04$  (0.29-0.91) and,  $3.41 \pm 0.26$  (2.26-4.15),  $0.46 \pm 0.04$  (0.24-0.93) and  $0.71 \pm 0.03$  (0.45-1.36) Bq, respectively. The values in parenthesis are ranges.

## Conclusion

The verification of dose limit compliance for members of the public demands that:

$$\frac{\text{External Dose}}{\text{Dose Limit}} + \frac{\text{Intake (ingested)}}{\text{ALI (ingestion)}} + \frac{\text{Intake (inhaled)}}{\text{ALI (inhalation)}} \leq 1 \quad (\text{Cember, 1988})$$

Based on obtained data above, there is non-compliance with the dose limit, since the first term of the compliance formula is much greater than unity. The calculated cancer mortality risk for external and internal exposure scenarios for  $^{226}\text{Ra}$  and  $^{232}\text{Th}$  were  $(1.67 \pm 0.33) \times 10^{-6}$  (0.00017%) and  $(3.41 \pm 0.14) \times 10^{-6}$  (0.00034%), respectively. The  $^{226}\text{Ra}$  radionuclide contributed about 96.09% of the risk in the external scenario with only 3.09% from the  $^{232}\text{Th}$  while in the internal, the  $^{226}\text{Ra}$  contributed only 70.38%. The combined external and internal (ingestion) risk is  $(5.08 \pm 0.36) \times 10^{-6}$ /year. It should be noted that the inhalation aspect has not been addressed and it is also an important radiation exposure pathway due to the presence of radioactive ore dust/radon gas in the atmosphere. Metal mining activities can be friendly to the inhabitants of the source area when the programme receives properly planned and carefully execution stages otherwise it is hazardous. To avoid radiation over exposure of the inhabitants via direct irradiation an intervention measure must be taken to mitigate the consequences of the potential radiation exposure due to the tin mining activities in the Jos environment.

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## Appendix

The accuracy of the work was checked via the analysis of the IAEA supplied gamma spectrometric reference materials.

	Certified Values (ppm)	This Work (ppm)
<sup>40</sup> K	448000.00 ± 3000.00	448007.77 ± 2459.06
<sup>226</sup> Ra	400.00 ± 2.00	402.86 ± 3.26
<sup>232</sup> Th	800.00 ± 16.00	800.11 ± 7.84

### Precision

<sup>40</sup> K	0.34%
<sup>226</sup> Ra	1.42%
<sup>232</sup> Th	1.31%

[Soil mass ≅ 350g]

[1 Hr counting time]