

# INVENTORY AND GEOCHEMICAL HOST PHASES OF NATURAL RADIONUCLIDES IN TIN MINING MATERIALS FROM NIGERIA

K Leopold<sup>1\*</sup>, A M Arogunjo<sup>2</sup>, A Giussani<sup>1</sup>, V Höllriegl<sup>1</sup>, U Oeh<sup>1</sup>, I Veronese<sup>3</sup>, U C Gerstmann<sup>1</sup>

<sup>1</sup>*Helmholtz-Zentrum München, German Research Center for Environmental Health, Institute of Radiation Protection, Ingolstädter Landstr. 1, 85764, Neuherberg, Germany*

<sup>2</sup>*Federal University of Technology, Department of Physics, P.M.B. 704, Akure, Nigeria*

<sup>3</sup>*Università degli Studi di Milano, Dipartimento di Fisica, via Celoria 16, 20133 Milano, Italy*

## Abstract

On the Nigerian Jos Plateau tin mining is extensively carried out in open pit style. All types of materials occurring there (raw materials, waste, and soil) were analysed radiometrically. For comparison, unaffected soils were also been investigated. The geochemical host phases of the natural radionuclides stored were determined by a sequential extraction procedure according to the European BCR-standard. It was found that especially easily mobilisable <sup>228</sup>Ra must be taken into consideration as a radioactive contaminant for the mining area.

Keywords: Tin mining, Nigeria, natural radionuclides, BCR-extraction procedure, gamma-spectrometry, TENORM

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\* corresponding author: Karsten Leopold, Helmholtz-Zentrum München, Institute of Radiation Protection, Ingolstädter Landstrasse 1, 85764 Neuherberg, Germany, Tel: 0049 89/3187-2205, Fax: 0049 89/3187-3323, karsten.leopold@helmholtz-muenchen.de

## 1. Introduction

Tin mining activities harmfully influence the environment due to deposition of large volumes of tailings (Remon et al., 2005), which significantly affect the natural composition of an ecosystem. The materials occurring in frame of tin mining activities are often to be considered as TENORM (“Technologically Enhanced Naturally Occurring Radioactive Materials”), whose radiation source is generally given by natural radionuclides of both the uranium and thorium decay series. Enhanced radiation exposures not only of workers but also of inhabitants of the mining and processing sites can be caused by mining, milling and processing of uranium and thorium bearing raw materials (UNSCEAR, 2000). This study is focused on the estimation of the environmental hazard dimensions due to the availability and mobilization potential of long-living natural radionuclides ( $^{238}\text{U}$ ,  $^{226}\text{Ra}$ ,  $^{210}\text{Pb}$ ,  $^{228}\text{Ra}$  and  $^{228}\text{Th}$ ) from tin, columbite, zircon, tin tailings, and soils. For this purpose, the 3-step extraction procedure proposed by the European Bureau Communautaire de Référence (BCR) was applied. Thereby, the geochemical host phases of these radionuclides are evaluable. This approach was successfully applied for radium isotopes in materials from Polish hard coal mining (Leopold et al., 2007).

## 2. Investigation methods

40 sub-samples have been taken between August and September 2006 in 3 regions (Bisichi, Jos, and Akure, sketch in figure 1), which were then mixed according to their material classification resulting in 8 investigated samples, so 5 sub-samples of a material gave 1 measured sample. The materials comprised the raw materials tin, zircon, columbite (Dress) and columbite (Burnt), the tailing waste and soils from the mining area Bisichi and from Jos and Akure city as unaffected samples. "Columbite (Dress)" indicates the original form of the ore, while "Columbite (Burnt)" is the result obtained after heating the processed ore in a

furnace for improving its mineral quality. In figure 1, the tailing sampling points at the settling ponds are shown. Furthermore, the fosses from open pit mining, where the raw material samples have been collected, can be clearly identified. The soils are taken in 10 cm depth at untreated and unfortified places within the cities assuming a homogenous radionuclide distribution. Despite the relatively high sampling uncertainty due to their short-range spatial variance (Perk, 2008), it is the only way to check the hazard dimensions in those anthropogenic areas.

The solid sample materials, which are all present in loose state, were dried for 24 h at 105 °C and then homogenized by sieving for the grain size fraction <2 mm. An aliquot was separated from that and measured by gamma-spectrometry for its initial radionuclide content.

## 2.1 BCR extraction procedure

The sequential extraction as proposed by the BCR was carried out according to Ure et al. (1993), whose handling scheme is presented in Table 1. In summary, three liquid fractions were obtained as representatives for the easily exchangeable (grain surface adsorbed, F1), reducible (iron and manganese oxides, F2) and oxidisable (sulphides and organic matter, F3) level. 10 g of solid were filled into acid cleaned polypropylene (PP) centrifugal cups. By the successive adding of several reagents, the radionuclides of different types of bonding were solved in those liquids. For the preparation of the chemical solutions needed, completely desalted water and chemicals of the purification degree “p.a.” were always used. The extraction shaking was done by an overhead-shaker at 50 rpm in accordance to the time limits mentioned in the procedure instruction. After each step, the suspension in the PP-cup was centrifuged at 4500 rpm for 5 min, in some cases the centrifuging time was extended to 20 min, if the suspension was not fixed on the bottom of the cup before. The separated extraction liquid was then decanted by a 45 µm membrane filter and transferred into a

Marinelli cup of 500 mL volume. In frame of the sequential BCR procedure, the reagent of the next step was filled in the same PP-cup, which contained the remaining solid material on the bottom from the precedent extraction step.

## 2.2 Gamma-spectrometry

Aliquots of the mixed mineral and soil samples were analysed by gamma-spectrometry at the Physics Department of Università degli Studi di Milano (Unimi), Italy and at the Radioanalytical Laboratory (RADLAB) of the Helmholtz-Zentrum München (HMGU), Germany. At both laboratories, negative-poled (Unimi: 35%, RADLAB: 25% rel. efficiency) high-purity germanium detectors (HPGe) of coaxial shape were used to perform the solid's measurements, whereas the BCR-liquids were exclusively analysed at the HMGU. The detection limit for  $^{226}\text{Ra}$  of the n-type detector at the RADLAB was calculated according to Currie (1968) and is given by 0.5 Bq/kg in case of solids and 0.05 Bq/L for the extraction liquids due to long measuring times of a week roughly, that of the other radionuclides are slightly higher. Canberra GENIE-VMS was the software used for analyses at HMGU and Ortec MAESTRO at Unimi. Each sample and standard was placed in cylindrical containers made of polypropylene/plexiglass (diameter 6 cm, height 3 cm) and sealed to avoid radon leakage. According to Leopold (2007), this approach has been validated by using a milled uranium ore which leads to radon concentrations in air of 33,390 Bq/m<sup>3</sup> (measured via LUCAS-cells) and is completely filled into a beaker and sealed. That beaker was then put into a bigger vessel equipped with two valves, so the initially aged and therefore radon-free air inside it could be easily and permanently transferred into a radon monitor (NITON Rad7). The vessel's air showed a maximum radon concentration of 31.6 Bq/m<sup>3</sup>, which means the radon loss from the beaker is lower than 0.1 % and can be accepted.

At HMGU, the solid samples were stored at least for 20 days after preparation to let establish secular equilibrium between  $^{222}\text{Rn}$  and its progenies, the BCR-extractions at least for 80 days to get equilibrated  $^{234}\text{Th}$  for the  $^{238}\text{U}$  determination before the gamma measurements were performed. The measurement time varied from two hours to six days depending on activity in the solid/extracts, detector efficiency, and background contribution. The system's detection efficiency for solids was determined by the calibration source "Sediment S2 1995" from the German BfG (Bundesamt für Gewässerkunde, Koblenz) according to Mundschenk (1996) and by QCY48 from Isotrak for liquids. Quality assurance was additionally performed by regular participations in national (e.g. BfS "Radionuclides in water") and international proficiency tests like IAEA-CU-2006-03 or NPL ERPTE 2007.  $^{238}\text{U}$  was determined by the gamma line at 1001 keV from  $^{234\text{m}}\text{Pa}$  in case of solids and at 63.3 keV emitted by  $^{234}\text{Th}$  for the extraction liquids,  $^{226}\text{Ra}$  via the emissions at 295 keV and 609 keV (corrected for coincidence summing) emitted by its progenies  $^{214}\text{Pb}$  and  $^{214}\text{Bi}$  respectively,  $^{228}\text{Ra}$  by the peaks at 911 keV and 968 keV emitted by  $^{228}\text{Ac}$  and  $^{228}\text{Th}$  by the line at 583 keV (corrected for coincidence summing) emitted by  $^{208}\text{Tl}$ . For the determination of  $^{210}\text{Pb}$  in solids at its low energy peak of 46.5 keV, at HMGU the sample was either measured in the container and corrected for self-absorption effects according to Cutshall et al. (1983) or, if not possible in case of the columbite, tin and zircon samples due to strong absorption caused by a very high Z, in a specific thin layer geometry of 2 mm height to 5 cm diameter material volume.

Prior to the gamma-spectrometric measurements, all the BCR-extracts were solidified in order to avoid radon accumulation in the Marinelli beaker's air-filled top and precipitation of particles on its bottom as well. After the liquid's volume was exactly determined, the solidification was done by adding agar-agar into the liquid heated up to 80 °C roughly, the Marinelli beaker was then put into boiling water for at least 10 min. Subsequently, it was allowed to cool down slowly. In order to determine radionuclide activity concentrations

dissolved in each extractant as a proportion of the starting solid material, it is necessary to calculate the volume activity concentration of the extraction liquids in relation to the activity concentration in the dry mass. That is possible because the amount of starting material is weighed carefully before the extraction is undertaken. Therefore, each extraction liquid's activity concentration represents a percentage of the starting material's dry mass activity concentration. The solid residuals from the extraction procedure were measured at HMGU for control in a volume of 40 mL by a well detector (p-type, 20.9% rel. efficiency) offering an almost  $4\pi$ -design.

### 2.3 Uncertainty budget

The uncertainties given for each result represent the count rate's statistical uncertainty by gamma-spectrometry, which quotes to  $1\sigma$ -reliability. An estimation of the average total combined uncertainty per peak in the extraction liquids takes into account the average statistical uncertainty of the count rate (10%), the average statistical uncertainty of the background (5%), the uncertainty of the calibration material (2%), the uncertainty of the liquid's volume determination (0.25%), and the uncertainty of the balance machine (0.05%). Thereby, the average total uncertainty is given by 11.4% per peak in the extraction liquid, whereas the maximum total uncertainty is 20.7%, because the maximum accepted statistical uncertainty of the count rate is 20%.

## 3 Results and discussion

### 3.1 Uranium and thorium activity concentration in mineral sands and soils

The activity concentrations of  $^{238}\text{U}$  and  $^{232}\text{Th}$  in mineral sands, waste and soil from Bisichi and Jos and Akure as background control are presented in Table 2. Strongly enhanced activity concentrations of both decay series  $^{238}\text{U}$  and  $^{232}\text{Th}$  as well as their progenies were

found in all the mineral ore samples and their waste products (tailings). The mined minerals tin, columbite and zircon are characterised by high radionuclide concentrations as a result of their lately crystallising in frame of the geological process called “magmatic differentiation”. During tin processing, the radionuclides are transferred into the waste, in case of the  $^{238}\text{U}$  decay series they are even enriched in the settled suspension (tailings).

It is eye-catching that there is a significant radioactive disequilibrium for the  $^{238}\text{U}$  decay series in the mineral sands, tailing waste, and Bisichi soil, whereas the  $^{232}\text{Th}$  decay series is well equilibrated, in case of columbite also in transitory equilibrium. This might be explained by the fact that the extraction process is performed in a reducing milieu, where uranium is insoluble but radium is removed (Kemski et al., 1996). But nevertheless, the  $^{232}\text{Th}$  series is still equilibrated despite the presence of one radium isotope ( $^{228}\text{Ra}$ ). The different mobility properties of the radium isotopes might be more reasonably explained on the basis of the alpha-recoil effect (Thomas, 1993). Indeed,  $^{226}\text{Ra}$  is formed from  $^{238}\text{U}$  through three alpha disintegrations, and the recoil effect may induce cracks in solid materials from which  $^{226}\text{Ra}$  isotopes may escape via liquids such as groundwater. In contrast,  $^{228}\text{Ra}$  within the  $^{232}\text{Th}$  series is preceded only by one alpha disintegration step with very long half-life ( $1.4 \cdot 10^{10}$  y). So, the lower probability of inducing cracks in the materials can give an explanation for less mobile  $^{228}\text{Ra}$ , its progenies and therefore equilibrated  $^{232}\text{Th}$  decay series.

The high activity concentrations in disequilibrium of the Bisichi soil sample indicate additionally the dimensions of radionuclide contamination of that area as a result of mining activities. In comparison, the activity concentrations of the Akure and Jos reference soils were generally much lower and equilibrated. However, it must be pointed out that the uranium concentration is enhanced not only in Bisichi (about 100 times higher) but also in Jos city (about two times higher) compared with Akure. This observation is supported by

elevated uranium activity concentrations found in well water of Bisichi (1.27 mBq/L) and Jos (3.28 mBq/L) city (Arogunjo et al., submitted).

### 3.2 Geochemical host phases of stored natural radionuclides

The geochemical host phases of the natural radionuclides stored in the materials occurring in frame of tin mining are shown in table 3. It is remarkable that despite the fact that columbite is an iron-manganese oxide according to  $(\text{Fe, Mn}) [\text{Ta, Nb}]_2\text{O}_6$  (Matthes, 1996), all investigated radionuclides are mainly found in the oxidisable fraction being representative for sulphides in both types, “Dress” (30 %) and “Burnt” (15 %) as well. Therefore, an oxidisable carrier must have impured the columbite ore. Heinrich (1962) mentioned an overgrowing or partly replacing by radioactive species or coating and veining by secondary minerals. It is also evident that  $^{228}\text{Ra}$  is significantly easily exchangeable from both columbite samples and zircon, too. This is surprising, because these are very stable minerals consisting of a strong crystal lattice, so  $^{228}\text{Ra}$  might be released from those impurities, too. The tin ore and the resulting waste tin tailing show almost all the radionuclides remaining in the residual, so the used extraction liquids are much too weak to interfere the crystal lattices and to remove compounds. But, nevertheless, both materials show the same behaviour in the radionuclides’ geochemical types of bonding. In case of the soils, there is a discrepancy between the strongly contaminated one from Bisichi and both the others, Jos and Akure, with almost no enhanced radionuclide concentrations: the Bisichi soil provides more than 20 % of the initial  $^{228}\text{Ra}$  content as easily exchangeable, which is not proven for both the others. In contrast, Jos and Akure soil are determined to have bonded almost all radionuclides mostly on organic matter and sulphides, respectively, which can be expected with regard to the general composition of soil.

#### **4 Conclusion**

The BCR-extraction procedure gives easily exchangeable amounts especially of  $^{228}\text{Ra}$  from the columbite samples and the zircon as well as the contaminated Bisichi soil. A pollution effect on the mining area's environment by  $^{228}\text{Ra}$  must be taken into consideration, but it is mainly provided by the raw materials in combination with smaller amounts from the contaminated soil and can, therefore, not be exclusively attributed to human mining activities.

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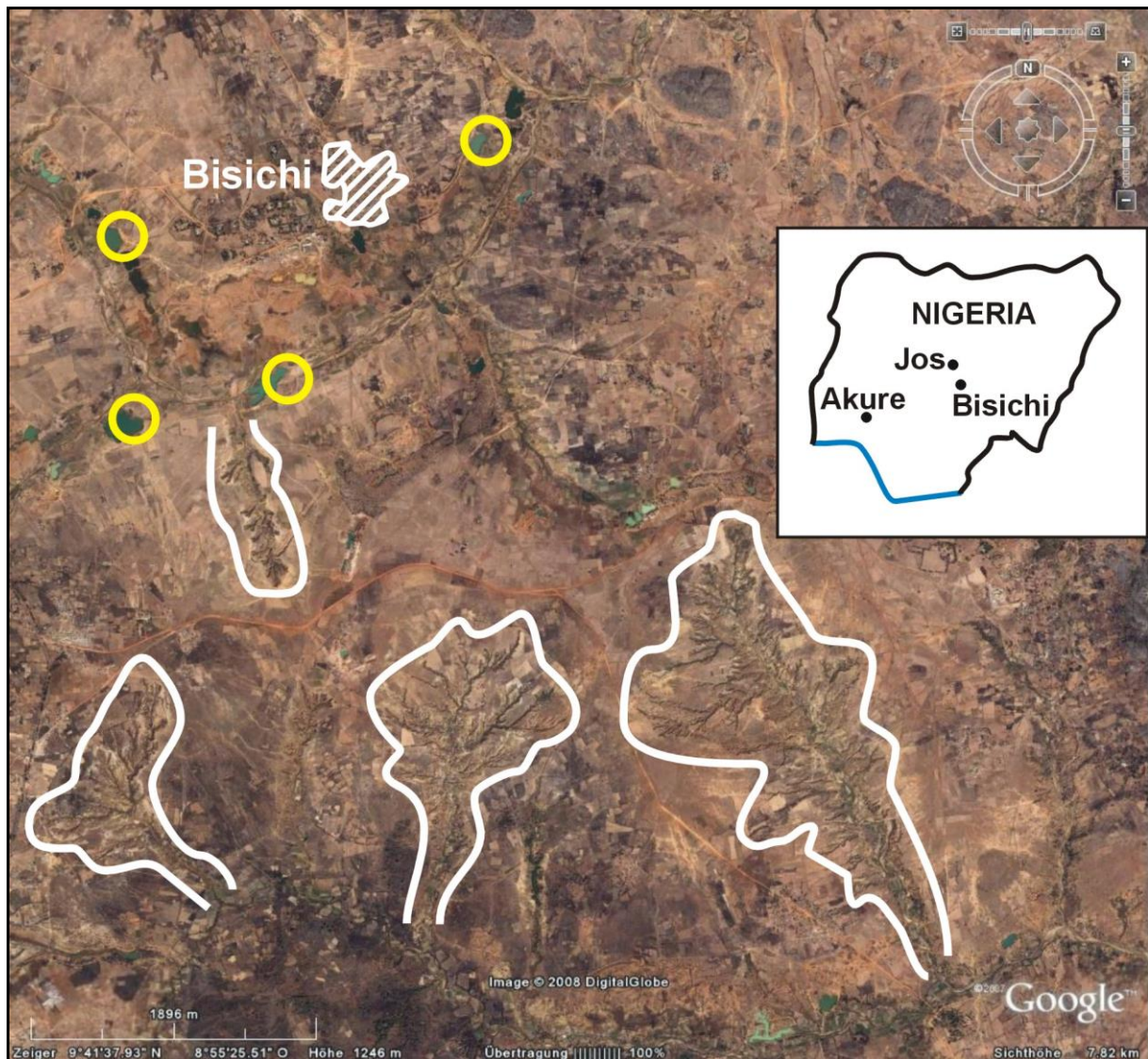


Figure 1: GOOGLE-Earth view of the Bisichi mining area; yellow circles mark tailing sampling points, southwards the fosses from open pit mining being whitely bordered are clearly identifiable.

Table 1. Scheme of the BCR extraction procedure

| fraction                         | procedure                                                                         |
|----------------------------------|-----------------------------------------------------------------------------------|
| starting material's volume: 10 g |                                                                                   |
| F1 (easily exchangeable)         | 400 mL of 0.11 mol/L $\text{CH}_3\text{COOH}$ , shaking time 16 h                 |
| F2 (reducible)                   | 400 mL of 0.1 mol/L $\text{H}_3\text{NOHCl}$ , pH 2, shaking time 16 h            |
| F3 (oxidisable)                  | a) $2 \times 100$ mL 35 % $\text{H}_2\text{O}_2$ , each heated up to near dryness |
|                                  | b) 500 mL of 1 mol/L $\text{CH}_3\text{COONH}_4$ , pH 2, 16 h shaking time        |

Table 2: Activity concentrations of  $^{238}\text{U}$ ,  $^{226}\text{Ra}$ ,  $^{210}\text{Pb}$ ,  $^{228}\text{Ra}$  and  $^{228}\text{Th}$  in mineral sands, waste and soil measured by gamma-spectrometry given in [kBq/kg]; each material consists of n=5 sub-samples.

| Material             | $^{238}\text{U}$  | $^{226}\text{Ra}$ | $^{210}\text{Pb}$ | $^{228}\text{Ra}$ | $^{228}\text{Th}$ |
|----------------------|-------------------|-------------------|-------------------|-------------------|-------------------|
| <u>Mineral sands</u> |                   |                   |                   |                   |                   |
| Columbite (Dress)    | $40 \pm 3$        | $14.2 \pm 1.0$    | $9.6 \pm 1.1$     | $92.0 \pm 1.6$    | $103.4 \pm 0.8$   |
| Columbite (Burnt)    | $30.0 \pm 1.9$    | $15.5 \pm 0.2$    | $9.35 \pm 0.14$   | $45.62 \pm 0.05$  | $57.9 \pm 0.5$    |
| Tin                  | $11.0 \pm 1.3$    | $7.4 \pm 0.6$     | $1.16 \pm 0.09$   | $32 \pm 4$        | $30.6 \pm 0.3$    |
| Zircon               | $51 \pm 3$        | $31 \pm 2$        | $10.31 \pm 0.15$  | $98 \pm 3$        | $94.6 \pm 0.8$    |
| <u>Waste</u>         |                   |                   |                   |                   |                   |
| Tin Tailing          | $22 \pm 2$        | $13.7 \pm 0.9$    | $7.3 \pm 0.6$     | $30.0 \pm 0.3$    | $29.4 \pm 0.3$    |
| <u>Soils</u>         |                   |                   |                   |                   |                   |
| Akure Soil           | $0.066 \pm 0.011$ | $0.041 \pm 0.010$ | $0.071 \pm 0.005$ | $0.053 \pm 0.001$ | $0.050 \pm 0.002$ |
| Jos city Soil        | $0.15 \pm 0.02$   | $0.109 \pm 0.005$ | $0.157 \pm 0.006$ | $0.059 \pm 0.004$ | $0.058 \pm 0.002$ |
| Bisichi Soil         | $8.7 \pm 0.5$     | $4.2 \pm 0.6$     | $4.60 \pm 0.11$   | $16.8 \pm 0.8$    | $18.10 \pm 0.12$  |

Table 3: Extracted activity concentrations of radionuclides per BCR-fraction and left in residual

| Radio-nuclide     | initial specific activity<br>[Bq/kg] | BCR-Extraction |             |        |            |             |        |            |             |      | residual activity concentration |        |
|-------------------|--------------------------------------|----------------|-------------|--------|------------|-------------|--------|------------|-------------|------|---------------------------------|--------|
|                   |                                      | 1. BCR         |             |        | 2. BCR     |             |        | 3. BCR     |             |      |                                 |        |
|                   |                                      | [Bq/L]         | [Bq/kg]     | [%]    | [Bq/L]     | [Bq/kg]     | [%]    | [Bq/L]     | [Bq/kg]     | [%]  | [Bq/kg]                         | [%]    |
| Columbite (Dress) |                                      |                |             |        |            |             |        |            |             |      |                                 |        |
| U-238             | 40,000 ± 3,000                       | < 1.2          | < 43        | < 0.1  | 8.9 ± 4.5  | 332 ± 168   | 0.8    | 66 ± 4.5   | 2,471 ± 168 | 6.2  | > 37,253                        | > 93   |
| Ra-226            | 14,200 ± 1,000                       | 26 ± 2.4       | 960 ± 89    | 6.3    | 17 ± 1.4   | 616 ± 52    | 4      | 27 ± 1.4   | 1,023 ± 52  | 6.7  | 12,610                          | 83     |
| Pb-210            | 9,600 ± 1,100                        | 8.8 ± 2        | 325 ± 74    | 3.4    | 56 ± 10    | 2,080 ± 374 | 21.7   | 60 ± 4.4   | 2,226 ± 165 | 23.3 | 4,939                           | 52     |
| Ra-228            | 92,000 ± 1,600                       | 713 ± 1.8      | 26,329 ± 66 | 28.3   | 281 ± 1.7  | 10,503 ± 64 | 11.3   | 438 ± 0.8  | 16,382 ± 30 | 17.6 | 39,866                          | 43     |
| Th-228            | 103,400 ± 800                        | 75 ± 0.9       | 2,760 ± 33  | 2.7    | 39 ± 0.9   | 1,455 ± 34  | 1.4    | 328 ± 0.7  | 12,267 ± 26 | 12   | 85,919                          | 84     |
| Columbite (Burnt) |                                      |                |             |        |            |             |        |            |             |      |                                 |        |
| U-238             | 30,000 ± 1,900                       | 2.3 ± 0.8      | 61 ± 21     | 0.2    | < 1        | < 27        | < 0.1  | 122 ± 3.2  | 3,173 ± 83  | 10.6 | > 26,699                        | > 89   |
| Ra-226            | 15,500 ± 200                         | 17 ± 0.2       | 447 ± 5.2   | 3      | 5.1 ± 0.1  | 139 ± 2.7   | 0.9    | 20 ± 0.4   | 524 ± 10    | 3.5  | 13,890                          | 93     |
| Pb-210            | 9,350 ± 140                          | 17 ± 2.1       | 432 ± 55    | 4.6    | 3.8 ± 1.4  | 104 ± 38    | 1.1    | 56 ± 1.7   | 1,457 ± 44  | 15.6 | 7,357                           | 79     |
| Ra-228            | 45,620 ± 50                          | 274 ± 0.8      | 7,137 ± 21  | 16.3   | 78 ± 0.4   | 2,119 ± 11  | 4.8    | 298 ± 0.9  | 7,769 ± 23  | 17.7 | 26,765                          | 61     |
| Th-228            | 57,900 ± 50                          | 31 ± 0.3       | 815 ± 7.8   | 1.7    | 6.7 ± 0.2  | 181 ± 5.4   | 0.4    | 199 ± 0.8  | 5,181 ± 21  | 10.  | 42,044                          | 87     |
| Tin               |                                      |                |             |        |            |             |        |            |             |      |                                 |        |
| U-238             | 11,000 ± 1,300                       | < 0.7          | < 28        | < 0.3  | < 0.2      | < 8.4       | < 0.1  | 3.4 ± 0.2  | 136 ± 8     | 1.2  | > 11                            | > 98   |
| Ra-226            | 7,400 ± 600                          | 0.1 ± 0.02     | 5.3 ± 0.8   | 0.1    | < 0.05     | < 2         | <0.03  | 0.6 ± 0.2  | 26 ± 8      | 0.4  | > 6,5                           | > 99.5 |
| Pb-210            | 1,160 ± 90                           | 0.3 ± 0.01     | 11 ± 0.4    | 0.9    | 0.5 ± 0.04 | 20 ± 1.6    | 1.7    | 1.5 ± 0.15 | 59 ± 6      | 5.1  | 1,070                           | 92     |
| Ra-228            | 32,000 ± 4,000                       | 36 ± 0.2       | 1,432 ± 8   | 5.3    | 14 ± 0.09  | 542 ± 3.6   | 2      | 21 ± 0.4   | 845 ± 16    | 3.1  | 24,192                          | 90     |
| Th-228            | 30,600 ± 3,00                        | 6.9 ± 0.2      | 275 ± 8     | 1.2    | 0.7 ± 0.05 | 30 ± 2      | 0.1    | 12 ± 0.4   | 488 ± 16    | 2.2  | 21,657                          | 97     |
| Zircon            |                                      |                |             |        |            |             |        |            |             |      |                                 |        |
| U-238             | 51,000 ± 3,000                       | < 1.1          | < 34        | < 0.07 | < 1.4      | < 43.1      | < 0.08 | 54 ± 3.6   | 1,615 ± 108 | 3.1  | > 49,697                        | > 97   |
| Ra-226            | 31,000 ± 2,000                       | 13 ± 1.9       | 393 ± 59    | 1.5    | 7.8 ± 0.4  | 242 ± 12    | 0.9    | 13 ± 0.5   | 378 ± 15    | 1.4  | 25,988                          | 96     |
| Pb-210            | 10,310 ± 150                         | < 0.95         | < 29        | < 0.3  | 4.8 ± 1.3  | 149 ± 40    | 1.4    | 25 ± 2.4   | 740 ± 72    | 7.2  | > 9,391                         | > 91   |
| Ra-228            | 98,000 ± 3,000                       | 632 ± 1.3      | 19,473 ± 40 | 20     | 412 ± 1    | 12,679 ± 31 | 13     | 377 ± 1.3  | 11,312 ± 39 | 11.8 | 52,236                          | 55     |
| Th-228            | 94,600 ± 800                         | 116 ± 0.9      | 3,562 ± 28  | 4.3    | 43 ± 0.6   | 1,318 ± 18  | 1.6    | 444 ± 1.3  | 13,336 ± 39 | 16.3 | 63,764                          | 78     |
| Tin tailing       |                                      |                |             |        |            |             |        |            |             |      |                                 |        |
| U-238             | 22,000 ± 2,000                       | < 0.3          | < 10        | < 0.04 | < 0.4      | < 12.8      | < 0.06 | 9.7 ± 0.15 | 286 ± 4.4   | 1.3  | > 21,621                        | > 98   |
| Ra-226            | 13,700 ± 900                         | 0.3 ± 0.06     | 9.7 ± 1.8   | 0.06   | 0.5 ± 0.05 | 15 ± 1.5    | 0.1    | 1.5 ± 0.1  | 44 ± 2.9    | 0.3  | 14,562                          | 99.5   |
| Pb-210            | 7,300 ± 600                          | < 0.4          | < 13        | <0.2   | 5.4 ± 0.7  | 161 ± 21    | 2.2    | 4.4 ± 1.5  | 129 ± 44    | 1.8  | > 6,997                         | 96     |
| Ra-228            | 30,000 ± 3,000                       | 81 ± 0.2       | 2,432 ± 6   | 8.2    | 49 ± 0.2   | 1,452 ± 4.8 | 4.9    | 58 ± 0.3   | 1,709 ± 8.8 | 5.8  | 23,958                          | 81     |
| Th-228            | 29,400 ± 300                         | 15 ± 0.13      | 462 ± 3.9   | 1.7    | 4.6 ± 0.09 | 138 ± 2.7   | 0.5    | 27 ± 0.25  | 800 ± 7.4   | 2.9  | 26,101                          | 95     |

Continuation of Table 3: Extracted activity concentrations of radionuclides per BCR-fraction and left in residual

| Radio-nuclide | initial specific activity<br>[Bq/kg] | BCR-Extraction |            |        |              |            |        |            |              |        | residual activity concentration |      |
|---------------|--------------------------------------|----------------|------------|--------|--------------|------------|--------|------------|--------------|--------|---------------------------------|------|
|               |                                      | 1. BCR         |            |        | 2. BCR       |            |        | 3. BCR     |              |        |                                 |      |
|               |                                      | [Bq/L]         | [Bq/kg]    | [%]    | [Bq/L]       | [Bq/kg]    | [%]    | [Bq/L]     | [Bq/kg]      | [%]    | [Bq/kg]                         | [%]  |
| Akure soil    |                                      |                |            |        |              |            |        |            |              |        |                                 |      |
| U-238         | 66 ± 11                              | < 0.43         | < 14.1     | < 21.4 | < 1.2        | < 41.6     | < 63   | < 0.7      | < 22.7       | < 34.3 | 66                              | 100  |
| Ra-226        | 41 ± 10                              | < 0.05         | < 1.6      | < 5.3  | < 0.1        | < 4.3      | < 14   | < 0.07     | < 2.5        | < 7.9  | 41                              | 100  |
| Pb-210        | 71 ± 5                               | < 0.2          | 6.6        | 9.3    | < 0.6        | < 21.8     | < 30.7 | 1.6 ± 0.6  | 52 ± 20      | 73     | > 19.2                          | > 27 |
| Ra-228        | 53 ± 1                               | 0.1 ± 0.02     | 3.3 ± 0.7  | 6.4    | 0.1 ± 0.02   | 4.5 ± 0.7  | 8.7    | 0.5 ± 0.08 | 15 ± 2.7     | 29     | 28.8                            | 56   |
| Th-228        | 50 ± 2                               | 0.5 ± 0.1      | 12 ± 3.3   | 24.7   | 0.08 ± 0.002 | 2 ± 0.07   | 3.9    | 1.4 ± 0.07 | 36 ± 2.4     | 71     | 0                               | 0    |
| Jos soil      |                                      |                |            |        |              |            |        |            |              |        |                                 |      |
| U-238         | 150 ± 20                             | 0.3 ± 0.01     | 10 ± 0.4   | 7      | 0.4 ± 0.02   | 16 ± 0.8   | 11     | 1.4 ± 0.4  | 56 ± 16      | 38     | 65                              | 44   |
| Ra-226        | 109 ± 5                              | < 0.1          | < 4.7      | < 4.1  | 0.2 ± 0.05   | 8.1 ± 2    | 7.1    | < 1.4      | < 56.5       | < 49.1 | > 45.7                          | > 40 |
| Pb-210        | 157 ± 6                              | < 0.9          | < 37.2     | < 23.7 | < 0.7        | < 26.9     | < 17.1 | 0.5 ± 0.05 | 21 ± 2       | 14     | > 71.6                          | > 46 |
| Ra-228        | 059 ± 4                              | 0.05 ± 0.001   | 1.9 ± 0.04 | 3.1    | 0.8 ± 0.08   | 31 ± 3.2   | 49     | 0.6 ± 0.1  | 23 ± 4       | 36     | 7.5                             | 12   |
| Th-228        | 58 ± 2                               | 0.2 ± 0.01     | 7.9 ± 0.4  | 14.4   | < 0.07       | < 2.8      | 5      | 0.4 ± 0.01 | 17 ± 0.4     | 30     | > 27.8                          | > 51 |
| Bisichi soil  |                                      |                |            |        |              |            |        |            |              |        |                                 |      |
| U-238         | 8,700 ± 500                          | < 1.5          | < 56.3     | < 0.6  | < 0.8        | < 29.3     | < 0.3  | 7.1 ± 1    | 265 ± 38     | 3.1    | > 8,349.2                       | > 96 |
| Ra-226        | 4,200 ± 600                          | 1.3 ± 0.1      | 49 ± 3.9   | 1.1    | 0.9 ± 0.2    | 35 ± 7.7   | 0.8    | 0.8 ± 0.2  | 31 ± 7.5     | 0.7    | 4,385                           | 97   |
| Pb-210        | 4,600 ± 110                          | < 2.2          | < 86.4     | < 1.9  | 1.2 ± 0.1    | 47 ± 3.9   | 1      | < 0.6      | < 22.6       | < 0.5  | > 4,444.3                       | > 97 |
| Ra-228        | 16,800 ± 800                         | 99 ± 0.5       | 3,819 ± 19 | 22.4   | 118 ± 0.3    | 4,536 ± 12 | 26.6   | 44 ± 0.3   | 1,665 ± 11.3 | 9.8    | 7,040                           | 41   |
| Th-228        | 18,100 ± 120                         | 10 ± 0.2       | 401 ± 7.7  | 2.4    | 9.1 ± 0.2    | 351 ± 7.7  | 2.1    | 89 ± 0.4   | 3,361 ± 15   | 20.5   | 12,307                          | 75   |