URANIUM AND THORIUM IN SOILS, MINERAL SANDS, WATER AND FOOD SAMPLES IN A TIN MINING AREA IN NIGERIA WITH ELEVATED ACTIVITY.

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Abstract

The activity concentrations of uranium and thorium have been determined in soils and mineral sands from the Nigerian tin mining area of Bisichi, located in the Jos Plateau, and from two control areas in Nigeria (Jos City and Akure) using high-purity germanium detectors (HPGe). High resolution sector field inductively coupled plasma mass spectroscopy (HR-SF-ICP-MS) was used to determine uranium and thorium in liquids and foodstuffs consumed locally in the mining area. The activities of uranium and thorium measured in the soils and mineral sands from Bisichi ranged from 8.7 kBq·kg^{-1} to 51 kBq·kg^{-1} for ^{238}U and from 16.8 kBq·kg^{-1} to 98 kBq·kg^{-1} for ^{232}Th, respectively. These values were significantly higher than those in the control areas of Jos City and Akure and the reference values reported in the literature, and even higher than the concentrations reported for areas of high natural radioactive background. Radionuclide concentrations in samples of the local foodstuffs and in water samples collected in Bisichi were found to be higher than UNSCEAR reference values. The results reveal the pollution potential of the mining activities on the surrounding areas.

Keywords: uranium, thorium, tin mining, soil, food, TENORM

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1. Introduction

The average activities of ^{238}U and ^{232}Th in the undifferentiated earth crust are in the range of 25 – 50 Bq·kg^{-1}, but, due to their large ion radius, both elements may be especially concentrated in late crystallising rocks such as granites and other alkaline magmatic ores, often accompanied by other incompatible elements like Rare Earth Elements (REE) (UNSCEAR, 2000).

Uranium is characterised by both radiotoxicity and chemical toxicity, but it is the latter which limits its exposure to humans (Oeh et al., 2007a) whereas thorium is to be considered as only radiotoxic. The health hazards associated with these radionuclides stem from their ability to accumulate in human tissues. During the nuclear transformation processes, the radionuclides emit gamma rays as well as high-LET charged particles, thereby causing intensive damage to the tissues where they are localized and, to a lesser extent, to the neighbouring organs.

Radionuclides of both the uranium and thorium decay series can be often present to a high degree in the materials occurring in frame of tin mining activities, which are then to be considered as TENORM (“technologically enhanced naturally occurring radioactive materials”).

The negative impact of tin mining activities on the environment is mainly due to the excavation of large amounts of sand and the eventual accumulation of a large volume of tailings (Banat et al., 2005; Remon et al., 2005; Akinlua et al., 2006; Birkefeld, 2006, Nyarko et al., 2006), which significantly alter the natural constituents of radionuclides in the soil and thus affect the terrestrial ecosystem. It has been observed that mining, milling and processing of uranium- and thorium-bearing minerals lead to enhanced radiation exposures not only to the workers but also to the inhabitants of the mining and processing sites (UNSCEAR, 2000; Lipsztein et al., 2001).

Indiscriminate and improper deposition of tailings, especially on steep slopes, increases their mobility and hence the risk of being transported to large inhabited areas (Henriques and Fernandes, 1991). Due to leaching and re-suspension processes, ^{238}U and ^{232}Th from abandoned dumping sites find their way in surface and ground water (Ragnarsdottir and Charlet, 2000). Consequently, this makes mine tailings a source of pollution to the ground- and surface-waters, and to the soil in their vicinities (Hector et al., 2006). The effect to man is of particular importance when lands that are used for crops production also serve as repository to tailings, thus increasing the risk of human exposure to TENORM. This may occur by inhalation of suspended dust in the air, direct dermal contact and/or by consumption of crops grown on the affected lands.

The tin mining site in Bisichi is located in the Jos Plateau, in the north-central part of Nigeria, and is about 25 km south of Jos City. The Jos Plateau is on the Nigerian basement complex, at about 1100 m above the sea level. The tin ore consists particularly of the mineral cassiterite and can be assigned to two geological epochs (Buchanan et al., 1971). The Pre-Cambrian mineralization is associated with the Older (Pan African) Granite Province and the cassiterite is stored in the pegmatites of the Basement Complex (Jacobson and Webb, 1946). The Jurassic intrusive sequences are represented by non-orogenic Younger Granites, which are outlined in circular or elliptical shape (Bowden et al. 1979). There, the cassiterite is preferably hold in the biotite-bearing phases either as disseminations at the roof zones of the granite bodies or as lodes and fracture controlled greisen veins (Bowden and Kinnard, 1984). The mining activities are exclusively focused on the Younger Granite Complexes due to easier access and started around 1904, shortly after airborne radiometric mapping revealed high deposit of cassiterite and columbite (niobium) ores. Accessory minerals like tantalite, zirconium, monazite, xenotime, and thorite, which have been known to have high concentration of uranium and thorium, were later discovered in the mid-1920s. This discovery led to increased mechanized mining and large mining pits were created due to soil excavation that resulted in ecological devastation of the region. At present, mechanized mining is no longer operational; however, illegal mining activities are present in the area. The method usually employed by illegal miners involves digging of wells to a depth ranging from 10 to 20 meters before tunnelling to different directions. Other methods usually employed by local villagers during the rainy season include surface and sub-surface mining processes in which drainage channels are created. The mineral ore, which is heavier than sand, will be left behind as water is allowed to run through the channel. This method also helps to detect areas where these minerals are located. The mining pits left behind by the mechanized activities are also being re-mined by the local people by channelling the accumulated water pond to other locations as the minerals are mined. The water collected in the dams created as a result of the mining activities is used as drinking water and for irrigation purposes by the communities living around the mining site. Agricultural practices have been operational and the main staple foodstuffs grown in the area are root tubers, cereals and vegetables. Moreover, the neighbouring residents have direct and regular access to these sites, as tailings are being used as building materials (Ademola and Farai, 2006). Hence, enhanced radiation burden posed by these practices remains a source of concern in both occupational and public radiation protection programs.

In view of the unregulated mining activities in the Jos Plateau, internal radiological impact and risk to the inhabitants and workers are of major interest. In some locations in Nigeria, radionuclides have been found in foodstuff and have been reported to be a potential health risk to the public through the dietary pathway (Arogunjo, 2003a; Arogunjo, 2003b; Arogunjo et al., 2005). Recently, extremely high concentrations of natural radionuclides were reported in foodstuffs from a mining site of the Jos Plateau called Bisitchi (Jibiri et al., 2007), and rough estimates of the resulting internal dose were performed. The map presented in Figure 1 of that manuscript locates the site of Bisitchi north-west of Jos City.

In order to monitor incorporation pattern of radionuclides, reliable knowledge of the daily intake from natural radionuclide sources and their metabolic behaviour is indispensable. In Nigeria, the ingestion rate of these radionuclides has not been studied as it has been done in other parts of the globe. The United Nations Scientific Committee on the Effects of Atomic Radiation summarizes reference values of thorium and uranium in environmental materials from other parts of the world with little or no data from Africa (UNSCEAR, 2000). The majority of the measurements on environmental and biological materials were carried out in the North Temperate Zone and may not truly reflect the global average (Santos et al., 2002).

The present study was aimed at evaluating the activity concentration of radionuclides of the uranium and thorium series in soils and mineral samples of the tin mining area of Bisichi and the eventual contamination in well and surface waters as well as in selected foodstuffs and liquids (including alcoholic and soft drinks) consumed by people working and living in that region.

2. Materials and Methods

2.1. Sample collection

Soil, well water, and tap water samples were collected in two locations in the Jos Plateau, specifically in the tin mining area of Bisichi and in the city of Jos, situated about 25 km north of the mining area. Additional samples were collected in the city of Akure, chosen to represent a control area, located 500 km (air distance) south of the Jos

Plateau and without any known mineral mining activity. Additionally, mineral ore samples, water from the mining pits, and food samples (purchased at a local market) were collected in Bisichi; mineral water in sachets, alcoholic and non-alcoholic drinks and cigarettes were purchased from street kiosks in Jos City and Akure. The soil samples were collected at a depth of 10 cm. At least five samples were taken from each location, subsequently pooled together and mixed to obtain a representative average of the desired location. Well water and fresh water samples were collected in 500-ml plastic bottles.

2.2. Sample preparation

2.2.1. Soil and mineral samples

All the loose solid samples were completely dried and then homogenized by sieving for the grain size fraction <2 mm. The materials were carefully weighed and then transferred into radon-densely sealed polyethylene/plexiglas bags to be ready for analysis both at the Physics Department of Università degli Studi di Milano, Italy and at the Helmholtz Zentrum München (HMGU), Neuherberg, Germany.

2.2.2. Food samples

The food samples collected from local farmers were carefully washed to prevent external contamination from soil and atmospheric depositions. Those samples in tuber form were cut to pieces with a knife and were allowed to dry to a constant weight at room temperature. The cereal samples were also kept at room temperature and other samples prone to biodegradation were oven dried at a temperature of 100 °C until a constant weight was achieved. The samples were carefully enclosed in polyethylene bags ready for transportation to Germany. Thereafter, each of the samples was weighted (range between 17.6 g and 506 g) with an analytical balance and was transferred into a tightly sealed polyethylene bag ready for analysis.

2.2.3. Liquid and other samples

The liquid samples, which include alcoholic and non-alcoholic drinks and mineral, well, stream and tap water were kept in tightly sealed plastic bottles and then transferred to the laboratory. The samples were also weighed (range between 100 g and 200 g) and transferred into polyethylene bottles, acidified by the addition of 50 $\mathrm{ml \cdot l^{-1}}$ of concentrated HNO_3 to avoid precipitation, and then stored frozen until analysis. The cigarette samples, which include three different brands, were separated into the three sections that make up the cigarette: tobacco, filter and paper. The various sections were weighed (range between 0.7 g and 17.2 g) and transferred into separate polyethylene bags until analysis.

2.3. Analysis

2.3.1. ICP-MS measurements

All the food, cigarettes and liquid samples were measured at the Central Analytical Service of HMGU, using high resolution sector field ICP-MS Model ELEMENT 1 (Finnigan MAT, Germany). The instrument parameters and the method applied have been described elsewhere (Roth et al., 2005; Oeh et al., 2007b). Prior to measurement, all the solid food and cigarette samples were digested using extraction with aqua regia according to the German standard method DIN 38414-S7. The food samples were first dried again, if needed, and then milled in a grinder and about 100 mg of the milled sample was mixed with 1 ml of nitric acid and pressure digested. Each sample was adjusted to 10 ml with MilliQ water and the measurements were carried out using ICP-

MS. The liquid samples were removed from the storage site, allowed to defrost at room temperature and diluted 1:2 in 5 % HNO_3. An internal standard solution (100 $\mu g \cdot l^{-1}$ of ^{193}Ir) was added to each sample to correct for matrix interferences. Additionally, uranium and thorium standard solutions were used to calibrate the instrument, and reagent blanks using deionised water were also measured at intervals during the entire measurement process.

2.3.2. Gamma-spectrometry measurements

The gamma-spectrometry measurements of the soil and mineral samples were carried out at the Physics Department of Università degli Studi di Milano, Italy and at the Radioanalytical Laboratory of HMGU, Germany. Negative-poled (n-type) high-purity germanium detectors (HPGe) of coaxial shape were used to perform the measurements. The detection limit of the instrument at the HMGU Laboratory is given by 0.5 $Bq \cdot kg^{-1}$ for ^{226}Ra. The obtained spectra were analysed by specific softwares (Canberra GENIE-VMS at HMGU, Ortec MAESTRO at Unimi). Samples and standards were placed into cylindrical containers made of polypropylene/plexiglass (diameter 6 cm, height 3 cm) and properly sealed for radon density. The measurements were performed at least 20 days after the sample preparation in order to let establish secular equilibrium between the parent nuclides uranium and thorium and their short-lived progenies, which were used for determination due to their intense gamma emissions. Uranium-238 was quantified using the gamma line at 1001 keV emitted by ^{234m}Pa; ^{226}Ra using the emissions at 295 keV and 609 keV emitted by its progenies ^{214}Pb and ^{214}Bi, respectively; ^{232}Th was determined by the peaks at 911 keV and 968 keV emitted by ^{228}Ac, ^{228}Th by the line at 583 keV emitted by ^{208}Tl. For the determination of ^{210}Pb at HMGU, the sample was measured in a specific thin layer geometry that enabled to make use of the low energy peak at 46.5 keV.

The measurement time ranged between two hours and six days depending on the activity in the sample, detector efficiency, background contribution etc. The systems were calibrated using reference standards from IAEA and the German PTB (Physikalisch-Technische Bundesanstalt, Braunschweig). Quality assurance was additionally guaranteed by regular participations in national and international intercomparison exercises.

3. Results and Discussion

3.1. Uranium and thorium activity in soils, mineral sands, waste and waters

The activity concentrations of ^{238}U, ^{232}Th, and of their progeny in soils, mineral sands and waste from Bisichi, and in control soils of Jos City and Akure are presented in Table 1. The activity concentrations of ^{238}U and ^{232}Th in water samples of the same regions are presented in Table 2.

It can be observed that the concentrations of radionuclides in the soil samples collected in the mining area of Bisichi are strongly enhanced with respect to the soils of Jos City and Akure, taken as control. Even higher activity concentrations of ^{238}U, ^{232}Th and of their progenies were found in all the mineral sands and their waste products (tailings). The mined minerals tin, columbite and zircon contain such high radionuclide concentrations due to their property of 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}U decay series they are even enriched in the settled suspension.

The mineral sands and waste materials are characterised by a significant radioactive disequilibrium for the ^{238}U decay series, whereas the ^{232}Th decay series is well equilibrated, in case of columbite also in transitory equilibrium. One possible explanation for the disequilibrium of the ^{238}U series is that the extraction process is performed in a reducing milieu, where uranium is insoluble, but radium is removed (Kemski et al., 1996). However, this would not explain the equilibrium in the ^{232}Th chain, as one radium isotope (^{228}Ra) is also present in this chain. The different mobility properties of the radium isotopes might be more reasonably explained on the basis of the alpha-recoil effect. Indeed, the transformation from ^{238}U to ^{226}Ra takes place through three alpha disintegrations, and the recoil effect may induce cracks in solid materials from which ^{226}Ra isotopes might escape. In the ^{232}Th series, only one alpha disintegration step with very long half-life ($1.4 \cdot 10^{10}$ y) leads to the formation of ^{228}Ra, the parent nuclide of ^{228}Ac and ^{228}Th. The lower probability of inducing cracks in the materials would explain why ^{228}Ra and its progeny are less mobile, and why ^{232}Th is in equilibrium with its progeny.

The highly active Bisichi soil samples also showed radioactive disequilibrium, indicating the dimensions of radionuclide contamination of that area as a result of the mining activities. In contrast, the activity concentrations of the reference soil taken at Akure and also of the one from Jos City were not only much lower but also equilibrated. It should be noted that, compared to the control soil of Akure, uranium concentration is slightly enhanced (about two times higher) also in Jos City. This observation is consistent with the elevated uranium activity concentration found in the well and tap waters of Jos City (Table 2), and could be attributed either to geological factors or to the circumstance that tin processing is operational in several locations in the city, so that contamination of ground water by tin tailings deposited around the city may be likely. Uranium concentration higher than those from the control area were also found in all the waters sampled in Bisichi except tap water, which came from a controlled source. On the contrary, thorium in all well and tap samples was found below the detection limit. This finding could be explained by the low solubility of thorium from soil as compared to the ability of uranium to form uranyl complexes with ground water. The activity concentrations of ^{232}Th were measurable only in the river and stream waters from Bisichi. These results are consistent with the possibility of direct ground water pollution by tin mining activity in the area.

3.2. Uranium and thorium activity in foodstuffs and beverages and evaluation of the intake

High activity values of ^{238}U and ^{232}Th were found in all the foodstuffs (see Table 2), especially in yam, cocoyam, potatoes, vegetables, and fish from the Bisichi river. The results highlighted the impact of tin mining on food grown in the area. Concentrations in beverages and drinking waters were on the contrary limited to few $mBq \cdot l^{-1}$.

It was not possible to assess the daily intake of these radionuclides in people working and living in Bisichi, as the activity concentrations of uranium and thorium were determined only in a sub-sample of the foodstuffs and beverages representative of Nigerian diet. The estimate of the total daily intake from data obtained on single food items is generally difficult, and highly standardised sampling procedures must be employed (Galletti et al., 2003). Moreover, radionuclide concentration can be modified during food processing and cooking (Jibiri et al., 2007), and the use of several minor ingredients could either be an additional contributor or an inhibitor to the bioaccessibility of the radionuclides for absorption. However, some indications on the exposure of the Bisichi population were obtained from the measurements presented in this work, since they covered a few of the most frequently consumed items (tubers, cereals), according to a survey conducted by the Nigerian Federal Office of Statistics (Federal Office of Statistics, 2006). In order to calculate the intake from a specific foodstuff, the radionuclide concentrations, measured on a dry-weight basis, had to be converted to fresh-weight using the corresponding dry-to-fresh weight ratios (Cierjacks, 2004; Karl et al., 2006). The annual consumption rates, the activity concentrations in the various foodstuffs, transformed into fresh-weight units, the estimated intake for ^{238}U and ^{232}Th and the resulting doses are presented in Table 3. It can be observed that the highest contributions come from tubers, fish and vegetables, due to the high consumption and/or the high concentration. With regard to leafy vegetables (spinach) no significant difference was found between the radionuclide concentration in the leaves and in the stem. Applying the effective dose coefficients of 0.045 $\mu Sv \cdot Bq^{-1}$ for uranium and of 0.23 $\mu Sv \cdot Bq^{-1}$ for thorium (UNSCEAR, 2000), the committed effective dose from the annual intake of ^{238}U and ^{232}Th are 1.0 μSv and 5.7 μSv, respectively. It should however be kept in mind that these values are only a minor portion of the ingestion dose, as higher doses (up to 2 orders of magnitude) can be delivered by the daughter radionuclides that are also present in the food and as such incorporated together with ^{238}U and ^{232}Th (Santos et al., 2002).

3.3. Uranium and thorium activity in cigarettes.

Cigarettes were also investigated, as they are known to be an additional source of uranium and thorium intake (Rawat et al., 1992; Santos et al., 1994). The activity concentrations of ^{238}U and ^{232}Th in sections of cigarettes are presented in Table 4. It is shown that ^{238}U activity concentrations were much higher in the wrapping paper than in filter or tobacco. In the case of ^{232}Th, tobacco is the constituent with the highest concentrations, whereas concentrations in the filter were low in two out of the three samples. Paper wraps are primarily made from cellulose fibres from wood pulp and other chemical additives like titanium oxide or zirconium oxides, the latter being an alternative in the event that titanium is not available or very expensive in the production of wrapping paper. Uranium and thorium radionuclides are known to be present in high concentrations in unpurified zirconia, and this may probably explain the values found in the wrapping paper. On the other side, filter is a mixture of charcoal and synthetic fibre materials, the later being a product of oil, coal or natural gas, which may be contributing to the activity in the material.

3.4. Comparison of data with literature

3.4.1. Concentrations in mineral and soil samples

Figure 1 compares the results obtained in this work with previous measurements performed in that same area using NaI(Tl) detectors (Ibeanu, 2003; Jibiri et al., 2007), with some representative regional mean values as presented by UNSCEAR (2000), and with concentrations measured in so-called "areas of high natural radioactive background" (also referred to as HINAR) like the plateau of Pocos de Caldas in Brazil (Amaral et al., 1992) or the Nieu Islands in South Pacific (Marsden, 1960). The bar indicated as UNSCEAR represents the world median concentration in soil (35 $Bq \cdot kg^{-1}$ for ^{238}U and 30 $Bq \cdot kg^{-1}$ for ^{232}Th). The maximum concentrations in normal soils reported by UNSCEAR are 690 $Bq \cdot kg^{-1}$ for ^{238}U and 260 $Bq \cdot kg^{-1}$ for ^{232}Th. The use of HPGe detector in the present works enabled to obtain greater accuracy in the determination of radionuclide concentration and more detailed information on the isotopic composition of the samples.

It can be clearly observed that the values measured in the soils of Jos City and of the control area of Akure do not significantly differ from the rest of the available data compiled by UNSCEAR (2000), whereas the levels of ^{238}U and ^{232}Th measured in the Bisichi mining area (panels i and ii of Figure 1) are higher than those reported for normal soils and also for the HINAR areas. The values reported for contaminated soils near a tailing dump site of Kuru Karama, approximately 7 km west of Bisichi, were between 52 and 1348 ppm for uranium and 245 and 12138 ppm for thorium (Ibeanu, 2003), i.e. 0.6-17 kBq·kg^{-1} for ^{238}U and 1.0-49 kBq·kg^{-1} for ^{232}Th assuming natural isotopic abundances. These ranges are in good agreement with the values observed in this work (8.7 kBq·kg^{-1} for ^{238}U and 16.8 kBq·kg^{-1} for ^{232}Th). Slightly lower concentration values (0.01–0.47 kBq·kg^{-1} for ^{238}U and 0.12–2.2 kBq·kg^{-1} for ^{232}Th) were observed in farm soils from a former tin mining site in the Jos Plateau (Jibiri et al., 2007). The map shown in that work locates however the sampling area (called Bisitchi) as being north-west of Jos City, i.e. far from the Bisichi mining site considered in this work and in (Ibeanu, 2003). The previous works conducted with samples from the Jos Plateau do not provide information on radioactive disequilibrium in the soils.

Uranium-238 and ^{232}Th activities in tailings and mineral sands present in the Bisichi area range from 11 to 51 kBq·kg^{-1} and from 30 to 98 kBq·kg^{-1}, respectively. Figure 2 shows the comparison of the Bisichi data obtained in this work with those reported for processed mineral sands in other parts of the world. Higher values of radionuclides activity of mineral sand found in Bisichi can be seen in the figure; however, the values are comparable to the typical ranges reported by UNSCEAR (2000) for raw zirconium sands (0.2-74 kBq·kg^{-1} for ^{238}U and 0.4-40 kBq·kg^{-1} for ^{232}Th). It is interesting to note that thorium activities were found to be always higher than those of uranium in mineral sands and tailings, as well as in the Bisichi soil, differently from what usually observed in non-contaminated environmental samples. Table 5 shows that the radioactive disequilibrium observed in the tailings and the relative enrichment of the main radionuclides ^{238}U, ^{226}Ra and ^{232}Th are in good agreement with others given in literature for tailings and sediments (Ziqiang, 1988); Chowdhury et al., 1999; Bahari et al., 2007).

3.3.2. Concentrations in water and foodstuffs and daily intake of ^{238}U and ^{232}Th

The activity concentrations of ^{238}U and ^{232}Th in water are presented in Fig. 3, compared with the UNSCEAR reference values of 1 mBq·l^{-1} and 0.05 mBq·l^{-1}, respectively. The measured concentrations of ^{238}U exceed the UNSCEAR reference only in the Bisichi river and in the well waters of Bisichi and Jos City. However, also in these cases, the activities are well below the ranges observed in many works published about the content of uranium in drinking waters: in bottled mineral waters for example, ^{238}U activity can exceed 100 mBq·l^{-1} (Martin Sanchez et al., 1999; Werner et al., 1999; Bagatti et al., 2003), and in private drilled wells it is not uncommon to find concentrations above 1 Bq·l^{-1}, even up to 150 Bq·l^{-1} (Limson Zamora et al., 1998; Hakonson-Hayes et al., 2002; Li et al., 2006). Also for ^{232}Th, the concentrations were definitely below the UNSCEAR reference values, with the exception of the two surface water samples in Bisichi.

Figure 4 reports the radionuclides' concentrations in foodstuffs (expressed on a fresh-weight basis) as compared with other studies elsewhere. It could be observed that some of the foodstuffs presented in this study have elevated activity concentration values compared to other studies, in particular rice, tubers, leafy vegetables and fish, therefore again highlighting the impact of mining activity in the area. Extremely higher values ranging between 4.5-85.5 Bq·kg^{-1} for ^{238}U and up to 89.8 Bq·kg^{-1} for ^{232}Th (expressed per unit dry weight) were reported by Jibiri et al (2007) for crops grown on contaminated soils north-west of Jos City. Such values are more than two orders of magnitude higher than in this work, and more than four orders of magnitude higher than the maximum values observed in non-contaminated areas.

Data on annual intake reported for ^{238}U by UNSCEAR (2000) in different countries range from 5.5-6.2 Bq in countries of North America, 3.2-57 Bq from Asian countries, and 4.4-16 Bq from countries in Europe. The value presented here (23.2 Bq) belongs to the upper range of the published data, and is a factor of four higher than the UNSCEAR reference value of 5.7 Bq. As already stated before, the estimates of intake for the people working and living in Bisichi presented here do not represent the whole diet, since concentration values were measured in an incomplete set of foodstuffs. The UNSCEAR estimates for thorium annual intakes range from 1.1-2.2 Bq, 0.6-9.3 Bq, and 1.2-2.2 Bq in Northern America, Asia and Europe, respectively. A reference value of 1.7 Bq was indicated. The incomplete estimate of the annual intake value of thorium for the inhabitants of Bisichi (24.7 Bq) is therefore a factor of 15 higher than the reference, but, on a daily base, lower than the amount ingested by people consuming therapeutic soils (healing earths) for curing specific disorders (Höllriegl et al., 2007).

The resulting committed doses due to ^{238}U and ^{232}Th do not exceed few µSv per year, thus being well below the levels indicated by (Jibiri et al., 2007).

4. Conclusions

The present work provides data on the activity concentration of radionuclides of the uranium and thorium series in soils and mineral samples of different areas in Nigeria as well as in foodstuffs and liquids consumed by people working and living in those regions.

The results revealed that the highest activity concentrations of ^{238}U and ^{232}Th were found in soils and mineral sands of the Bisichi tin mining area, located on the Jos Plateau. The values (up to 8.7 kBq·kg^{-1} for ^{238}U and up to 16.8 kBq·kg^{-1} for ^{232}Th) agree with previous measurements conducted in the area and are significantly higher than in other so-called areas of high natural radioactive background as e.g. the plateau of Pocos de Caldas in Brazil or the Nieu Islands in South Pacific.

On the contrary, the values measured in the soils in further distances from the mining area of the Jos Plateau (Jos City and Akure) are well within the normal variability range, which is according to UNSCEAR up to 690 Bq·kg^{-1} for ^{238}U and up to 260 Bq·kg^{-1} for ^{232}Th.

The activity concentrations of ^{238}U and ^{232}Th for all collected Nigerian water samples were generally within the UNSCEAR reference values, with the exception of the samples in Bisichi. However, even in these samples, the uranium activities were well below the ranges observed in many works published about the content of uranium in drinking waters.

The results of the measurements in foodstuffs indicated elevated activity concentrations of ^{238}U and ^{232}Th in some foods compared to UNSCEAR reference values, which may be a result of the impact of the mining activity in the surrounding area.

On the basis of the food measurements annual intake values of ^{238}U and ^{232}Th were estimated being at the upper range of reference data from literature. However, the resulting committed doses do not exceed few µSv per year, thus being well below the levels published in a recent paper for the same population.

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Figure captions

Fig. 1. Activity concentrations of ^{238}U and ^{232}Th in soils. Please note the logarithmic scale of the y axis. Panel i): this study. Panel ii): other studies conducted in the same area. Panel iii) UNSCEAR listing of non-contaminated soils (UNSCEAR, 2000). Panel iv) other areas with high natural radioactive background. (a) (Jibiri et al., 2007); (b) (Ibeanu, 2003); (c) (Amaral et al., 1992); (d) (Marsden, 1960).

Fig. 2. Comparison between the activity concentrations of ^{238}U and ^{232}Th in mineral sands and tailings and literature data from different parts of the world: (a) (Hartley, 2001); (b) (Deng et al., 1997); (c) (Haridasan et al., 2001); (d) (Mohanty et al., 2003); (e) (Hofman et al., 2000).

Fig. 3. Activity concentrations of ^{238}U (upper panel) and ^{232}Th (lower panel) in waters. The symbols “X” represent measurements below the detection limit. The horizontal lines refer to the reference values indicated by UNSCEAR (UNSCEAR, 2000).

Fig. 4. Comparison between the activities of ^{238}U and ^{232}Th in foodstuffs measured in the present and in previous studies. A: Cereals, B: Legumes, C: Tubers, D: Vegetables. (a) (Santos et al., 2002); (b) (Shiraishi, 2000); (c) (Pietrzak-Flis et al., 1997); (d) (Fisenne, 2000); (e) (Pietrzak-Flis et al., 2001). The UNSCEAR data correspond to the reference values as given in Table 15, pages 124-125 of the UNSCEAR report (UNSCEAR, 2000).

Table 1

Activity concentrations of ^{238}U, ^{226}Ra, ^{210}Pb, ^{232}Th and ^{228}Th in soils, mineral sands and waste determined by gamma-spectrometry.

Sample Name	Activity concentration (kBq·kg^{-1})				
	^{238}U$^{(a)}$	^{226}Ra$^{(b)}$	^{210}Pb	^{232}Th$^{(b)}$	^{228}Th$^{(a)}$
Soils					
Soil (Akure)	0.066 ± 0.011	0.041 ± 0.010	0.071 ± 0.005	0.053 ± 0.001	0.050 ± 0.002
Soil (Jos City)	0.15 ± 0.02	0.109 ± 0.005	0.157 ± 0.006	0.059 ± 0.004	0.058 ± 0.002
Soil (Bisichi)	8.7 ± 0.5	4.2 ± 0.6	4.60 ± 0.11	16.8 ± 0.8	18.10 ± 0.12
Mineral sands (Bisichi)					
Columbite (dress) $^{(c)}$	40 ± 3	14.2 ± 1.0	9.6 ± 1.1	92.0 ± 1.6	103.4 ± 0.8
Columbite (burnt) $^{(c)}$	30.0 ± 1.9	15.5 ± 0.2	9.35 ± 0.14	45.62 ± 0.05	57.9 ± 0.5
Tin	11.0 ± 1.3	7.4 ± 0.6	1.16 ± 0.09	32 ± 4	30.6 ± 0.3
Zircon	51 ± 3	31 ± 2	10.31 ± 0.15	98 ± 3	94.6 ± 0.8
Waste (Bisichi)					
Tin Tailing	22 ± 2	13.7 ± 0.9	7.3 ± 0.6	30.0 ± 0.3	29.4 ± 0.3

(a) Results of the HMGU measurements ± measurement uncertainties

(b) Average of the gamma-measurements at the two laboratories ± SE

(c) "Columbite (dress)" refers to the original processed form of the ore; while "Columbite (burnt)" is the result obtained after heating in a furnace the processed ore in order to improve its mineral quality.

Table 2.:

Activity concentrations of ^{238}U and ^{232}Th in waters, beverages and foodstuffs measured by ICP-MS

Sample description	Activity concentration (mBq·kg^{-1})	
	^{238}U	^{232}Th
Waters		
River water (Bisichi)	1.82±0.03	0.25±0.02
Stream water (Bisichi)	1.00±0.01	0.22±0.02
Well water (Bisichi)	1.27±0.01	< 0.002
Well water (Jos City)	3.28±0.02	< 0.002
Well water (Akure)	0.41±0.01	< 0.002
Tap water (Bisichi)	0.050±0.001	< 0.002
Tap water (Jos City)	0.246±0.004	< 0.002
Beverages		
Sachet water (Jos)	0.18±0.01	< 0.002
Sachet water (Akure)	0.77±0.01	< 0.002
Hot alcoholic drink	0.48±0.01	< 0.002
Guilder alcoholic drink	0.58±0.24	0.03±0.04
Harp alcoholic drink	0.70±0.01	0.04±0.02
Maltina	0.30±0.01	0.015±0.023
Vegetables[a]		
Cabbage	32.4±0.3	70.3±1.7
Cucumber	105±2	128±3
Spinach	1873±19	1279±8
Beans and Cereals[a]		
White Bean	33.9±0.4	62.0±1.6

Green Bean	455±5	439±11
Rice	49.6±0.8	96±2
Maize	21.0±0.2	57.6±1.5
Tubers[a]		
Cocoyam	266±3	298±8
Yam	248±3	496±13
Irish Potato	465±4	1060±30
Sweet Potato	375±3	1100±30
Animal[a]		
Fish (River Bisichi)	1360±20	576±14

[a] Dry weight

Table 3. The annual consumption rates, the activity concentrations (fresh weight) in the foodstuffs and the resulting committed doses for ^{238}U and ^{232}Th

Food samples including liquids	Consumption (kg·y^{-1})	^{238}U			^{232}Th		
		Concentration (mBq·kg^{-1})	Annual intake (Bq·y^{-1})	Committed dose (μSv·y^{-1})	Concentration (mBq·kg^{-1})	Annual intake (Bq·y^{-1})	Committed dose (μSv·y^{-1})
Yam	75	53	3.98	0.179	107	8.03	1.85
Leafy vegetables	60	187	11.2	0.505	128	7.68	1.77
Rice	26.4	43.2	1.14	0.051	84	2.22	0.510
Maize	20.7	4.2	0.087	0.004	11.5	0.24	0.055
White Bean	18.6	3.4	0.063	0.003	6.2	0.12	0.027
Fish	15	272	4.08	0.184	115	1.73	0.397
Sweet Potato	14.4	81	1.17	0.053	237	3.41	0.785
Cocoyam	6.5	57	0.37	0.017	64	0.42	0.096
Cabbage	5.0	3.9	0.02	8.8e-4	8.4	0.042	0.010
Irish Potato	3.2	100	0.32	0.014	226	0.72	0.166
Cucumber	3.0	4.2	0.013	5.7e-4	5.1	0.015	0.004
Green Bean	0.02	45.5	0.9e-3	4.1e-5	43.3	0.9e-3	2.0e-4
Water/Beverages/Alcohol	584*	1.27	0.74	0.033	0.1	0.058	0.013
TOTAL	**-**	**-**	**23.2**	**1.0**	**-**	**24.7**	**5.7**

* Estimated in the present study

Table 4

Activity concentrations of ^{238}U and ^{232}Th in cigarettes measured by ICP-MS

Sample description	Activity concentration (mBq·kg^{-1})	
	^{238}U	^{232}Th
Yes (Filter)	96.3±1.0	348±9
Yes (Paper)	1419±16	217±6
Yes (Tobacco)	175±2	349±8
Aspen (Filter)	26.1±0.6	1.3±0.5
Aspen (Paper)	7750±70	108±3
Aspen (Tobacco)	153.0±1.9	279±7
Super kings (Filter)	27.7±0.3	10.1±0.7
Super kings (Paper)	6060±60	84±2
Super kings (Tobacco)	348±5	890±20

Table 5

Examples of radionuclide ratios for ^{238}U, ^{226}Ra and ^{232}Th in tin tailings and sediments

Sample site	$^{226}Ra/^{238}U$	$^{232}Th/^{238}U$	$^{232}Th/^{226}Ra$
Bisichi (this study)	0.73	1.59	2.19
Malaysia (Bahari et al., 2007)			
Discharge point 1	0.47	1.27	2.72
Discharge point 2	0.42	1.56	3.71
Mean	0.59	1.49	2.92
China (Ziqiang, 1988)	0.81	1.45	1.8
Bangladesh (Chowdhury et al., 1999)	0.95	1.97	2.09

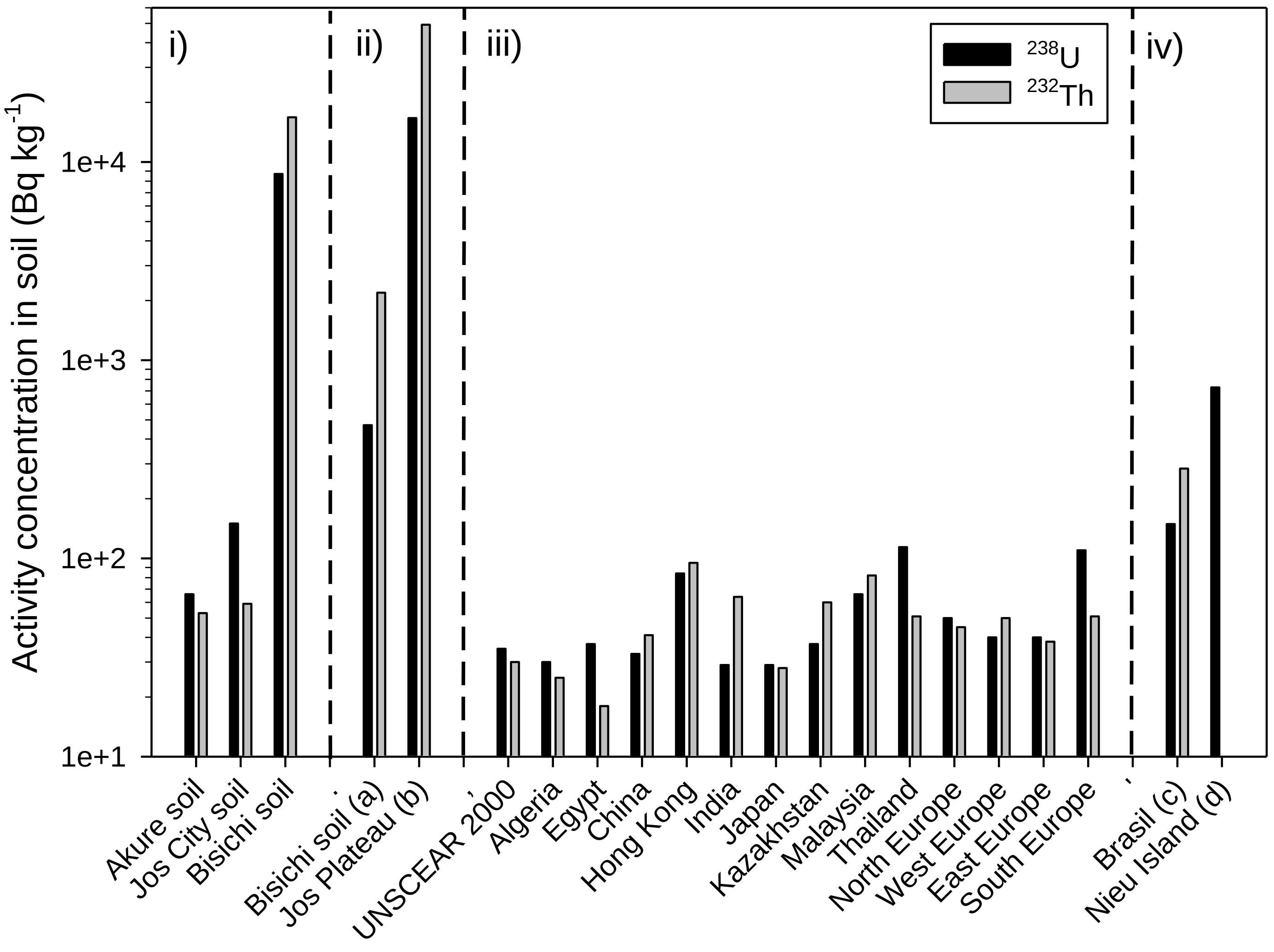

i)
ii)
iii)
iv)
^{238}U
^{232}Th
Activity concentration in soil (Bq kg^{-1})
1e+1
1e+2
1e+3
1e+4
Akure soil
Jos City soil
Bisichi soil
Bisichi soil (a)
Jos Plateau (b)
UNSCEAR 2000
Algeria
Egypt
China
Hong Kong
India
Japan
Kazakhstan
Malaysia
Thailand
North Europe
West Europe
East Europe
South Europe
Brasil (c)
Nieu Island (d)

FIGURE 1

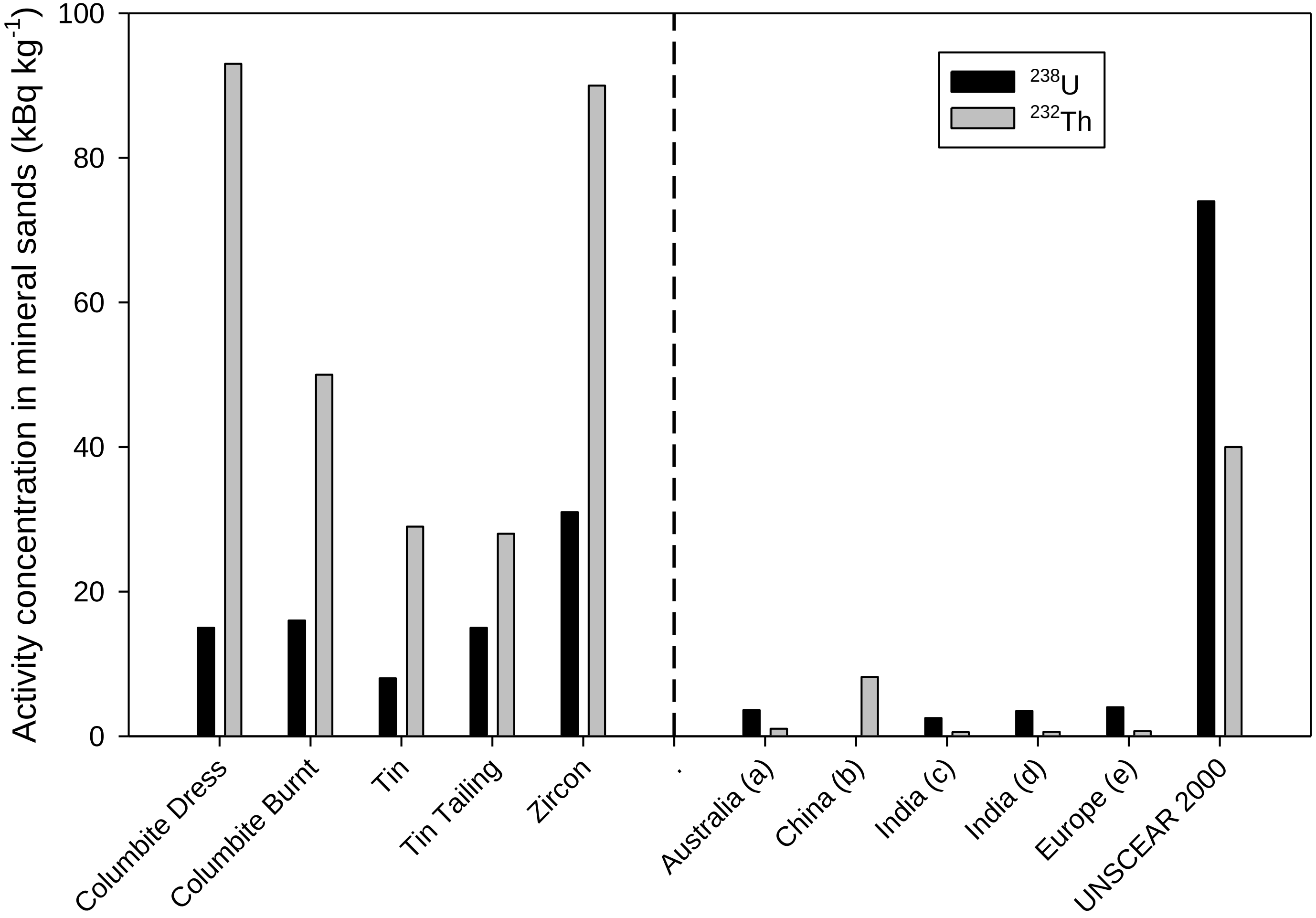
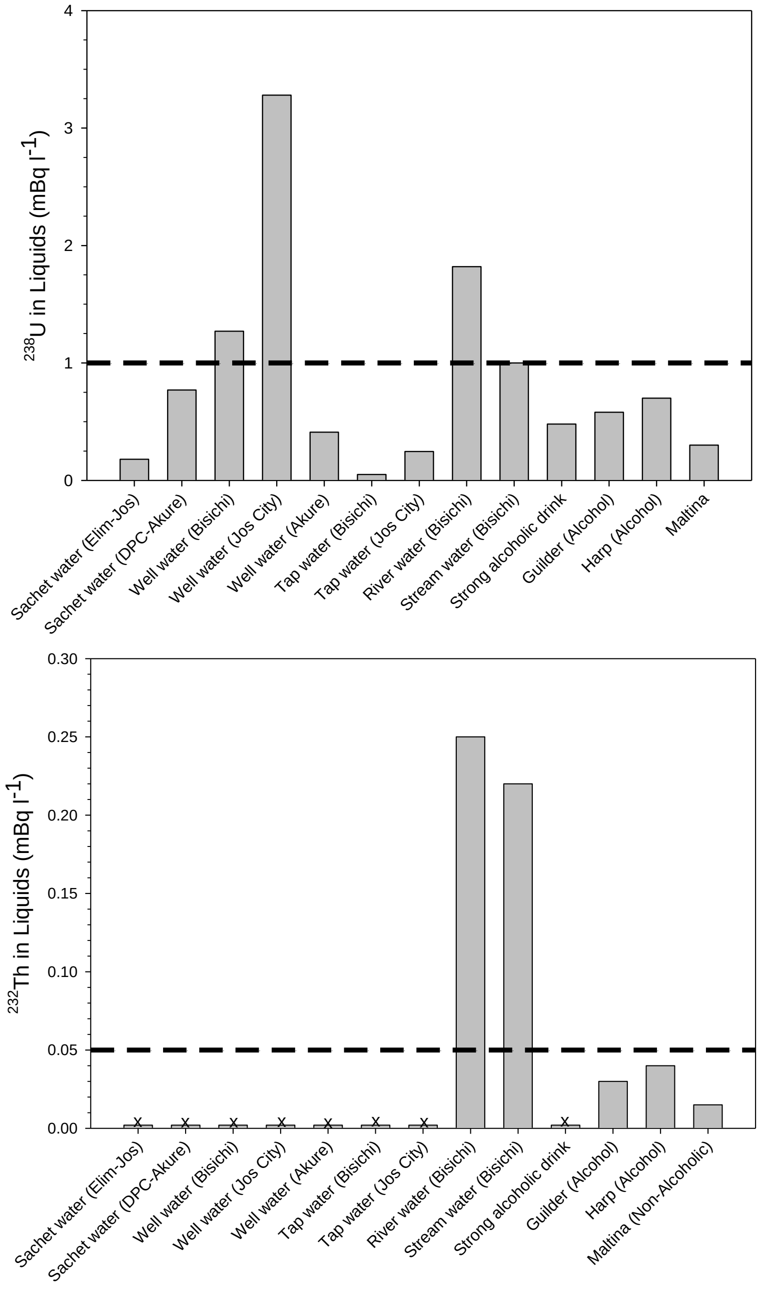

Activity concentration in mineral sands (kBq kg^{-1})
0
20
40
60
80
100
^{238}U
^{232}Th
Columbite Dress
Columbite Burnt
Tin
Tin Tailing
Zircon
.
Australia (a)
China (b)
India (c)
India (d)
Europe (e)
UNSCEAR 2000

FIGURE 2



^{238}U in Liquids (mBq l^{-1})
4
3
2
1
0
Sachet water (Elim-Jos)
Sachet water (DPC-Akure)
Well water (Bisichi)
Well water (Jos City)
Well water (Akure)
Tap water (Bisichi)
Tap water (Jos City)
River water (Bisichi)
Stream water (Bisichi)
Strong alcoholic drink
Guilder (Alcohol)
Harp (Alcohol)
Maltina
^{232}Th in Liquids (mBq l^{-1})
0.30
0.25
0.20
0.15
0.10
0.05
0.00
x
Sachet water (Elim-Jos)
Sachet water (DPC-Akure)
Well water (Bisichi)
Well water (Jos City)
Well water (Akure)
Tap water (Bisichi)
Tap water (Jos City)
River water (Bisichi)
Stream water (Bisichi)
Strong alcoholic drink
Guilder (Alcohol)
Harp (Alcohol)
Maltina (Non-Alcoholic)

FIGURE 3

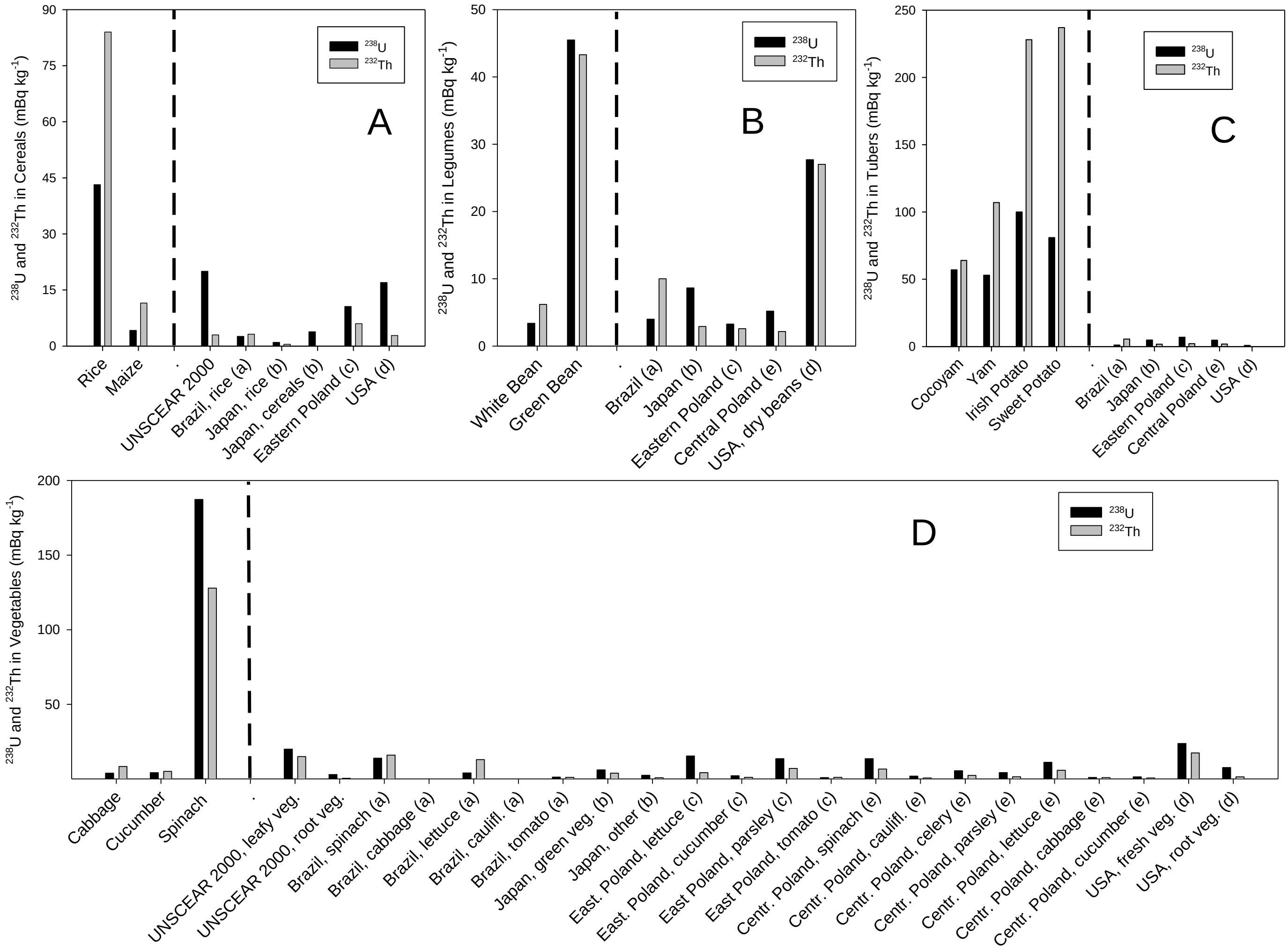

A
90
75
60
45
30
15
0
^{238}U and ^{232}Th in Cereals (mBq kg^{-1})
^{238}U
^{232}Th
Rice
Maize
UNSCEAR 2000
Brazil, rice (a)
Japan, rice (b)
Japan, cereals (b)
Eastern Poland (c)
USA (d)
B
50
40
30
20
10
0
^{238}U and ^{232}Th in Legumes (mBq kg^{-1})
^{238}U
^{232}Th
White Bean
Green Bean
Brazil (a)
Japan (b)
Eastern Poland (c)
Central Poland (e)
USA, dry beans (d)
C
250
200
150
100
50
0
^{238}U and ^{232}Th in Tubers (mBq kg^{-1})
^{238}U
^{232}Th
Cocoyam
Yam
Irish Potato
Sweet Potato
Brazil (a)
Japan (b)
Eastern Poland (c)
Central Poland (e)
USA (d)
D
200
150
100
50
^{238}U and ^{232}Th in Vegetables (mBq kg^{-1})
^{238}U
^{232}Th
Cabbage
Cucumber
Spinach
UNSCEAR 2000, leafy veg.
UNSCEAR 2000, root veg.
Brazil, spinach (a)
Brazil, cabbage (a)
Brazil, lettuce (a)
Brazil, caulifl. (a)
Brazil, tomato (a)
Japan, green veg. (b)
Japan, other (b)
East. Poland, lettuce (c)
East. Poland, cucumber (c)
East Poland, parsley (c)
East Poland, tomato (c)
Centr. Poland, spinach (e)
Centr. Poland, caulifl. (e)
Centr. Poland, celery (e)
Centr. Poland, parsley (e)
Centr. Poland, lettuce (e)
Centr. Poland, cabbage (e)
Centr. Poland, cucumber (e)
USA, fresh veg. (d)
USA, root veg. (d)

FIGURE 4