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Intake of ^{238}U and ^{232}Th through the consumption of foodstuffs by tribal populations practicing slash and burn agriculture in an extremely high rainfall area

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ABSTRACT

The concentration of naturally occurring radionuclides ^{232}Th, ^{238}U was determined using Instrumental Neutron Activation Analysis (INAA) in different food groups namely cereals, vegetables, leafy vegetables, roots and tubers cultivated and consumed by tribal population residing around the proposed uranium mine. The study area is a part of rural area K. P. Mawthabah (Domiasiat) in the west Khasi Hills District of Meghalaya, India located in the tropical region of high rainfall that remains steeped in tribal tradition without much outside influence. Agriculture by Jhum (slash and burn) cultivation and animal husbandry are the main occupation of the tribal populations. A total of 89 samples from locally grown food products were analyzed. The concentration of ^{238}U and ^{232}Th in the soil of the study area was found to vary 1.6–15.5 and 2.0–5.0 times respectively to the average mean value observed in India. The estimated daily dietary intake of ^{238}U and ^{232}Th were 2.0 μg d^{-1} (25 mBq d^{-1}) and 3.4 μg d^{-1} (14 mBq d^{-1}) is comparable with reported range 0.5–5.0 μg d^{-1} and 0.15–3.5 μg d^{-1} respectively for the Asian population.

1. Introduction

The presence of primordial radionuclides in human habitats has always been a source of prolonged exposure. Measurements of naturally occurring radionuclides in the environment can be used as a reference baseline to evaluate the impact caused by nuclear and non-nuclear activities (Amiro et al., 1996). There have been several studies of naturally occurring radionuclide concentrations in foodstuffs from normal background areas, such as those for ^{238}U (Azam and Prasad, 1989; Fisenne et al., 1987; Hamilton, 1972; Long, 1987; Yu and Mao, 1995), ^{226}Ra (Fisenne et al., 1987; Pietrzak-Flis et al., 1997; Radhakrishna et al., 1996; Shukla et al., 1994; Toader, 1993; Yu and Mao, 1995), ^{232}Th (Fisenne et al., 1987; Pietrzak-Flis et al., 1997) and ^{210}Pb (Carvalho, 1995; Lalit et al., 1980; Pietrzak-Flis et al., 1997; Smith-Briggs and Potter, 1986). The daily ingestion of the ^{238}U and ^{232}Th has been reported to be 1.9 μg and 3 μg respectively for ICRP Reference Man (Iyengar et al., 2004a) whereas proposed UNSCEAR values for the entire world (global average) populations are 15.6 mBq (1.3 μg) for ^{238}U and 4.6 mBq (1.1 μg) for ^{232}Th (UNSCEAR, 2000).

The current study was carried out in Meghalaya, one of the seven states in the northeastern part of India, where 85.5% of the population are tribal (National Centre for Health Statistics, 1987). The study area, known as Kylleng Pyndengsohiong (KP) Mawthabah (Domiasiat), Meghalaya, India, consists mainly archean rock formations containing rich deposits of valuable minerals such as coal, limestone and uranium. The region represents a unique set of topographic, economic, sociologic, and cultural conditions that differs from the rest of the country and most of the rest of the world (Parvathi and Goswami, 1989). The study area is also a part of rural area that remains steeped in tribal traditions without much outside influence. Agriculture by *Jhum* (slash and burn) cultivation and animal husbandry are the main occupations of the tribal populations (Ramakrishnana and Patnaik, 1992). Shifting cultivation occurs during December/January and then land is cleared by way of cutting forest vegetation, allowing it to dry in the field, setting fire to the dried vegetation in March/April and cleaning the slope of the remaining trash. Many crops such as rice (paddy), maize and topical beans are planted during the first year and in the second year usually rice (paddy) is grown. In Meghalaya, where industrialization is virtually non-existent, a major portion of the population depends on agriculture for their livelihood, and their diet is

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supplemented by food gathered from the forest (Agrahar-Murugkar and Pal, 2004).

The Domiasiat area receives an average annual rainfall of about 12,000 mm. This predominantly affects the geochemical behaviour of the surface soil. This unique climate condition favours elements such as Th, Sc, V, Cr and Nb to change into more stable oxidation states due to acid and well-aerated soils resulting in high accumulation in soil (Marques et al., 2004). The area is identified as a rich uranium deposit for India. In India most of data concerning long lived naturally occurring radionuclides has been obtained from southern part of the country (Shanthi et al., 2010).

The unique climate and tribal diets in the area make this an important study site, and provide the opportunity to contribute to the world literature on radionuclide intake under lifetyles that are different from those usually considered in risk assessment (e.g., Klos and Albrecht (2005).

2. Materials and methods

2.1. Study area and sampling

Domiasiat lies in plateau state is famous for its rain, mist and rolling hills and glens. It is bounded on the North, East and West by Assam and on the South by Bangladesh. Meghalaya lies between 25°47″–26°10″ North latitude and 89°45″–92°45″ East longitude. Different geological components with diverse lithological, structural and metallogenic complexities are present, and there are identifying areas of commercially important mineral deposits. The Southern slopes of Khasi Hills receive maximum rainfall (12,000 mm per year) in Sohra or Mawsynram and these are known as highest rainfall areas of the world (Fumie Murata et al., 2007). Physiographically, the area forms a westerly sloping undulating hilly terrain with an average elevation of 1400 m above mean sea level. The climate is tropical and humid with temperature ranging between 2° to 28 °C with maximum during May–June and falling to 2 °C in the month of January. The Atomic Mineral Directorate for Exploration and Research (AMD), responsible for exploration of uranium, discovered large sandstone type, near-surface uranium ore deposits at Domiasiat.

The sampling programme focused on the diet of the people who live near the proposed uranium-mining site. The agricultural practices adopted by the local tribes are different from the conventional farming technique. The generally consumed locally grown vegetables, cereals and fruit were sampled from different villages covering 10 km radius around proposed mining site. The samples were categorized in different food groups such as cereals, leafy vegetables, non-leafy vegetables, roots and tubers, fruits and flesh food. For the fish samples which were consumed by the local population, sampling sites were in the river Kynshi, a major tributary of the Jadukata river. It originates from the West Khasi Hills district of Meghalaya and flows near the uranium ore deposits of the study area. Beef samples were collected from local village market. The soil, vegetation/vegetables, food crops and flesh food are the main sample matrices to be studied, based on the diets reported by Agrahar-Murugkar and Pal (2004). A total of 89 samples were collected. Sample collection and preservation was done as per HASL-300, 1990 guidelines and the International Geological Correlation Programme (IGBP-1992) (UNESCO, 1995; Eden and Bjorklund, 1994; Harley J. H., 1988; Hiroshige et al., 1977). The depth of soil taken was 20 cm as per IAEA-TECDOC-1616 (Fesenko et al., 2009; Fesenko and Voigt, 2009). Nine villages were selected for sampling, namely Umdohlun, Wahkaji, Phalangdiloin, Domiasiat, Kylleng, Syngkai, Mawthabah, Nongbah Jynrin, and Nongtynger (Fig. 1). In addition to the present study, the data will act as a baseline for monitoring during mining and post mining operations.

2.2. Sample processing and analytical techniques

Samples were washed and peeled, when necessary dried in air and weighed for determination of the fresh mass. After that they were oven dried at 80 °C for approximately 16 h. Then the samples were ashed for 24 h in oven at 450 °C. Ashed weight of the samples was recorded. Instrumental Neutron Activation Analysis (INAA) was used for determination of uranium (Yaprak et al., 1998). In INAA, about 60–100 mg of the homogenized, ashed and powdered samples together with certified reference material were irradiated for 7 h. After a suitable delay, the samples were measured for 50,000 s in a gamma spectrometer containing an HPGe detector (25% relative efficiency and 1.9 keV resolution) coupled to a PC based 8k multichannel pulse height analyzer. The analyzer had reproducible sample to detector geometry. Energy calibration and efficiency evaluation of the high resolution gamma-ray spectrometer was done by using standards (RGK-1, RGTh-1, RGU-1 and IAEA-152) obtained from International Atomic Energy Agency (IAEA). The decay of radionuclides was followed to confirm the identity of the isotopes being measured. The analysis of ^{238}U was made by measuring the radioisotopes ^{239}Np formed in the n,γ reaction of ^{238}U. The peaks used for analysis were the 277.5 and 228.2 keV peaks of ^{239}Np (Puranik and Jha, 2010). The analysis of ^{232}Th was carried out by using the 311.9 keV of ^{233}Pa. The concentration was evaluated by comparing net counts of uranium and thorium in samples with those obtained from the certified reference standard IAEA-140/TM sea plant.

Spatial distributions of ^{238}U and ^{232}Th in the soil samples were measured by gamma spectrometry as described by War et al. (2008). The gamma spectrometric system comprised of a high resolution HPGe detector, amplifier and PC based MCA. The detector is a coaxial 190cc P-type high purity germanium detector (manufactured by Eurisys Mesures, France). It has a resolution of 2 keV for 1.332 MeV gamma energy of ^{60}Co and a relative efficiency of 50% compared to NaI (Tl) detector. The output of the detector was analyzed using a PC based 8k multichannel analyzer system. The detector was surrounded throughout by 7.6 cm thick lead shield to reduce the surrounding background natural radiation. The gamma spectra of all samples were analyzed using SAMPO code software package. The efficiency calibration of the system was carried out using IAEA standard source of uranium ore (RGU-1) in geometry available for sample counting. The source was counted for sufficiently long time for good statistics. Photo peak areas of the most prominent and well-separated photo peaks were calculated. For energy calibration and relative efficiency calibration, ten prominent gamma energies from ^{226}Ra, ^{214}Pb and ^{214}Bi were used. Each sample was measured for an accumulating period of 50,000 s (13.88 h). An empty cylindrical plastic container of the same geometry as the sample was placed in the detection system and counted for 50,000 s in order to collect the background count rates. Assuming the daughter products of U and Th were in equilibrium, the ^{238}U activity of a sample was estimated from the gamma emissions of ^{214}Pb (352 keV) and ^{214}Bi (609.3 keV, 1764 keV). The gamma ray energies from ^{212}Pb (238.6 keV), ^{228}Ac (911 keV) and ^{208} Tl (583.2 keV) were used to estimate the concentration of ^{232}Th. ^{235}U was not considered in the present study since its natural abundance is only 0.72% of the total uranium content. The activity concentrations were calculated from the intensity of each gamma line, taking into account the mass of the sample, the branching ratio of the γ-decay, the time of counting and the efficiency of the detector. Daily intake of uranium and thorium was calculated using the average food consumption rate of local

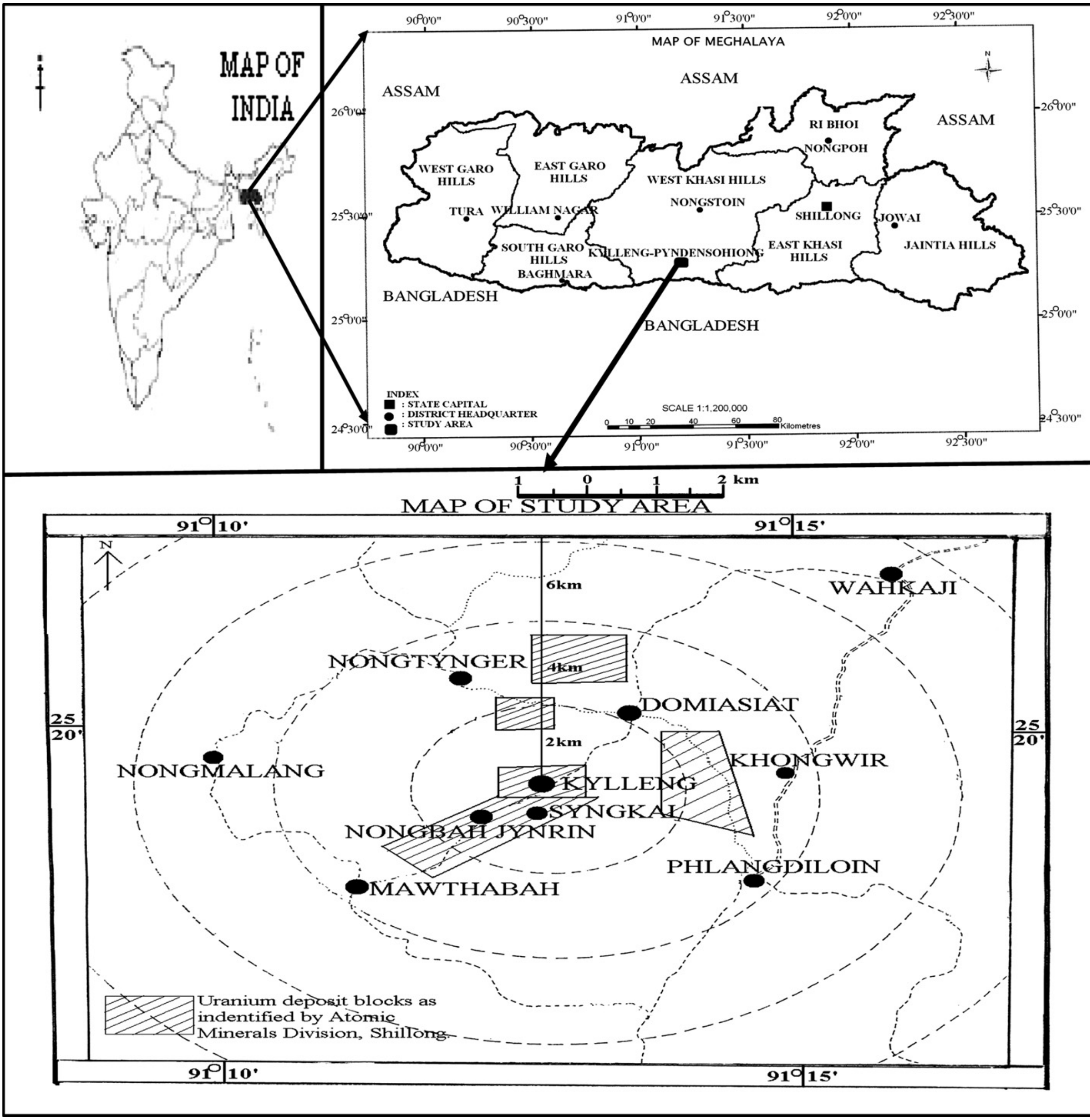

Fig. 1. Map showing location of the study area.

population (Table 3) and the weighted mean concentration of uranium and thorium in each food type.

3. Results and discussion

Natural radioactivity (Table 1) of ^{238}U and ^{232}Th in top soil from study area was found to vary from 32.1 to 980 Bq kg^{-1} and 68 to 441 Bq kg^{-1} respectively, showing maxima at Kylleng. The mean of natural ^{238}U and ^{232}Th activity at Umdohlun, Wahkaji, Domiasiat and Phlangdiloin was found to be 1.6, 1.7, 2.3, 2.3 times and 2.0, 2.3, 2.2, 2.1 times respectively as that of the countrywide average of India. At the other locations Nongtynger, Mawthabah, Nongbah Jynrin, Kylleng and Syngkai, the mean concentration of natural ^{238}U and ^{232}Th was found to vary from 5.5 to 15.5 times and 3.2 to 5.0 times respectively as compared to that of India (Table 1). The higher ratio of the mean value of natural ^{238}U in comparison to Indian and global mean value of ^{238}U in soil reflects that the soil in this area is under the influence of weathering of the uranium containing rock. Natural radioactivity levels in Indian soils were earlier reported by Mishra and Sadasivan (1971) using gamma spectrometry. Their study gives information about the distribution of terrestrial radioactivity in India and the radiation dose likely to be delivered to the population inhabiting in these areas. Countrywide averages were calculated by excluding the contributions from high background

Table 1
Range and mean (Bq kg^{-1}) of naturally occurring radionuclides in top soil from various locations at Domiasiat.

Location	No of samples	^{238}U		^{232}Th	
		Range	Mean	Range	Mean
Umdohlun	11	36–76	48	68–206	128
Wahkaji	12	32.1–87.2	53	95.9–264	147
Phlangdiloin	12	41.4–108.8	72	81.6–264	133
Domiasiat	13	32.7–120	71	105–215	142
Kylleng	13	126.2–980	480	171–399	314
Syngkai	14	101–488	270	104–353	230
Nongbah Jynrin	12	114–517	250	135–281	214
Mawthabah	12	100–459	220	162–441	257
Nongtynger	12	98.8–224	170	93.6–282	203
India		7.0–81	31	14–160	63
Global		16–110	35	11.0–64	30

areas. The concentrations of ^{232}Th and ^{238}U in normal areas vary between 3.5–24.7 Bq kg^{-1} and 2.3–25.5 Bq kg^{-1} respectively. The estimated average global concentration level of ^{238}U and ^{232}Th in soil is 35 Bq kg^{-1} and 30 Bq kg^{-1} respectively (UNSCEAR, 2000). In an independent study carried out by Kamath et al. (1996) in the Indian context, these values were 31 Bq kg^{-1} and 63 Bq kg^{-1} respectively.

Table 2 gives results of analysis (wet weight) of ^{238}U and ^{232}Th in different food products. The ^{238}U and ^{232}Th in leafy vegetables ranged from 3.7 to 8.4 μg kg^{-1} and 6.5 to 12 μg kg^{-1} respectively, in cereals from 0.6 to 3 μg kg^{-1} and 1.3 to 4 μg kg^{-1} respectively, in roots and tubers from 0.4 to 17.4 μg kg^{-1} and 1.1 to 20 μg kg^{-1} respectively and in other vegetables from 3.2 to 9 μg kg^{-1} and 0.4 to 14 μg kg^{-1} respectively. As usually observed in environmental samples, the concentration of radionuclides were better represented by the log normal distribution and the central tendency thus represented by geometric mean (Santos et al., 2002). The geometric means of ^{238}U and ^{232}Th present in flesh, fruit, leafy vegetables, cereals, roots and tubers, and other non-leafy vegetables were found to be 11, 1.0, 6.1, 2.2, 7.8, 6.7 μg kg^{-1} and 14.9, 0.9, 9.3, 2.8, 14, 6.0 μg kg^{-1} respectively. The highest ^{238}U concentrations were found in flesh food and lower levels in locally grown vegetables, cereals, and fish. Shukla et al. (1993) studied concentrations of ^{232}Th in wheat and rice samples collected quarterly from Bombay market for a period of five years. Traces of ^{137}Cs activity were observed in wheat and rice samples in 1987 but in other years it was below the detection limit (0.007 Bq kg^{-1}; 5 kg sample size). The ^{226}Ra values both in wheat and rice were also reported to be below the detection limit (0.019 Bq kg^{-1}) in all the years. The ^{228}Th concentration in rice varied from <0.002 to 0.887 Bq kg^{-1} whereas the same in wheat ranged from <0.002 to 0.224 Bq kg^{-1}. Lal et al. (1983) employed a nuclear track etch technique to estimate trace contents of ^{238}U in most commonly used cereals and pulses grown in various parts of India and reported ^{238}U content in rice varied from 64 to 257 μg kg^{-1} and the same in wheat varied from 71 to 384 μg kg^{-1}. Concentrations of naturally occurring radionuclides in food vary widely because of the variable background levels, prevailing environmental conditions and the characteristics of the different types of plants.

Total intake of ^{238}U and ^{232}Th by the population residing at the Domiasiat and the adjoining area was calculated by summing individual consumption rate of the food item contributing towards total food (g d^{-1}) multiply by weighted mean concentration (μg kg^{-1}) of radionuclide present in each food item. The people living within the 10 km radius of proposed uranium mine are not connected by any means of transportation so that the food from other parts of the country is not accessible to them. They depend solely on the locally grown crops and vegetation grown in forest, a situation described in the risk assessment literature as a high degree of autarchy (Klos and Albrecht, 2005). Table 3 gives the total intake of ^{238}U and ^{232}Th through foodstuff consumed by the local population of Domiasiat. In the present study the intake of ^{238}U and

Table 2
Concentration of uranium and thorium (on wet weight basis) in some foodstuff most frequently consumed by local population in K.P. Mouthabah area.

Foodstuff		Percentage moisture content	Scientific name	No. of samples	Uranium (μg kg^{-1})		Thorium (μg kg^{-1})	
					Mean	Range	Mean	Range
Cereal	Rye	14	*Secale cereale*	5	2.6 ± 0.15	0.6–3.0	4.0 ± 0.12	1.3–4.0
	Maize	15	*Zea mays*	4	0.6 ± 0.05	(2.2)	3.6 ± 1.44	(2.81)
	Rice	14	*Oryza sativa*	6	3.0 ± 0.20		1.3 ± 0.06	
Leafy vegetables	Lettuce	96	*Latuca indica*	4	3.7 ± 0.18	3.7–8.4	6.5 ± 0.26	6.5–12.0
	Mustard leaves	91	*Brassica*	4	8.4 ± 0.25	(6.05)	12 ± 0.36	(9.25)
Non-leafy Vegetables	French beans	73	*Phaseolus domesticus*	6	3.2 ± 0.16	3.2–9.0 (6.68)	14.0 ± 0.42	0.4–14.0 (5.97)
	Gobi	89	*Brassica oleraceae capitata*	5	9.0 ± 0.27		0.8 ± 0.06	
	Cucumber	96	*Cucumis sativum*	4	9.0 ± 0.30		0.4 ± 0.04	
Roots and tubers	Raddish	91	*Raphanus sativus*	4	15.0 ± 0.45	0.4–17.4 (7.83)	1.1 ± 0.11	1.1–20.0 (13.52)
	Garlic Chives	59	*Allium sativum*	4	17.7 ± 0.53		9.2 ± 0.82	
	Carrot	87	*Dacus carota*	4	0.4 ± 0.03		1.3 ± 0.12	
	Sweet potato	78	*Ipomea battatus*	5	3.7 ± 0.15		16.0 ± 0.56	
	Solanum	75	*Solanum nigrum*	5	4.7 ± 0.28		20.0 ± 0.64	
	Potato	79	*Solanum tuberusum*	6	4.7 ± 0.24		20.0 ± 0.64	
	Yam	72	*Colocasea* sp.	5	4.0 ± 0.28		20.0 ± 0.80	
Fruits	Papaya	95	*Carica papaya*	4	1.0 ± 0.10	0.8–1.2 (1.0)	1.1 ± 0.10	0.7–1.6 (0.9)
Flesh food	Beef	71	–	4	24.0 ± 0.72	6.4–24.0	30.0 ± 0.90	8.9–30.0
	Fish	77	–	10	6.4 ± 0.38	(11.4)	8.9 ± 0.10	(14.92)

Values in ( ) indicates geometric mean.

Table 3
Total daily intake of ^{238}U, ^{232}Th through different foodstuffs.

Sr. no.	Foodstuff	Average consumption rate by local population[a] (g d^{-1})	Intake of uranium (μg d^{-1})	Intake of thorium (μg d^{-1})
1	Cereals	390 ± 14	0.86	1.10
2	Leafy vegetables	110 ± 3.3	0.66	1.02
3	Non-leafy vegetables	4.8 ± 1.7	0.03	0.02
4	Roots and tubers	54.4 ± 2.9	0.42	0.73
5	Fruits	20.3 ± 2.1	0.02	0.18
6	Flesh foods	21.5 ± 8.6	0.24	0.31
Total intake (μg d^{-1})			2.01	3.36

1 μg U = 12.6 mBq, 1 μg Th = 4.07 mBq.
[a] Agrahar-Murugkar and Pal, 2004.

^{232}Th was found to be 2.0 μg d^{-1} (25 mBq d^{-1}) and 3.4 μg d^{-1} (14 mBq d^{-1}) compared to the intake of 0.8 μg d^{-1} and 2.2 μg d^{-1} by adult urban Indian population respectively (Dang et al., 1998). The average per capita intake of uranium in food has been reported to be 1.3 μg d^{-1} (Fisenne et al., 1987) and 2–3 μg d^{-1} (Singh, 1990) in the USA, 1.6–15 μg in China and 1.5 μg d^{-1} in Japan (Nozakit, 1970). Iyengar et al. (2004a) showed variation of more than an order of magnitude in intake rate of different Asian countries, the highest mean daily intake was 3.5 μg in Bangladesh and lowest 0.15 μg was reported from Philippines. The daily intake of ^{238}U and ^{232}Th is comparable with the ICRP reference man value of 1.9 μg and 3 μg respectively (Iyengar et al., 2004b). Thus, despite the unique climate and high autarchy of the diets, the total intakes of ^{238}U and ^{232}Th were not very different from other situations.

4. Conclusion

The concentration of radionuclides ^{238}U and ^{232}Th was analysed in soil and locally grown food collected from agriculture field using gamma spectrometry and INAA technique respectively. The concentration of ^{238}U in the Kylleng and Syngkai was found to be 13.7 and 7.7 times higher than the world average indicating the area is under influence of weathering from uranium containing rock.

Among the locally grown food products the highest concentration of ^{238}U and ^{232}Th was observed in roots and tubers followed by non-leafy vegetables, leafy vegetables and cereals. Due to high consumption rate, cereal is the major intake source of these two radionuclides. The present study is the first ever approach to understand the ^{238}U and ^{232}Th intake by tribal population depends on the food grown in the agricultural field prepared using Jhum cultivation in the proposed uranium-mining site Kylleng Pyndengsohiong (KP) Mawthabah (Domiasiat), Meghalaya, India. The daily dietary intake of ^{238}U and ^{232}Th were 2.0 μg d^{-1} (25 mBq d^{-1}) and 3.4 μg d^{-1} (14 mBq d^{-1}) is comparable with range 0.54–5.06 μg d^{-1} and 0.15–3.54 μg d^{-1} respectively for the Asian population reported by Iyengar et al. (2004b).

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