Radioactivity in food crops from high-background radiation area in southwest India

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The study was carried out to evaluate radioactive concentration in food crops grown in naturally high-background radiation areas in southwest India. Seventeen varieties of food crops were collected from different parts of Kanyakumari district. The gross alpha and beta activities of the collected samples were measured using alpha scintillation counter and low beta counter respectively. The alpha activity was maximum in tapioca (497 ± 72 Bq kg^{-1}) and the beta activity was maximum in paddy grain (10,946 ± 583 Bq kg^{-1}). The gamma activity of the food samples was studied by measuring the activity concentration of the radionuclides (^{226}Ra, ^{228}Th, ^{238}U and ^{40}K) in the food crops. The radioactivity content of the food crops from high-background radiation area was higher when compared to similar samples collected from low-background radiation area. The daily radionuclide intake from the food crops grown and consumed by the public was 127.696 Bq and daily internal dose resulting from ingestion of radionuclides in food was 2.34 μSv.

Keywords: Annual dose, daily intake, food crops, radionuclide, Southwest India.

TERRESTRIAL radiation varies from place to place depending upon the variation of radionuclide concentration in soil. Areas where the natural background radiation is higher than normal are called high-background radiation areas. Such areas are prevalent throughout the globe and in our country. Areas with unusually high-background radiation are found in Yangjiang, China, Guarapari, Brazil and Ramsar, Iran. Some areas of Ramsar, a city in northern Iran, have among the highest known background radiation level in the world[1]. High-background radiation areas are of great interest because they present anomalous conditions in their geological and geochemical features and consequently in the background radiation levels. In India, we have high-background radiation areas along the southwest coast[2]. According to Paul and Gupta[3], the monazite deposits occur throughout the erstwhile south Travancore region comprising parts of Kerala and Kanyakumari district, Tamil Nadu. The presence of monazite deposits on the coastal areas of Kerala and Tamil Nadu is due to the weathering of rocks in Nilgris hills and western ghats[4].

The earth and atmosphere contain varied levels of radioactivity due to chain decays of natural radionuclides uranium-238 (^{238}U) and thorium-228 (^{228}Th) and singly occurring radionuclides such as potassium-40 (^{40}K)[5]. Soil features, geological formations and human activities related to radiation and radioactivity are important factors enhancing the background levels of natural radiation[6]. Natural radioactive elements are transferred and cycled through natural processes and between the various environmental compartments by entering into ecosystems and human food chains. Vegetables may be subjected to direct and indirect contamination of uranium-series radionuclides. Use of fertilizers leads to elevation of uranium series nuclides in vegetables[7].

Naturally occurring radionuclides of Th and U are significant contributors of ingestion dose and are present in the biotic system of plants, animals, soil, water and air. Distribution of these radionuclides in different parts of the plant depends on the chemical characteristics and several parameters of the plant and soil. Presence of radioactivity in plant organs has been reviewed by various workers[8,9]. Studies on the radioactivity of the consumable parts of a vegetable assume importance as it is necessary to estimate the ingestion dose to the public.

In this study, we determined the radioactive concentration of the radionuclides ^{226}Ra, ^{228}Th, ^{238}U and ^{40}K (which are either alpha emitters, beta emitters or both) in food crops and in the associated soil collected throughout Kanyakumari using radiation counting methods. We focused on the selected food crops (mango, banana, papaya, guava, pomegranate, Indian caper, drumstick, ladies finger, cucumber, Indian acalypha, tomato, coconut, paddy and tapioca) because they are commonly available and consumed in southwest India.

Experimental work

Sample collection

For the present study, 28 sites (Figure 1) were identified in Kanyakumari. From each site, 4–5 soil samples of approximately 1 kg (wet weight) were collected and analysed

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for radioactivity. About 2 kg (fresh weight) of each food crop selected were also collected and analysed for radioactivity. The work was carried out from February 2007 to May 2008. Plant samples were collected from agricultural farms and vegetable gardens in the region. Kanyakumari is the southern most district of Tamil Nadu and located between 77°15′E and 77°36′E long. and 8°03′N and 8°35′N lat.

Measurement

The soil samples were dried at 150°C and sealed in plastic vessels. These samples were kept at room temperature for about four weeks so that the ^{226}Ra series was able to reach radioactive equilibrium. About 50 mg of the soil sample from each site was crushed into fine powder using agate motor and spread as a fine layer in an aluminium planchet and its gross alpha activity was measured using alpha scintillation counter with ZnS (Ag) detector. Gross beta activity was measured using low beta counting system of 2.5 cpm background (ECIL model/K2700B). The percentage of monazite was measured using Geiger Muller counter coupled to a radiation counting system (ECIL model/RCS 4027 A).

The food crop samples were first washed under running water and then in distilled water to remove all attached sand and dust particles. Samples were weighed and dried in the oven at 140°C for 48 h and the dry weights were recorded. Dried samples were ashed using Muffle furnace at 700°C for 8 h. About 0.02 g of ash was spread as a fine layer in an aluminum planchet and its gross α and gross β activity were measured using the alpha and low beta counter respectively.

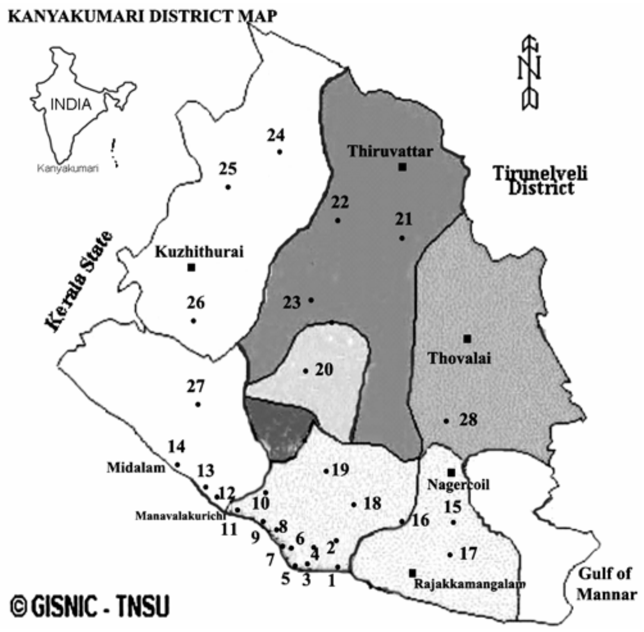

Figure 1. The studied locations in the Kanyakumari District.

The concentration of gamma ray emitting radioisotopes in the samples was measured by employing high efficiency 48 mm × 48 mm NaI (Tl) detector. Assuming that the two primordial radionuclides ^{238}U and ^{228}Th were in secular equilibrium with their corresponding decay products, the ^{238}U, ^{228}Th activity concentration was calculated through 1764 keV of ^{214}Bi and 2614.5 keV of ^{208}Tl respectively. ^{40}K activity was calculated through 1460 keV[10]. The gamma ray energies used to estimate the concentration of ^{226}Ra were ^{214}Bi at 609 keV and 1120 keV[11]. The gamma spectrum was recorded using a PC based multichannel analyser and processed using the NETSWIN software.

Results and discussions

Radioactivity in soil

The activity concentration of soils of 28 sites was estimated. Fourteen sites show appreciable activity concentration of radium, thorium, uranium and potassium compared to the global average values of 28.0, 28.0, 35.0 and 410.0 Bq kg^{-1} respectively[12]. These sites are categorized as high- and low-background radiation areas because the radioactivity was found to be greater than the global mean value (Table 1) and they are located in nearby region of a narrow band of 10 km long via Manavalakurichi. Also, these high-activity regions have the highest monazite content (Figure 2). The gross alpha activity of the high-background radiation area soil ranges from 12,350 ± 270 to 17,270 ± 475 Bq kg^{-1}. And the gross beta activity ranges from 38,420 ± 290 to 53,940 ± 1120 Bq kg^{-1}. The maximum alpha and beta activities were observed in the samples collected from location 14 (Midalam).

The mean values of activity concentration of the radionuclides ^{226}Ra, ^{228}Th, ^{238}U and ^{40}K in the high-background radiation area soil are 44.07 ± 7.07, 215.14 ± 13.9, 67.76 ± 9.14 and 1585.47 ± 62.44 Bq kg^{-1} respectively.

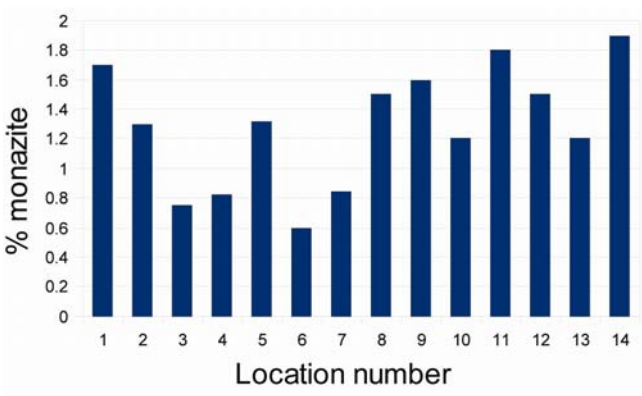

Figure 2. Percentage of monazite concentration vs locations of high-background radiation area.

Table 1. Comparison of average activity concentration of the natural radionuclides

^{226}Ra	^{228}Th	^{238}U	^{40}K	Region	Reference
44.07 ± 17.07	215.0 ± 13.9	67.76 ± 9.14	1585.4 ± 64.4	HBRA	Present study
9.99 ± 1.07	21.59 ± 2.7	23.33 ± 2.63	288.63 ± 32.34	LBRA	Present study
28	28	35	410	World average	UNSCEAR[12]

HBRA, High-background radiation area; LBRA, Low-background radiation area.

All the values are greater than the global average values[12]. The average activity levels of the radionuclides ^{226}Ra, ^{228}Th, ^{238}U and ^{40}K of the soils from the low-background radiation are 9.99 ± 1.07, 21.59 ± 2.77, 23.33 ± 2.63 and 288.63 ± 32.34 Bq kg^{-1} respectively. The variation among the activity levels in soils of different places (Figure 3) may be attributed to the wide variations in geological formations of different types of soil[5]. The statistical analysis (Pearson's correlation, $R^2 = 0.4618$) shows that the thorium concentration presents significant differences in relation to radium activity concentration in soil of high-background radiation area as shown in Figure 4 *a*. But in the soils of low-background radiation areas, the thorium activity is well related to the activity of radium ($R^2 = 0.889$) as given in Figure 4 *b*.

Radioactivity in food crops

The results of these activity concentration measurements on the food crops at the studied site are presented in Table 2. Wide variation in concentration of radionuclides was found in different species of plants and the same species also. A slight modification in any of the factors like physico-chemical characteristics, clay and pH of the soil in which they grow may lead to variability in concentration of the radionuclide, in same food crops or among different food crops samples. Among the 18 food crops collected and analysed from the high-background radiation area, alpha activity was maximum in tapioca (497 ± 72 Bq kg^{-1} fresh) and minimum in Indian caper (Ivy gourd or kovai; 116 ± 14 Bq kg^{-1}). The gross beta activity has been found maximum in paddy grain (10,946 ± 583 Bq kg^{-1}) and minimum in drumstick (190 Bq kg^{-1}). Among the 12 food crops studied from the low-background radiation area, gross alpha was maximum for banana (80.2 ± 10.1 Bq kg^{-1}) and gross beta activity was maximum for Indian acalypha (leafy vegetable; 1032.24 ± 12.4 Bq kg^{-1} fresh). The activity concentration of radionuclides in majority of the samples from these areas was below detecting level (BDL) and so it is not included here.

The activity concentration of ^{226}Ra for vegetables varies between 0.064 ± 0.03 (tomato) and 1.227 ± 0.24 Bq kg^{-1} (drumstick) and it is similar to the results of Lalit and

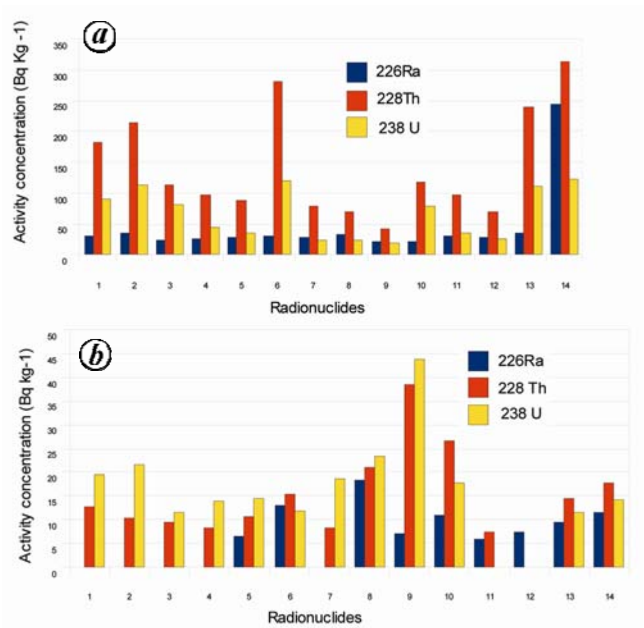

Figure 3. Activity concentration of the ^{226}Ra, ^{228}Th, ^{238}U radionuclides in the soils from (***a***) high-background radiation area; (***b***) low-background radiation area.

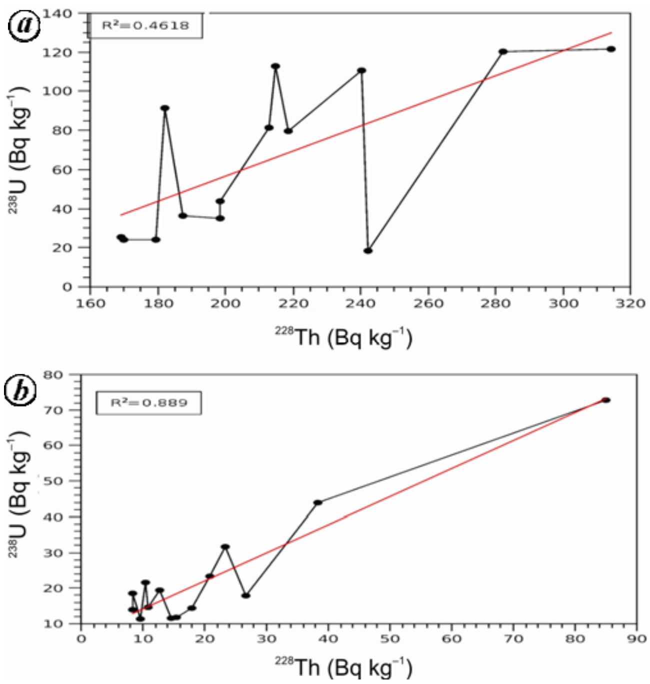

Figure 4. Correlation between ^{228}Th and ^{238}U in the soil of (***a***) high-background radiation area; (***b***) low-background radiation area.

Table 2. Activity concentration of radionuclides in the food crops of high-background radiation area (Bq kg^{-1} fresh)

Food crop	Gross α	Gross β	^{226}Ra	^{228}Th	^{238}U	^{40}K
Fruits						
Mango	165 ± 11	300 ± 43	1.21 ± 0.02	4.5 ± 0.03	0.61 ± 0.02	50.44 ± 8.2
Ripe mango	187 ± 24	323 ± 38	1.32 ± 0.5	3.2 ± 0.2	0.53 ± 0.04	56.23 ± 10.2
Cherry fruit	194.4 ± 21	493 ± 32.5	–	–	–	–
Banana	290 ± 32	600 ± 73	0.094 ± 0.02	0.965 ± 0.4	0.12 ± 0.04	136.2 ± 41.1
Papaya	130 ± 19	1260 ± 136	0.271 ± 0.08	0.184 ± 0.0	BDL	59.56 ± 21.1
Guava	194 ± 17	380 ± 47	–	–	–	–
Pomegranate	186 ± 28	425 ± 53	–	–	–	–
Vegetables						
Indian caper	116 ± 14	240 ± 19	1.05 ± 0.21	5.3 ± 0.02	0.34 ± 0.05	68.69 ± 5.4
Drumstick	133 ± 11	190 ± 23	1.23 ± 0.24	1.7 ± 0.4	0.15 ± 0.02	78.7 ± 9.3
Ladies finger	236 ± 25	495 ± 57	0.378 ± 0.04	0.42 ± 0.2	0.26 ± 0.01	72.8 ± 7.4
Cucumber	226 ± 30	508 ± 62	0.097 ± 0.03	0.14 ± 0.06	0.07 ± 0.03	29.64 ± 9.1
Indian acalypha	214 ± 0.7	2263 ± 343	0.948 ± 0.02	1.5 ± 0.02	0.43 ± 0.041	31.46 ± 2.4
Tomato	336 ± 27	987 ± 89	0.064 ± 0.03	0.17 ± 0.01	0.08 ± 0.02	71.92 ± 8.4
Curry leaves	145.7 ± 9	624.2 ± 21	–	–	–	–
Coconut	127 ± 17	593 ± 73	0.214 ± 0.05	0.34 ± 0.02	BDL	58.4 ± 12.4
Grains						
Rice	347 ± 43	10946 ± 583	3.07 ± 0.02	34.3 ± 11.3	BDL	120.8 ± 2.1
Tuber						
Tapioca	497 ± 72	1267 ± 129	5.42 ± 0.02	107 ± 24.1	BDL	181.1 ± 14.3

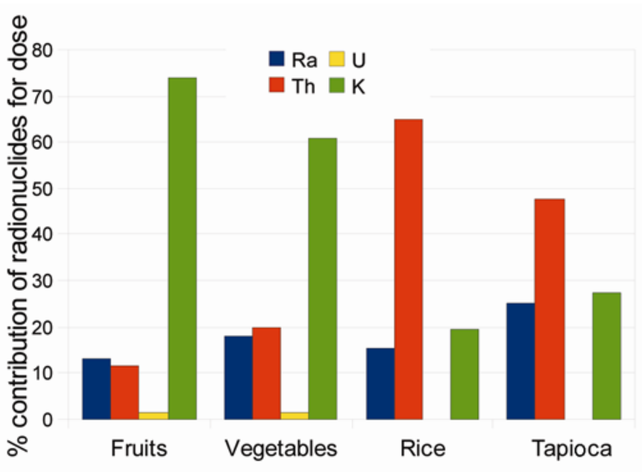

Figure 5. Percentage contributions of radionuclides of interest to effective dose.

Shukla[2]. The ^{226}Ra activity in fruits varies between 0.094 ± 0.02 (banana) and 1.32 ± 0.52 (ripe mango). Fruits and vegetables like cucumber, Indian caper and tomato also have low concentration of these radionuclides. According to this study, tubers (tapioca) have maximum ^{226}Ra activity compared to vegetables or fruits and this result is in good agreement with Mistry *et al.*[13]. Most of the fruit samples from low-background radiation areas have negligible radioactivity and the radioactivity variation could not be studied. The activity concentration of ^{228}Th ranged from 0.14 ± 0.06 (cucumber) to 107.4 ± 24.1 Bq kg^{-1} (tapioca) for the high-background radiation area samples. In the high-background radiation area samples ^{238}U activity varies from BDL to 0.61 ± 0.02 Bq kg^{-1} (mango).

The activity concentration of ^{40}K was less in fruits and vegetables when compared to cereals and tubers. The level of ^{40}K in paddy was 320.28 ± 32.1 Bq kg^{-1} and for tapioca 181.1 ± 14.3 Bq kg^{-1}. The activity concentration of ^{40}K for the studied samples ranged from 29.64 ± 9.1 (cucumber) to 320.28 ± 32.1 Bq kg^{-1} (tapioca). This result agrees with the world range reported by Maul and O'Hara[14] for ^{40}K concentration from 40 to 240 Bq kg^{-1}.

The ^{228}Th concentration was higher than ^{226}Ra and ^{238}U because it is influenced by the ingrowth of ^{228}Th from ^{228}Ra and ^{228}Ac taken up by plants[15]. The concentration of potassium was found to be very high compared to radium and this may be attributed to poor migration characteristics of radium from the substrate to the vegetables in the concerned environment. Potassium is a macronutrient, so the concentration may be high. It may be expected that the soil characteristics favour the mobilization of potassium and its subsequent migration into the plant.

Daily radioisotope intakes from south Indian foods

The major diet of south Indians consists of rice, tapioca, vegetables and fruits. The estimated annual radioisotope intakes from the food consumed are presented in Table 3.

The radioisotopes with the highest daily intake was ^{40}K (107.94 Bq) followed by ^{228}Th; and food with the highest amount of intake is rice. The daily radionuclide intake by ingestion of food was 127.696 Bq of which 84.5% was from ^{40}K. The relative contribution of naturally occurring radioisotopes measured in this study to dose is presented in Figure 5. Potassium-40 is usually of limited interest because as an isotope of an essential element, it is homeostatically controlled in the human cells. As a result, the

Table 3. Annual radionuclide intake and effective ingestion dose

Food crop	Consumption (kg)	Annual radionuclide intake (Bq)				Dose (μSv)
		^{226}Ra	^{228}Th	^{238}U	^{40}K	
Mango	10	12.1	45	6.1	504.4	8.94
Ripe mango	20	26.4	64	10.6	1124.66	17.06
Banana	40	3.76	0.09	4.8	5448	34.71
Papaya	20	5.42	3.68	–	1191.2	8.67
Indian caper	25	26.26	132.5	8.5	217.2	16.25
Drumstick	10	12.27	17	1.5	787	8.49
Ladies finger	25	9.45	10.5	6.5	1820	14.12
Cucumber	35	3.4	4.9	2.45	1037.4	7.54
Indian acalypha	9	8.53	13.5	3.87	283.14	4.52
Tomato	20	1.28	3.4	1.6	1438.4	9.47
Coconut	20	4.28	6.8	–	1168	8.54
Rice	150	460.5	5145	–	18042	569.75
Tapioca	35	189.7	959	–	6338.5	144.3
Annual intake		763.34	6405.3	45.92	39,399.84	852.44

body content of ^{40}K is determined largely by its physiological characteristics rather than its intake. For rice and tapioca, the high radioisotope intake was not due to high radioisotopes concentration but due to the high consumption; rice being the staple food of south Indians.

Conclusion

The mean values of activity concentration of the radionuclides ^{226}Ra, ^{228}Th, ^{238}U and ^{40}K in high-background radiation area soils are 43.07 ± 7.07, 146.14 ± 13.9, 67.76 ± 9.14 and 1572.07 ± 64.44 Bq kg^{-1} respectively. A systematic study was made to evaluate the concentration of radionuclides ^{226}Ra, ^{228}Th, ^{238}U and ^{40}K in locally grown food crops in high- and low-background radiation areas. The activity concentration of ^{226}Ra for vegetables varies between 0.064 ± 0.03 (tomato) and 1.227 ± 0.24 Bq kg^{-1} (drumstick). The radioactivity concentration of ^{228}Th and ^{40}K was found to be very high in paddy grains grown in the high-background radiation areas. Tapioca (edible tubers) accumulates more radionuclides than vegetables. For the estimation of committed effective dose received by the members of the public from such consumables, it is to be ensured that the samples are grown in that particular area only. This study concludes that the committed effective dose received by general public living in high-background radiation area due to the consumption of the fruits, vegetables, rice and tapioca is 852.45 μSv. The daily radionuclide intake by an adult in southwest India through ingestion of food crops is nearly 127.71 Bq/day and daily dose will be 2.33 μSv/day.

1. Andrew Karam, P., WM'02 Conference, Tucsan, AZ, 2002.
2. Lalit, B. Y. and Shukla, V. K., Natural radioactivity in food stuffs from natural high background area of Southern India. Proceedings of 2nd Special Symposium on Natural Radiation Environment, 1982, pp. 43–49.
3. Paul, A. C. and Gupta, V. K., *Radiat. Prot. Environ.*, 1998, **3 and 4**, 173–177.
4. Victor Rajamanickam, G., *Handbook of Placer Deposits*, New Academic Publishers, New Delhi, 2000.
5. Saleh, I. H., Hafez, A. F., Elanany, N. H., Motaweh, H. A. and Naim, M. A., Radiological study on soils, foodstuff and fertilizers in the Alexandria region, Egypt. *Turk. J. Eng. Environ. Sci.*, 2007, **31**, 9–17.
6. ColmeneroSujo, L. *et al.*, Uranium-228 and Th-232 series concentrations in soil, radon-222 indoor and drinking water concentration and dose assessment in the city of Aldama, Chihuahua, Mexico. *J. Environ. Radioact.*, 2004, **77**, 205–219.
7. Ekdal, E., Karali, T. and Sac, M. M., ^{210}Po and ^{210}Pb in soils and vegetables in Kucuk menders basin of Turkey. *Radiat. Measurements*, 2004, **41**, 72–77.
8. Markose, P. M., Studies on the Environment Behaviour of Radium from Uranium Mill Tailings, Ph D thesis, University of Mumbai, 1990.
9. Jibiri, N. N., Farai, I. P. and Alausa, S. K., Activity concentration of ^{226}Ra, ^{228}Th and ^{40}K in different food crops from a high background radiation area in Bitsichi, Jos Plateau, Nigeria. *Radiat. Environ. Biophys.*, 2007, 53–59.
10. Jibiri, N. N. and Banlole, O. S., Soil radioactivity and radiation absorbed dose rates at roadsides in high traffic density areas in Ibadan metropolis, South western Nigeria. *Radiat. Prot. Dosimetry*, 2006, **118**, 453–458.
11. Min-Seok Choi, *et al.*, Daily intakes of naturally occurring radioisotopes in typical Korean foods. *J. Environ. Radioact.*, 2008, **99**, 1319–1323.
12. UNSCEAR, Sources and effects of ionizing radiation. United Nations Scientific Committee on the Effect of Atomic Radiation, United Nations, New York, 2000.
13. Mistry, K. B., Bharatan, K. G. and Gopal Ayengar, A. R., Radioactivity in the diet of population of the Kerala coast including monazite bearing high radiation areas. *Health Phys.*, 1970, **19**, 535–542.
14. Maul, P. R. and O'Hara, J. P., Background radioactivity in environmental materials. *J. Environ. Radioact*., 1989, **9**, 265.
15. Pietrzak Flis, Z., Rosiak, L., Suplinska, M. M., Chrzanowski, E. and Dembinsk, S., Daily intake of ^{238}U, ^{234}U, ^{232}Th, ^{230}Th, ^{228}Th and ^{226}Ra in the adult population of Central Poland. *Sci. Total Environ*., 2001, **273**, 163–169.

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