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Assessment of ingestion dose due to radioactivity in selected food matrices and water near Vizag, India

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Abstract Activity concentrations of ^{238}U, ^{232}Th, ^{226}Ra, ^{40}K and ^{137}Cs were measured in milk, egg, fruit and fish samples collected around a proposed site for setting up nuclear facilities, near Vishakhapatanam. The activity concentrations of the radionuclides ranged from 0.002 to 10.6, 0.002 to 2.8, 0.1 to 7.2, 3 to 110.8, 0.03 to 3 mBq g^{-1} for ^{238}U, ^{232}Th, ^{226}Ra, ^{40}K and ^{137}Cs considering analysed food matrices. Natural uranium was measured in drinking water samples and the values were below 15 ppb. The average ingestion dose was 2.07 ± 2.01, 2.81 ± 4.38, 7.66 ± 8.24, 1.28 ± 0.84 and 0.04 ± 0.05 μSv year^{-1} for ^{238}U, ^{232}Th, ^{226}Ra, ^{40}K and ^{137}Cs in milk, egg, fruit, fish and water. The ingestion dose received was the highest for milk, due to its high ingestion rate. It was observed that ^{226}Ra is the largest contributor of measured radionuclides in this study for the different food matrices analysed due to its high dose conversion factor. The study was carried out as a part of baseline data generation for this region with which future changes in the radiological scenario can be compared.

Keywords Radionuclide · Food matrix · Baseline data · Activity concentration · Ingestion dose

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Introduction

Natural radiation and radioactivity is a fact of life. Naturally occurring radioactive materials are present in all compartments of the environment. Cosmic, terrestrial rays, inhalation and ingestion of radionuclides through air, water and food materials are the different sources of natural radiation [1]. Radioactivity enters the human body mainly by inhalation of radon and thoron and their decay products [2] and also by ingestion of primordial radionuclides and their progeny, like, ^{40}K, ^{238}U series and ^{232}Th series radionuclides [3, 4]. Soils have different concentrations of radionuclides depending on their formation from the parent rocks, geographical location and by geochemical processes [5]. The radionuclides that are present in soils and fertilizers find their way to the human body via the food chain by atmospheric dispersion, gravitational settling, plant uptake and various other geochemical processes.

Foodstuffs are known to contain natural and artificial radionuclides that, after ingestion, contribute to an effective internal dose. The average natural radiation exposure to global population is 2.4 mSv year^{-1} [6]; whereas for Indian population it is reported as 2.3 mSv year^{-1} [7]. It has been estimated that nearly one-eighth of the mean annual effective dose due to natural sources is caused by the intake of food [6]. Hence it is extremely important to monitor the levels of radioactivity in foodstuffs to assess the ingestion dose to members of the public.

The present study was carried out as a part of the baseline survey around a proposed site for setting up nuclear facilities, near Vishakhapatanam (also known as Vizag). It is a prerequisite to establish the baseline radiological data at any nuclear facility before its commencement. The study area is situated around 17°31.417′N and 83°02.143′E in the South Eastern part of India, in the state

Fig. 1 Map of the study area (*source* Google maps)

of Andhra Pradesh. Granitic rocks that serve as good geochemical hosts for U, Th and K are predominant in Vizag. A major portion of this region is occupied by Proterozoic rocks covering a vast time span from 3,800 to 570 million years. The crust in this region shows an enrichment of rare earths with which U and Th are invariably associated in the form of minerals like monazite, samarskite, fergusonite and allanite. Sandstones, limestones, quartzites, slates with poor amounts of U, Th and K are also present [8].

This paper discusses the measurement of radionuclides in common foodstuffs and surface and ground water in the study area with high resolution gamma spectrometry (HRGS) system and laser fluorimetry and calculation of the consequent ingestion dose. Studies on natural radioactivity in food matrices have been reported from certain regions in India [9–12], but there is little data for this part of the country. Hence, this study will serve as the baseline for this area, with which future measurements can be compared to check any possible changes in the radiological scenario of this region.

Sampling and analytical techniques

Sample collection and processing

Foodstuffs were selected according to the consumption practices of local population residing around the study area. Samples were collected from local markets at different locations in the study area as shown in Fig. 1. Foodstuffs such as milk, fruits, eggs and fish were collected and processed according to the IAEA recommendations [13]. Food samples were washed with ultrapure water to remove soil and dust particles. Sample fresh weights were taken and they were put in a furnace for 24 h at 300 °C for ashing (preconcentration). Ash samples were powdered, homogenised and then transferred to plastic containers of standard geometry (polyethylene bottles of 6.5 cm diameter and 7.5 cm height). Water samples collected from the study area included surface water and ground waters. 5 L of water samples were each evaporated up to 250 mL and transferred to the standard containers. Food samples were sealed in standard geometry for 1 month, after drying and

homogenization, to ascertain secular equilibrium between ^{226}Ra and ^{224}Ra and their respective daughter products. Samples were then counted on a gamma spectrometric system for calculation of radionuclide concentrations. Water samples were analysed by Laser Fluorimetry technique for estimation of total uranium concentration.

Radioactivity measurement

An n-type coaxial high-purity vertical germanium detector, of 100 % relative efficiency (DSG, Germany) was used for all the measurements. The detector was surrounded by 7.5 cm thick lead shield with inside dimensions 36 cm (l), 35 cm (b) and 36 cm (h). The resolution of the HRGS was 2.5 keV at 1,332 keV gamma energy of ^{60}Co. IAEA Certified Reference Materials, RGU-I (^{235}U and ^{238}U and their daughter products) and RGTh-I (^{232}Th and daughter products), were used for the energy and efficiency calibration of the HRGS [14]. The gamma spectra were acquired for 1,00,000 s and subjected to pulse height analysis by 8 K PC based MCA. Other details of the counting system and associated electronics are given elsewhere [15]. The energy calibrated detector was used for counting the standards (RGU-I and RGTh-I) with known activities. This enabled the calculation of detector efficiencies at particular energies, which were then subjected to curve fitting procedures to get efficiency versus energy plot. Identical geometry of the samples and standards were ensured by filling containers to the same volumes. The density correction factors were then applied to correct the difference in the densities of samples and standards. IAEA-330 (spinach) and IAEA-445 (spiked water) were analysed for quality control purposes. The obtained results were all within 95 % confidence interval of the recommended values.

The different chemical behavior of uranium and its daughters is responsible for their dissimilar transport behavior in the environment and differential plant uptake. Hence it is unlikely that uranium and its daughter radium will be in equilibrium in environmental matrices; eliminating the possibility of measurement of uranium from the gamma lines of ^{226}Ra daughters. Since ^{238}U emits a very weak low energy γ photon (49.56 keV, 0.064 %) it is measured by the γ rays of its immediate daughter product ^{234}Th at 63.29 and 92.59 keV [16]. The 186.2 keV peak of ^{226}Ra (γ 3.59 %) was used for the estimation of ^{226}Ra after correcting for the contribution from ^{235}U.

The ^{208}Tl and ^{228}Ac radionuclides were measured by the γ energies as given in Table 1 for the estimation of ^{232}Th. The background spectra recorded were used to correct the net γ-ray photopeak areas for the isotopes of interest.

Activity concentration of each radionuclide was calculated using the following equation:

$$A(\mathrm{Bq/kg}) = C(\mathrm{counts})/(E * \gamma * M * T) \quad (1)$$

Table 1 Radionuclide energies and gamma ray intensities used in this study [33]

Radionuclide	Measured nuclide	Energy (keV)	Branching intensity (%)
^{238}U	^{234}Th	63.6	3.75
^{226}Ra	^{226}Ra	186	3.56
^{232}Th	^{208}Tl	583.2	30.7
^{232}Th	^{208}Tl	2,614.5	35.6
^{232}Th	^{228}Ac	911.2	26.2
^{137}Cs	^{137}Cs	662	85.2
^{40}K	^{40}K	1,460.8	10.6

where C is the background subtracted net counts of the sample, E is the efficiency of the detector for the specific gamma ray energy, γ is the absolute transition probability of that specific gamma decay, M is the mass of sample in kg and T is the counting time in seconds.

Laser fluorimetry

Laser fluorimeter is a compact analytical instrument based on the principle of measuring the fluorescence of uranyl complex enhanced by addition of sodium pyrophosphate as fluorescence enhancement reagent. A sealed-off nitrogen laser is the excitation source which emits a very intense, short lived pulse (7 ns) of ultra violet light with wavelength 337.1 ± 0.1 nm. The maximum energy is 20 μJ at a repetition rate of 10 pulses s^{-1}. This excites the uranyl complex in the aqueous medium. A PMT measures the green lights from excited uranium complex at 496–565 nm (4 peaks). The organic matter present in natural water also fluoresces when excited by the nitrogen laser, but has very short life time (<100 ns). The fluorescence of uranyl complex has a longer life time (>25 μs). By measuring the delayed fluorescence signal (a few microsecond after the laser pulse), the unwanted fluorescence of organic compounds are ignored and only the fluorescence of uranyl complex is collected by the time gated PMT. In addition to this, the fluorescence of organic matters has a wavelength maximum around 400 nm whereas that of uranyl complex is around 500 nm. Therefore, the wavelength filters at 450 nm significantly curtail the interferences of organic matters. The water samples were analyzed by standard addition method, in order to avoid the matrix effect. Sodium pyrophosphate ($Na_4P_2O_7{\cdot}10H_2O$) was used as fluorescence enhancement reagent, as well as a complexing agent. The details analysis protocol are given elsewhere [17].

Estimation of committed effective dose (CED)

Radiation dose due to intake of radionuclides through ingestion pathway was calculated using IAEA dose conversion

factors for adults [18]. The daily dietary intake was estimated by using prescribed food intake rates as given by IAEA [19]. The daily intake values considered were 100 mL milk, 20 g fruits, 6 g eggs and 12 g fish. The daily water intake was considered as 4.05 L [19]. The annual radiation ingestion dose was calculated by using the following formula [20].

$$\text{Ingestion dose } \left(\text{mSv year}^{-1}\right) = \text{Conc.}\left(\text{mBq g}^{-1}\right) \times \text{Intake } \left(\text{g year}^{-1}\right) \times \text{DCF}\left(\text{Sv Bq}^{-1}\right) \quad (2)$$

where *DCF* is the dose conversion factor to convert activity to dose for corresponding intake.

Results and discussions

Activity concentrations

The activity concentrations of ^{238}U, ^{232}Th, ^{226}Ra, ^{40}K and ^{137}Cs were 2.58 ± 2.26, 1.11 ± 1.19, 1.78 ± 0.67, 8.78 ± 11.55 and 0.24 ± 0.44 mBq g^{-1} in milk, 2.84 ± 3.37, 0.38 ± 0.22, 1.94 ± 1.20, 24.46 ± 8.54 and 0.93 ± 1.23 mBq g^{-1} in eggs, 5.30 ± 3.63, 0.80 ± 0.77, 5.06 ± 1.35, 43.78 ± 55.99 and 0.35 ± 0.31 mBq g^{-1} in fruits and 0.22 ± 0.38, 0.36 ± 0.6, 0.79 ± 0.59, 29.63 ± 14.52 and <0.03 mBq g^{-1} in fish, respectively. The data are represented by Box-whisker plot in Fig. 2. It can be observed from the figure that ^{232}Th and ^{137}Cs values lie within a narrow range compared to those of ^{238}U and ^{226}Ra. The activity concentrations of ^{40}K lie within a wider range. The wide range of data values in the foodstuffs can be attributed to the variety of food stuff, the area/location of origin of the foodstuff and the transfer of a radionuclide to a particular food item. The lowest concentrations of radionuclides (except ^{40}K) were observed for fish compared to other food items, as can be noted from Table 2. Highest concentrations were observed for ^{40}K and lowest for ^{137}Cs in the food materials. The U concentrations measured in drinking water samples were all below the guideline value of 15 ppb set by WHO [21] and 60 ppb set by AERB [22]. 79 % of the drinking water samples had ^{238}U activity concentrations below 50 mBq L^{-1} as shown in Fig. 3. This constituted eleven out of fourteen samples. These values are comparable with other reported values worldwide except a few high vales [6].

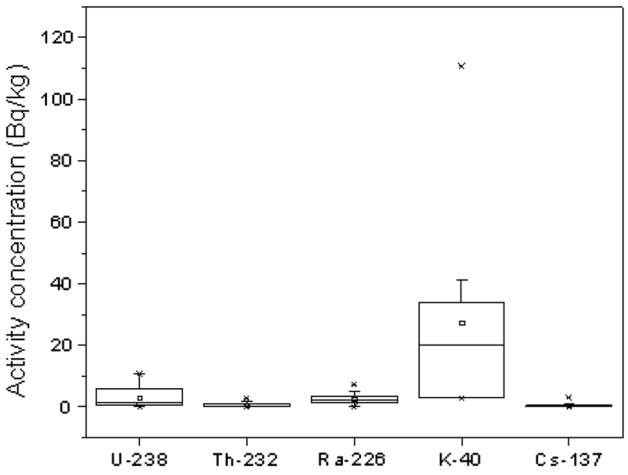

Fig. 2 Box plot of radionuclide activity concentrations in foodstuffs

The data obtained from the present study are compared to activity concentrations of radionuclides obtained from studies carried our worldwide in Table 3. The values are comparable with that reported for India in UNSCEAR [6]. The activity concentrations of radionuclides are in the range of the global values but lesser than the reported data in a high background radiation area in Nigeria [23, 24]. The values in this region are comparable with those reported in Punjab and Bombay in India but lesser than those from a HBRA in Odisha [12]. This can be attributed to the geochemistry of the region and the food items considered [25]. The baseline values observed at this site can be compared with other baseline studies carried out at Tamil Nadu [37] and Tarapur [38] in India and Lebanon [36]. In Tamil Nadu and Tarapur the studies have been carried out before the commencement of nuclear power plants, similar to the present study. Rao et al. [39] have reported data from

Table 2 Activity concentrations of radionuclides in food matrices

Food matrix	Statistical parameter	^{238}U	^{232}Th	^{226}Ra	^{40}K	^{137}Cs
Milk	Average	2.58 ± 2.26	1.11 ± 1.19	1.78 ± 0.67	8.78 ± 11.55	0.24 ± 0.44
(mBq g^{-1})	Range	1.00–5.9	0.25–2.8	1.10–2.70	3.00–26.10	0.03–0.90
Egg	Average	2.84 ± 3.37	0.38 ± 0.22	1.94 ± 1.2	24.46 ± 8.54	0.93 ± 1.23
(mBq g^{-1})	Range	0.05–6.60	0.10–0.60	0.10–3.30	14.90–33.90	0.03–3.00
Fruit	Average	5.30 ± 3.63	0.80 ± 0.77	5.06 ± 1.35	43.78 ± 55.99	0.35 ± 0.31
(mBq g^{-1})	Range	1.50–10.60	0.10–2.00	3.60–7.20	3.00–110.80	0.03–0.70
Fish	Average	0.22 ± 0.38	0.36 ± 0.6	0.79 ± 0.59	29.63 ± 14.52	<0.03
(mBq g^{-1})	Range	0.002–0.66	0.002–1.05	0.21–1.39	13.36–41.27	<0.03

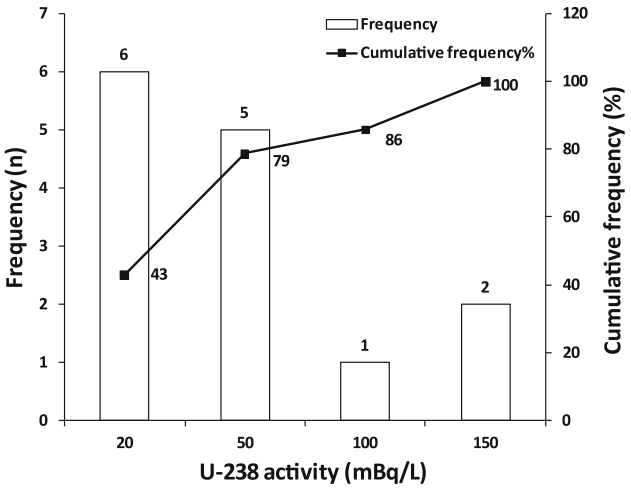

Fig. 3 Frequency distribution of uranium activity concentrations in drinking waters

Tarapur after 25 years of commencement of the facility, comparing their data with pre-operational values. This shows the significance of baseline studies and their utility in the years to come, after the commencement of a facility.

Committed effective dose

The average CEDs for the different radionuclides have been tabulated in Table 4. The highest CEDs were obtained for milk. This is due to the higher ingestion rate for milk (100 g day^{-1}) compared to the other food matrices analysed, as given by IAEA [19]. The average ingestion dose received by ^{238}U and ^{232}Th in milk, eggs, fruits and fish make up for a total CED of 17.53 μSv year^{-1}, which is higher than the reported annual effective dose of 0.54 μSv year^{-1} due to ingestion by the Asian population [26]. The CEDs due to ^{238}U, ^{232}Th, ^{226}Ra, ^{40}K and ^{137}Cs

Table 3 Range of activity concentrations of radionuclides in food matrices, in mBq g^{-1}, reported from global studies

Location	^{238}U	^{232}Th	^{226}Ra	^{40}K	^{137}Cs	References
Tenerife, Spain	<0.09	–	0.03–0.47	35–380	0.01–0.42	[30]
Turkey	–	ND–10.54	15.96–52.80	491.62–2,324.51	–	[34]
Tanzania	5.02–13.23	3.82–4.08	–	24.67–48.79	5.57	[31]
Jos Plateau, Nigeria	–	BDL–89.8	BDL–83.5	BDL–684.5	–	[23]
Nigeria	1.47–39.5	3.5–10.5	–	9.9–298	ND	[24]
Lebanon	–	–	–	31.1–120.9	0.09–0.1	[29]
Tehran, Iran	0.6–15.6	–	6.0–1,153.3	–	–	[35]
Cameroon	–	1.50	2.30	140.40	–	[32]
Lebanon	–	–	–	6.9–868	0.04–2.5	[36]
Bombay, India	–	0.02–1.26	0.01–1.16	45.9–649	–	[9]
Punjab, India	0.38–4.6	–	–	–	–	[10]
Tamil Nadu, India	–	0.09–4.59	0.02–0.23	36–380.6	–	[37]
Tarapur	–	–	0.003–0.8	–	0.03–7.1	[38]
India	0.017–77	–	–	–	–	[6]
Odisha, India	0.3–32.0	0.3–2.0	0.4–28.2	14.3–956.9	0.2–5.4	[12]
Vizag, India	0.002–10.6	0.002–2.8	0.1–7.2	3–110.8	0.03–3	Present study

ND not detected

Table 4 The average CEDs for different radionuclides in various food matrices

Food matrix	CED (μSv year^{-1})				
	^{238}U	^{232}Th	^{226}Ra	^{40}K	^{137}Cs
Milk	4.23 ± 3.72	9.34 ± 9.99	18.14 ± 6.85	1.99 ± 2.61	0.12 ± 0.21
Egg	0.28 ± 0.33	0.19 ± 0.11	1.19 ± 0.73	0.33 ± 0.12	0.03 ± 0.03
Fruit	1.74 ± 1.19	1.34 ± 1.29	10.34 ± 2.76	1.98 ± 2.53	0.03 ± 0.03
Fish	0.04 ± 0.07	0.36 ± 0.6	0.97 ± 0.72	0.8 ± 0.39	<0.001
Water	4.08 ± 4	–	–	–	–

were 10.37, 11.24, 30.65, 5.10 and 0.18 μSv/y considering milk, eggs, fruits, fish and water intake. These values are comparable with the CED values reported for Jaduguda [11] and Tamil Nadu [36] region. A total CED of 53.46 μSv year^{-1} was obtained considering all radionuclides in milk, eggs, fruit and fish. This is lower than that of 180.08 μSv year^{-1} in Pakistan [27], 109.83 μSv year^{-1} in Korea [28], 186 μSv year^{-1} in Lebanon [29], 362 μSv year^{-1} in Tenerife, Spain [30], 360 μSv year^{-1} in Tanzania [31], 476 μSv/y in Jos Plateau, Nigeria [23], 700 μSv year^{-1} in Cameroon [32]. This is because of low intake of the food matrices considered in this study. The large variation in CEDs is mainly due to the geochemical and crustal characteristics of the environment and the dietary and lifestyle choices of the local population.

Contribution of the individual radionuclides to the CEDs in different food matrices are represented in Fig. 4. The ^{238}U, ^{232}Th, ^{226}Ra, ^{40}K and ^{137}Cs contributions ranged from 2.03 ± 3.42 to 13.86 ± 16.44, 8.7 ± 8.35 to 27.62 ± 29.56, 44.62 ± 33.18 to 66.99 ± 17.9, 5.87 ± 7.73 to 36.91 ± 18.09 and 0.07 ± 0.001 to 1.31 ± 1.73 %, respectively. The high degree of associated uncertainty is due to the high variation of radionuclide activity concentrations in a given matrix considering the number of samples of that matrix analysed during this study. This high degree of variation in radionuclide activity concentration is typical for environmental samples and may occur due to preferential uptake and accumulative properties of a radionuclide in a matrix which again depends on the soil characteristics of the region. It can be observed that the highest dose is obtained from ^{226}Ra in all food matrices. Although the concentration of ^{226}Ra is smaller in the matrices than ^{40}K, its contribution is higher due to the higher value of DCF for adults (2.8 × 10^{-7} Sv Bq^{-1}) [18] as compared to the other radionuclides.

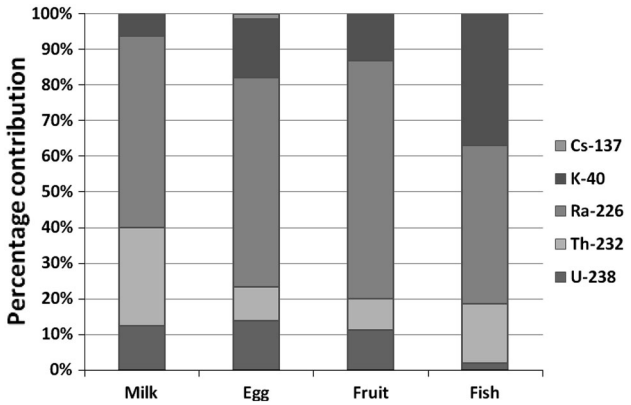

Fig. 4 Contribution of radionuclides to the ingestion dose from food

Contribution of the radionuclides to the average CED combining all the food matrices analysed is shown in Fig. 5. It can be observed from the figures that that ^{226}Ra is the major contributor, making up 56 and 60.3 % for the non-vegetarian and vegetarian diets of the dose considering the food matrices analysed. This is due to its higher DCF value. The smallest contribution is due to ^{137}Cs. This is due to its lower concentration in the food matrices, apart from its small DCF value (1.3 × 10^{-8} Sv Bq^{-1}) [18]. It can also be observed that the contribution of ^{226}Ra, ^{238}U and ^{232}Th increase from non-vegetarian to vegetarian food, whereas contribution of ^{40}K and ^{137}Cs reduces. This is because of the variation of their corresponding concentrations in the food matrices and their variable intake rates.

Conclusion

Activity concentrations of ^{238}U, ^{232}Th, ^{226}Ra, ^{40}K and ^{137}Cs were measured in various food matrices and the

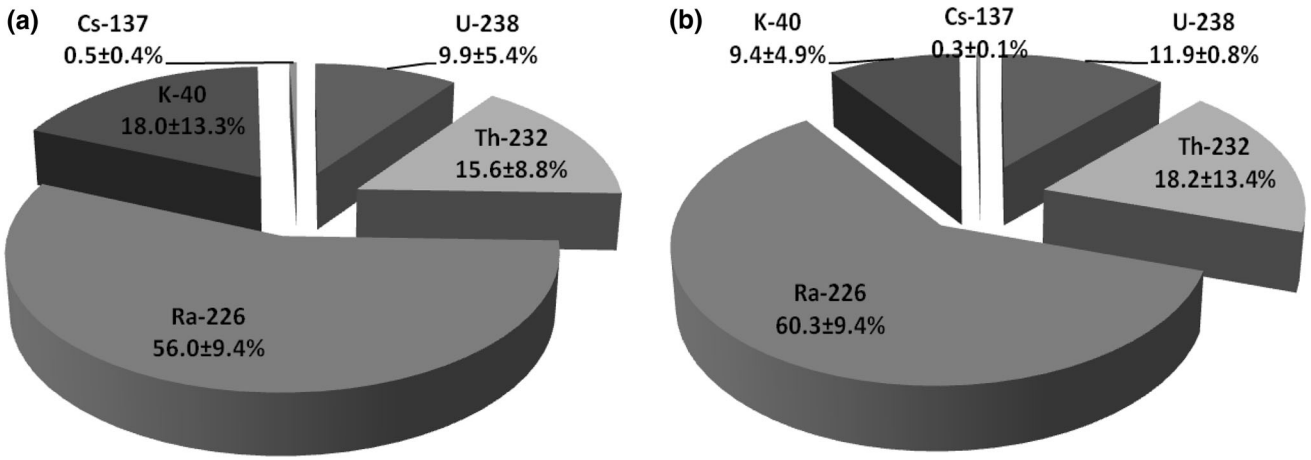

Fig. 5 Contribution of radionuclides to the ingestion dose from (**a**) non-vegetarian: milk, egg, fruit and fish and (**b**) vegetarian: milk and fruit items

corresponding ingestion dose was evaluated. The activity concentrations of the radionuclides ranged from 0.002 to 10.6, 0.002 to 2.8, 0.1 to 7.2, 3 to 110.8, 0.03 to 3 mBq g^{-1} for ^{238}U, ^{232}Th, ^{226}Ra, ^{40}K and ^{137}Cs considering milk, eggs, fruit and fish. The U concentrations measured in drinking water samples were all below the guideline value of 15 ppb set by WHO. The activity concentrations were comparable with reported values in studies carried out globally. Highest ingestion doses were obtained from milk, due to its higher intake rate. The total CED of 53.46 μSv $year^{-1}$ was obtained for milk, fruit, fish and eggs. The highest contributor to the average ingestion dose was ^{226}Ra for the different food matrices analysed due to its high value of dose conversion factor. The contribution of ^{226}Ra, ^{238}U and ^{232}Th increase from non-vegetarian to vegetarian food, whereas contribution of ^{40}K and ^{137}Cs reduces.

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