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The use of total diet study for determination of natural radionuclides in foods of a high background radiation area

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ABSTRACT

The activity concentrations of ^{40}K, ^{210}Pb, ^{210}Po, ^{226}Ra, ^{228}Ra, ^{228}Th, ^{230}Th, ^{232}Th, ^{234}U, and ^{238}U were determined in 82 food samples, grouped into 20 food groups according to the Brazilian Total Diet, which reflects the dietary habits of a population, for the rural and urban areas of Poços de Caldas city, a High Background Radiation Area. The highest activity concentration found in the food samples was due to ^{40}K being present in all types of food. Among the other radionuclides, high activity concentrations were found for ^{210}Pb in beans and salt, ^{210}Po in fish, ^{226}Ra and ^{228}Ra in nuts and seeds. The main food groups that contributed most to the effective dose, in urban and rural regions, were beans and beverages. The effective doses, due to the ingestion of the analysed food groups, were of 0.44 and 0.60 mSv y^{-1} and the lifetime cancer risks were 1.6 × 10^{-3} and 2.3 × 10^{-3} for the urban and rural Poços de Caldas population, respectively.

1. Introduction

Considerable attention has been given worldwide to the study of high background radiation areas (HBRAs) (Aliyu and Ramli, 2015; Veerasamy et al., 2021; Hosoda et al., 2021). Some of these areas are found in China, India, Iran, United States, Canada, and Brazil (Obodovskiy, 2019). A HBRA is defined as "an area or a complex of dwellings where the sum of exposures from cosmic radiation and natural radioactivity of soil, indoor and outdoor air, water and food intake result in an annual effective dose to the public above the defined level of the global average of 2.4 mSv y^{-1} (UNSCEAR, 2000). HBRAs have been classified into four levels: low (<5 mSv y^{-1}); medium (5–20 mSv y^{-1}), high (20–50 mSv y^{-1}); and very high (>50 mSv y^{-1}) (Hendry et al., 2009). Some of these areas have been under study for many years in order to determine the risks and effects of chronic low dose long term exposure for natural radiation (Sohrabi, 2013; Aliyu and Ramli, 2015; Sun and Carr, 2005; Punniyakotti and Ponnusamy, 2018).

In Brazil, the Poços de Caldas Plateau, located in the Southeast region, has a peculiar geological formation characterized by a huge alkaline intrusion where uranium and thorium anomalies can be found, and is recognized as a HBRA. These anomalies are presented in Fig. 1 and it can be seen that most anomaly areas are localized in rural regions (Lauria et al., 2012; Bossew et al., 2015). The arithmetic dose value, for external gamma exposure in the Poços de Caldas urban region is 0.98 mSv y^{-1}, and in the rural region, 1.09 mSv y^{-1}, nevertheless, in one of the anomaly points the external gamma radiation reach dose values as high as 95 mSv y^{-1} (Projeto Planalto de Poços de Caldas, 2009).

Besides the external exposure, food ingestion is also a concern for those living in a HBRA. In addition to food contamination by toxins and toxic elements, the amount of radionuclides present in all foods, incorporated into the food chain from soil, water, and air, must also be taken into consideration to ensure food safety requirements (Brasil, 2013).

The natural radionuclide concentrations in food and water can contribute significantly to the effective internal dose after ingestion and can vary according to several factors, such as local geology, climate and agricultural practices (Shanthi et al., 2010). Therefore, it is important to determine the levels of natural radionuclides from the uranium and thorium series in food.

The World Health Organization (WHO) recommends the Total Diet Study (TDS) as the most suitable method to estimate the dietary intakes

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of contaminants and nutrients for a country or a large population area (WHO, 2002). The TDS is an approach to estimate and monitor dietary exposures that can be present in food at levels that can adversely affect the health of consumers (IBGE, 2011). Although recommended by the WHO, only a few studies in Brazil have used the TDS approach for dietary surveys (Avegliano et al., 2015; Rosa et al., 2015; Ambrogi et al., 2016).

In this study a TDS based on the evaluation of a food list was conducted to evaluate the dose resulting from ingestion of natural series radionuclides from local and marketed food consumption. The food list was composed of 82 types of food divided into 20 groups, reflecting the dietary habits of the Brazilian population from the South-eastern region where the HBRA of Poços de Caldas Plateau is located. Additionally, the lifetime cancer risk due to the ingestion of these radionuclides was estimated.

2. Experimental

The food consumption data and information were obtained from the national Household Budget Survey (HBS) conducted by the Brazilian Institute of Geography and Statistics in 2008–2009 (IBGE, 2011). The food list was based on the personal consumption, over a 24-h period, of food consumed in and out of the household. A total of 14,078 households were surveyed in two non-consecutive days. From them, 3064 were randomly selected as the sub-sample for the analysis of personal food consumption to obtain data on 7302 inhabitants with an age of 10 or more years, in the urban and rural areas of the Brazilian Southeast region (IBGE, 2017; Avegliano et al., 2015).

2.1. *Food group preparation*

The foods were grouped by their similarities and nutritional composition, resulting in a list composed of 20 groups presented in Table 1. Each food group was prepared table-ready, which means that, if necessary, the food was prepared following the normal preparation habits of the studied population. The preparation includes discarding non-edible portions (bones, fat, fruit seeds, etc.) and cooking when necessary. No seasonings, oils or other condiments were added in the preparation. The foods that compose the diet of the urban region were purchased in the retail shops and markets of the city of Poços de Caldas. The food of the rural region diet, whenever possible, was purchased on small farms and ranches, such as eggs and vegetables. After the table-ready preparation, items belonging to each food group were mixed in equal amounts to compose the sample. The types of foods that make up each group and the total consumption, in g day^{-1}, are shown in Table 1.

Table 1
Consumption data of ready-to-consume food groups.

Food Group	Food Items	Total Consumption (g. day^{-1})
Cereals	Polished rice; corn; whole rice	190.9
Beans	Beans; green beans	223.6
Vegetables	Potato; cauliflower; tomato; lettuce; cassava; pumpkin; cabbage; carrot; chayote; sweet potato; French fries; cucumber	78.51
Fruits	Orange; banana; apple; papaya; tangerine; water melon; mango, fruit salad; acai berry, grape; pineapple	81.89
Nuts and seeds	Nuts and seeds	0.2
Flours, pasta and bakery products	French loaf; pasta-based mixtures; wheat flour; instantaneous pasta; pasta; stuffing; breakfast cereal; cassava flour; whole wheat bread	108.27
Cakes and biscuits	Cakes; cookie; salted biscuit; stuffed cookies	29.89
Cattle meat	Rump	67.1
Pork	Slender leg	9.7
Poultry	Chicken	33.0
Fish	Sardine; codfish; sardine in oil	14.9
Industrialized meat and offal	Sausage; cooked sausage; mortadella; ham; bovine liver	14.29
Eggs	Chicken egg	9.90
Dairy products	Pasteurized cow whole milk; natural yogurt; white cheese; fruit smoothie; low fat milk	76.0
Sweets	Chocolate; powder chocolate; pudding; crystallised fruit sweet; ice cream	23.8
Oils and fats	Soil oil	7.60
Beverages	Alcoholic beverages; beer; wine; fruit juice; coke soft drink; milk beverages; soy beverages; coffee; tea	563.71
Industrialized products	Pizzas; snacks; appetizer; sandwiches; soups; tomato sauce; mixture	80.1
Salt	Salt	4.6
Water	Drinking tap water	2000.0
Total Consumption		3617.96

Approximately 500 g of each group were dried at 75 °C till constant weight and pulverized. Approximately 50 g were sealed in acrylic pots and used for ^{40}K determination; 20 g for ^{210}Po determination and approximately 100 g was burnt in order to obtain the ash at 450 °C for 24 h in a muffle furnace. The ash samples were dissolved in nitric and perchloric concentrate acids. The final solution was evaporated and the salts dissolved with 8 M nitric acid and then divided into two parts, one

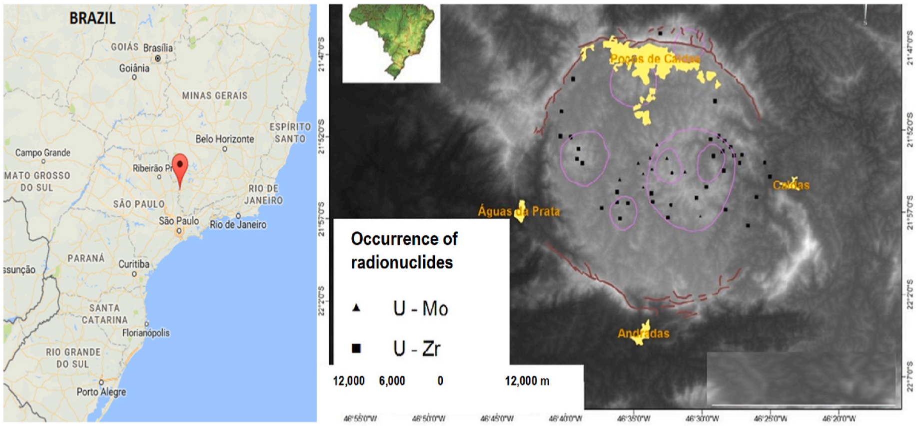

Fig. 1. Location of the city of Poços de Caldas and the Plateau map showing urban areas in yellow and the anomaly points of radionuclide occurrences with black dots. The pink circles refer to volcanic faults, probably associated with radioactive anomalies formation. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

for the determination of ^{210}Pb, ^{226}Ra and ^{228}Ra and the other, for ^{228}Th, ^{230}Th, ^{232}Th, ^{234}U, and ^{238}U determination.

2.2. Radiochemical and radiometric analysis

Potassium-40 activity concentration was determined by measuring its gamma ray transition at 1460.8 keV using a high purity germanium (HPGe) detector, model GX4519, from Canberra Industries. The detector resolution is 1.9 keV for the transition of ^{60}Co at 1332 keV and the relative efficiency, 45%.

For the determination of ^{210}Po, ^{209}Po tracer was added to the sample prior to the dissolution with nitric acid and spontaneously deposited on silver discs. The activity concentration was determined by alpha spectrometry measuring the alpha particle energies at 4.88 MeV from ^{209}Po and 5.30 MeV from ^{210}Po using an Alpha Analyst spectrometer for 150,000 s.

The determination of ^{226}Ra, ^{228}Ra and ^{210}Pb, was carried out in one part of the solution obtained by the ash dissolution as cited above. The separation and measurement followed the procedure described in Knupp et al. (2014). Briefly, solutions of stable Pb^{2+} (20 mg mL^{-1}) and Ba^{2+} (20 mg mL^{-1}) were used as carriers. After a series of dissolutions and precipitations, Ra-isotopes were co-precipitated as barium and radium sulphate, $Ba(Ra)SO_4$, and ^{210}Pb as lead dichromate, $PbCr_2O_7$. The samples were stored for 30 days to await the secular equilibrium between ^{226}Ra, ^{228}Ra and ^{210}Pb with their short half-lives' descendants ^{222}Rn, ^{228}Ac, and ^{210}Bi, respectively. The chemical recovery was obtained by gravimetry and the activity concentrations were quantified using an Ultra Low Level Alpha and Beta total counting with a gaseous flow proportional detector model S5-XLB Tennelec (Canberra Industries) for 120 min.

For the determination of U and Th, the tracers ^{229}Th and ^{232}U were added and the separation followed the procedure described in Taddei et al. (2013) and Rosa et al. (2015). Described briefly, Th separation was carried out using Dowex 1 × 2 anionic exchange resin and U separation using a chromatography column UTEVA. Uranium and thorium were electrodeposited on polished silver plates using an electrical current of 1.0 A to thorium and 1.2 A to uranium for 60 min and analysed in an Alpha Analyst spectrometer with 12 PIPS (Passivated Implanted Planar Silicon) detectors from Canberra Industries for 200,000 s. The alpha particles energies of 5.41 MeV for ^{228}Th, 4.90 MeV for ^{229}Th, 4.67 MeV for ^{230}Th, and 4.01 MeV for ^{232}Th were used for the quantification of thorium isotopes and the alpha particles energies of 4.31 MeV for ^{232}U, 4.74 MeV for ^{234}U, and 4.19 MeV for ^{238}U were used to quantify the uranium isotopes.

2.3. Radioanalytical control

The Reference Material IAEA- Soil 327 (IAEA – International Atomic Energy Agency, 2001) was analysed for quality control of the results (Table 2). Reliable accuracy was obtained in the determination of the natural radioisotopes. Only ^{40}K and ^{210}Pb presented a relative error higher than 10%.

Table 2
Results of ^{226}Ra ^{228}Ra, ^{210}Pb, U and Th radioisotopes in IAEA-Soil-327.

Radionuclide	Activity ± Uncertainty (Bq. kg^{-1})	Certified Value (Bq. kg^{-1})	95% Confidence Interval	Relative Error (%)
^{40}K	688 ± 17	621	612–630	10.8
^{210}Pb	49.0 ± 7.0	58.8	53.9–63.7	16.7
^{210}Po[a]	59.7 ± 3.4	58.8	53.9–63.7	1.5
^{226}Ra	36.5 ± 1.3	34.1	32.7–35.5	7.0
^{228}Ra	35.0 ± 4.0	38.7	37.8–39.6	9.6
^{228}Th	37.0 ± 1.7	38.2	37.2–39.2	3.1
^{230}Th	33.3 ± 1.5	34.1	32.4–35.8	2.3
^{232}Th	37.6 ± 1.7	38.7	37.2–40.2	2.8
^{234}U	33.4 ± 1.7	31.9	30.4–33.4	4.7
^{238}U	33.2 ± 1.7	32.9	31.4–34.2	0.9

[a] Value of ^{210}Pb considering radioactive equilibrium.

Hierarchical Cluster Analyses and Principal Component Analysis were used for data interpretation using Statistica 7 software. These analyses assemble objects based on their similarities, sorting cases into groups or clusters, resulting in a strong association among members of the same cluster and a weak association among members of different clusters, as well as highlighting the variables that most contribute to the observed variance.

2.4. Statistical analysis

Hierarchical Cluster Analyses and Principal Component Analysis were used for data interpretation using Statistica 7 software. These analyses assemble objects based on their similarities, sorting cases into groups or clusters, resulting in a strong association among members of the same cluster and a weak association among members of different clusters, as well as, highlight the variables that most contribute for the observed variance.

3. Results and discussion

3.1. Activity concentration of natural radionuclides in the food groups

The activity concentration of ^{238}U and ^{232}Th series radionuclides and ^{40}K obtained in the diets of both Poços de Caldas populations are presented in Tables 3–5.

Uranium intake is considered to mainly occur by ingestion of food and water, with typical ^{238}U activity concentration in the order of tens and rarely hundreds of mBq kg^{-1} (Garcia et al., 2006; Carvalho and Oliveira, 2010; Desideri et al., 2019).

Uranium-238 activity concentration measured in the food group samples varied from <6 to 91 mBq kg^{-1} and <6–110 mBq kg^{-1} in the urban and rural areas, respectively. Several groups presented ^{238}U activities lower than the detection limit and the highest values were detected in cake and biscuits groups. For comparison, Kam et al. (2016) reported the lowest levels of ^{238}U in leguminous plants or vegetables, and the highest activity concentration in wheat products with a value of 680 mBq kg^{-1} in foods consumed in Turkey.

Most of the food groups presented an activity concentration of ^{234}U very similar to that observed for ^{238}U. Similar results can also be observed in the determination of uranium isotopes in different types of food from vegetal or animal origin (Štrok and Smodiš, 2011a; Pearson et al., 2016, Trdin and Benedik, 2017). This fact indicates that disequilibrium should not be expected for these isotopes in the food chain and in processed food. The isotope ^{230}Th, on the other hand, presented values below the detection limit in most of the urban group samples and the highest activity concentration in the cakes and biscuits group.

Most of the food groups possess ^{232}Th values below 10 mBq kg^{-1} (Table 4). Similar behaviour is also observed between the isotopes ^{232}Th and ^{230}Th although belonging to different natural decay series. Likewise, for ^{238}U, high concentrations for this nuclide were found for the groups of cakes and biscuits, fish, and the group of nuts and seeds.

The difference in the activity concentration between the U and Th decay series radionuclides in food products depends on their mobility in soils and their transfer to plants, which in turn depends on the type of soil and plant, climate, relief, vegetation season, and the radionuclide content in soil, which in HBRA's is higher than average (Shtangeeva, 2010).

In the present samples, ^{226}Ra and ^{228}Ra isotopes presented higher concentrations than their parent series, ^{238}U and ^{232}Th, respectively, in all food groups and in both regions. In the present study the values obtained for ^{226}Ra varied from <20 mBq kg^{-1} (beverage and water groups) to 2300 mBq kg^{-1} (nuts and seeds group) for the urban region and from <20 mBq kg^{-1} (water) to 2600 mBq kg^{-1} (nuts and seeds

Table 3

Activity concentration of ^{238}U series radionuclides determined in the food groups.

Foods Groups	^{238}U (mBq kg^{-1})		^{234}U (mBq kg^{-1})		^{230}Th (mBq kg^{-1})		^{226}Ra (mBq kg^{-1})		^{210}Pb (mBq kg^{-1})		^{210}Po (mBq kg^{-1})	
	Urban	Rural	Urban	Rural	Urban	Rural	Urban	Rural	Urban	Rural	Urban	Rural
Cereal	<6	<6	<10	<10	<6	<6	30 ± 10	150 ± 5	106 ± 10	96 ± 30	120 ± 30	180 ± 20
Beans	<6	11 ± 2	<10	20 ± 10	<6	<6	270 ± 70	260 ± 70	1200 ± 300	1400 ± 200	110 ± 30	150 ± 20
Vegetables	<6	<6	<10	<10	<6	<6	340 ± 70	1600 ± 20	70 ± 20	70 ± 30	130 ± 30	160 ± 20
Fruits	11 ± 2	11 ± 2	11 ± 2	<10	<6	11 ± 2	90 ± 40	210 ± 60	170 ± 60	230 ± 70	130 ± 50	160 ± 20
Nuts and Seeds	74 ± 5	110 ± 20	120 ± 10	190 ± 20	30 ± 10	74 ± 5	2300 ± 200	2600 ± 200	230 ± 60	320 ± 80	230 ± 30	470 ± 20
Flours, Pasta and Bakery Products	13 ± 2	11 ± 2	22 ± 2	<10	10 ± 4	13 ± 2	180 ± 60	320 ± 70	230 ± 80	260 ± 70	200 ± 30	210 ± 20
Cakes and Biscuits	91 ± 6	20 ± 2	80 ± 10	20 ± 10	60 ± 10	91 ± 6	340 ± 70	330 ± 60	30 ± 10	30 ± 10	150 ± 30	180 ± 20
Cattle Meat (beef)	<6	<6	10 ± 2	<10	<6	<6	80 ± 40	90 ± 40	200 ± 70	240 ± 70	140 ± 30	120 ± 30
Pork	6 ± 1	6 ± 1	10 ± 1	<10	<6	6 ± 1	70 ± 30	80 ± 30	230 ± 80	330 ± 70	70 ± 20	170 ± 20
Poultry	26 ± 3	6 ± 1	30 ± 3	<10	<6	26 ± 3	30 ± 10	40 ± 10	<20	120 ± 30	100 ± 30	200 ± 20
Fish	40 ± 3	64 ± 5	50 ± 4	70 ± 10	30 ± 5	40 ± 3	50 ± 20	230 ± 60	600 ± 100	900 ± 200	5050 ± 280	7130 ± 260
Industrialized Meat and Offal	10 ± 2	9 ± 2	20 ± 2	<10	<6	10 ± 2	40 ± 10	50 ± 10	1380 ± 300	1300 ± 300	610 ± 50	630 ± 50
Eggs	<6	<6	<10	<10	<6	<6	150 ± 50	760 ± 110	<20	450 ± 900	120 ± 30	310 ± 30
Dairy Products	<6	<6	10 ± 02	<10	<6	<6	60 ± 20	70 ± 30	30 ± 10	120 ± 40	120 ± 30	180 ± 30
Sweets	20 ± 2	15 ± 2	30 ± 3	20 ± 10	20 ± 10	20 ± 2	80 ± 30	90 ± 30	<20	<20	150 ± 30	180 ± 20
Oils and Fats	40 ± 10	37 ± 9	50 ± 10	50 ± 10	<6	40 ± 10	80 ± 30	80 ± 30	1100 ± 300	1000 ± 300	340 ± 90	420 ± 100
Beverages	7 ± 1	9 ± 2	<10	20 ± 10	<6	7 ± 1	<20	140 ± 50	280 ± 70	250 ± 40	80 ± 20	950 ± 20
Industrialized Products	12 ± 2	24 ± 3	20 ± 2	30 ± 10	<6	12 ± 2	100 ± 40	240 ± 60	500 ± 100	400 ± 40	90 ± 20	100 ± 20
Salt	<6	20 ± 7	60 ± 10	40 ± 10	<6	<6	30 ± 10	50 ± 20	6400 ± 200	5950 ± 170	320 ± 10	290 ± 80
Water	<6	<6	<10	<10	<6	<6	<20	<20	<20	<20	<10	<10

group) for the rural region. Generally, the ^{226}Ra activity concentrations were higher in rural than in urban food groups with the higher differences observed in cereals, vegetables, fruits, fish and egg groups. Similar results were observed for ^{228}Ra, for which higher activity concentrations were observed for most of the food groups from rural compared to urban regions. The higher differences in the activity concentrations were found in almost the same groups as ^{226}Ra.

Thorium-228 shows the same behaviour as its predecessor (^{228}Ra), with higher activity concentrations in rural than in urban food groups. The only notable exception is for the nuts and seeds group, presenting much higher activity concentrations in urban samples.

Activity concentrations in *Bertholletia excelsa* Brazilian nut were found in the order of tens of Bq kg^{-1} for ^{226}Ra and in the order of hundreds of Bq kg^{-1} for ^{228}Ra, showing the ability of seeds in accumulating this element (Armelin et al., 2016). Besides the Ra-isotopes, high activity concentrations in the nuts and seeds group were also found for the radionuclides ^{228}Th, ^{234}U and ^{238}U. Overall, the activity concentrations in Brazilian nut groups are up to a thousand times higher than those in other foods (Mazokopakis and Liontiris, 2017). Higher activity concentrations of the radionuclides from the ^{232}Th series than for the ones of the ^{238}U series, were also observed in different species of Brazilian beans (Silva et al., 2021), and in various types of food (Lauria et al., 2009; Ross et al., 2013).

The isotope ^{210}Pb is the grandfather of ^{210}Po. These radionuclides are the ones with the highest radiotoxicity in the ^{238}U decay chain, being of great concern from the radiation protection viewpoint (Štrok and Smodiš 2011b). Comparing the different food groups, high activity concentrations of ^{210}Pb were found in beans, cattle meat, pork, fish, industrialized meat and offal, oils and fats, industrialized products, and salt for both rural and urban region samples. The well-known capacity for ^{210}Po to be accumulated by most marine organisms was also observed in this study (Hansen et al., 2022; Ahmed et al., 2021). The activity concentrations of this radionuclide in the fish group were 5100 mBq kg^{-1} and 7100 mBq kg^{-1} in the urban and rural areas, respectively. The same tendency was observed in an Italian daily diet study showing a decreasing trend from seafood to meat and milk products (Meli et al., 2014).

The global average for the activity concentration found in food groups, reported by the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR, 2000), due to the intake of ^{238}U and ^{232}Th nuclides decay series is shown in Table 6. The activity concentrations observed in this study in fruits and fish products are slightly higher than these values while grains and vegetables are lower. For U isotopes, it can be observed that groups composed of fruits, nuts and seeds, and fish are slightly higher than the global average. Radium isotopes, ^{226}Ra and ^{228}Ra, were higher in milk products, grain products (beans), meat, vegetables, fruits of both regions and fish products of the rural region. Higher values than those presented by UNSCEAR – United Nations Scientific Committee on the Effects of Atomic Radiation, Ionizing (2000) for ^{210}Pb and ^{210}Po were observed in the groups of beans, vegetables, fruits, meat (cattle and pork), milk, and fish.

Potassium-40 is a radionuclide that occurs naturally in a fixed ratio to the stable potassium. It is an essential element for all living beings and humans, uniformly distributed in the body following food intake, and its concentration is controlled by homeostatic and metabolic processes (Madruga et al., 2020). In the Poços de Caldas food groups from the urban region, the ^{40}K activity concentration varied from 6.6 Bq kg^{-1}, in water, to 155 Bq kg^{-1}, in the pork meat group. The food groups from the rural region presented generally higher ^{40}K levels than those from the urban regions, mainly for nuts and seeds, fish and industrialized products.

3.2. Determination of effective dose by food intake

The effective doses of radionuclides from U and Th decay series from food intake by urban and rural adult population groups were calculated

Table 4
Activity concentration of ^{232}Th series radionuclides determined in the food groups.

Foods Groups	^{232}Th (mBq kg^{-1})		^{228}Ra (mBq kg^{-1})		^{228}Th (mBq kg^{-1})	
	Urban	Rural	Urban	Rural	Urban	Rural
Cereal	<10	<10	26 ± 3	250 ± 60	120 ± 10	190 ± 10
Beans	<10	30 ± 10	<20	460 ± 140	350 ± 20	590 ± 20
Vegetables	<10	10 ± 10	550 ± 20	1530 ± 340	370 ± 20	680 ± 40
Fruits	10 ± 3	10 ± 10	30 ± 10	120 ± 50	140 ± 10	210 ± 10
Nuts and Seeds	30 ± 10	<10	3880 ± 990	4180 ± 560	4050 ± 150	870 ± 120
Flours, Pasta and Bakery	10 ± 4	<10	260 ± 90	300 ± 130	150 ± 20	390 ± 20
Cakes and Biscuits	60 ± 10	<10	<20	120 ± 30	300 ± 20	290 ± 40
Cattle Meat (beef)	<10	20 ± 10	1010 ± 270	1100 ± 140	100 ± 10	400 ± 60
Pork	<10	10 ± 10	70 ± 20	150 ± 30	40 ± 10	360 ± 40
Poultry	<10	<10	540 ± 90	890 ± 120	30 ± 10	190 ± 10
Fish	30 ± 4	20 ± 10	180 ± 70	270 ± 90	120 ± 10	300 ± 40
Industrialized Meat and Offal	<10	10 ± 10	60 ± 40	80 ± 20	250 ± 10	370 ± 50
Eggs	<10	<10	210 ± 90	1330 ± 420	80 ± 10	500 ± 80
Dairy Products	<10	<10	190 ± 70	150 ± 40	50 ± 01	870 ± 170
Sweets	<10	10 ± 1	320 ± 90	420 ± 90	70 ± 10	770 ± 20
Oils and Fats	<10	20 ± 1	410 ± 20	380 ± 30	310 ± 50	450 ± 70
Beverages	<10	<10	130 ± 50	150 ± 30	30 ± 10	30 ± 10
Industrialized Products	<10	<10	200 ± 70	380 ± 100	70 ± 10	570 ± 20
Salt	<10	<10	1300 ± 40	1000 ± 250	260 ± 40	210 ± 40
Water	<10	<10	<20	<20	<10	<10

Table 5
Activity concentration of ^{40}K determined in the food groups.

Foods Groups	^{40}K (Bq kg^{-1})	
	Urban	Rural
Cereal	134 ± 15	180 ± 13
Beans	111 ± 15	257 ± 23
Vegetables	54 ± 13	221 ± 19
Fruits	47 ± 10	168 ± 11
Nuts and Seeds	110 ± 12	319 ± 16
Flours, Pasta and Bakery	16 ± 4	32 ± 4
Cakes and Biscuits	46 ± 9	49 ± 12
Cattle Meat (beef)	135 ± 17	152 ± 19
Pork	155 ± 17	184 ± 12
Poultry	128 ± 20	180 ± 13
Fish	118 ± 15	268 ± 24
Industrialized Meat and Offal	75 ± 14	96 ± 13
Eggs	28 ± 7	41 ± 7
Dairy Products	49 ± 10	52 ± 11
Sweets	133 ± 15	142 ± 11
Oils and Fats	52 ± 3	54 ± 3
Beverages	17 ± 4	21 ± 4
Industrialized Products	142 ± 15	207 ± 18
Salt	96 ± 10	108 ± 10
Water	6.6 ± 0.3	7 ± 1

using Equation (1) (IAEA – International Atomic Energy Agency, 1999). Due to the biological half-life of stable potassium, the effective dose derived from ^{40}K is considered to be kept constant with a value of 0.165 mSv y^{-1} for adults (Fasae and Isinkaye, 2018; UNSCEAR – United Nations Scientific Committee on the Effects of Atomic Radiation, Ionizing, 2000).

$$E = \sum_{j}\sum_{f} C_{fj}.M_{j}.T.\ h_{j} \tag{1}$$

Where: E_A is the committed effective dose (mSv) per group. C_{fj} is the average concentration (Bq kg^{-1}) of a radionuclide j in each group of food. M_{jA} is the mass (kg day^{-1}) of a food item f consumed per day. T is the total time (days) considered. h_{jA} is the ingestion dose coefficient (mSv Bq^{-1}) of a radionuclide j.

The effective dose resulting from the ingestion, by adults, of each determined radionuclide and the annual effective dose are presented in Table 7. Dose coefficients were taken from ICRP – International Commission on Radiological Protection (2012). The dose distributions according to each food group are shown in Figs. 2 and 3. Water dose values were not included in these figures since the activity concentrations for all the analysed radionuclides were below the detection limits. In this study, only tap water was analysed since it is the main source of water for the Poços de Caldas population, even in the rural region.

The highest doses of uranium isotopes (Fig. 2a) came from the consumption of beverages, and cakes and biscuit groups, both with almost the same contribution, while the highest dose contribution of ^{230}Th is due to the ingestion of cakes and biscuits, flour, pasta and bakery products, and industrialized product groups in the Poços de Caldas diet.

The highest dose from uranium decay series radionuclides arises from ^{210}Pb by the ingestion of beverages and beans, from the ingestion of ^{210}Po, due to fish and beverages, and for ^{226}Ra, due to the ingestion of beans and vegetables (Fig. 2b). From the ^{232}Th series, ^{228}Ra is the nuclide that contributes the most to the total dose and the highest ones are observed for the ingestion of beverages, cattle meat, cereals and vegetables (Fig. 2c). Higher doses of ^{228}Th are due to beans, vegetables, and beverages ingestion. ^{232}Th is the nuclide with the lowest contribution to the dose from its own series.

For the rural food groups, the dose contribution from the uranium isotopes (Fig. 3a) presents almost the same values as the urban ones, except in beverages where the ^{234}U dose is almost twice that provided by ^{238}U. The highest contributions of ^{230}Th are due to flour, pasta and bakery products, cattle meat, and vegetables. Considering ^{226}Ra, ^{210}Pb and ^{210}Po (Fig. 3b), the highest doses are due to the ingestion of ^{210}Pb, present in beans and beverages. Polonium-210 contribution is mainly due to beverage and fish consumption, while the main contribution of ^{226}Ra is due to vegetables and beverages. The thorium series radionuclide dose contribution presents, in the rural area, the same behaviour that is observed for the urban region, with the dose of $^{228}\text{Ra} > {}^{228}\text{Th} > {}^{232}\text{Th}$.

Despite the nut and seed group presenting the highest activity concentration for Ra-isotopes and ^{228}Th, its consumption, including *Bertholletia excels* nuts, is low and not enough to contribute with a significant dose for both urban and rural regions. Beans and beverages consumed in both regions contribute to the highest doses for ^{210}Pb, but only for the rural group is ^{210}Po more significant in beverages. The contribution of ^{228}Ra is more pronounced in the rural than in the urban group.

The dendrograms for the food groups analysed for urban and rural regions are shown in Figs. 4 and 5. Putting the cut in 40% of the maximum distance (Dlink/Dmax), both food group samples are grouped in three subgroups. For the urban samples, the first subgroup (4a) is characterized by the dose contribution arising from ^{228}Ra. The second subgroup (4b), by the low dose contributions of all radionuclides analysed, and the third subgroup (4c) by the dose distribution arising from ^{210}Pb. For the rural samples, the first subgroup (5a) is characterized by

Table 6
Mean activity concentration in food according to UNSCEAR – United Nations Scientific Committee on the Effects of Atomic Radiation, Ionizing (2000)

Products	Activity concentration (mBq kg^{-1})							
	^{238}U	^{230}Th	^{226}Ra	^{210}Pb	^{210}Po	^{232}Th	^{228}Ra	^{228}Th
Milk	1	0.5	5	15	15	0.3	5	0.3
Meat	2	2	15	80	60	1	10	1
Grain	20	10	80	50	60	3	60	3
Leafy vegetables	20	20	50	80	100	15	40	15
Roots and fruits		0.5	30	30	40	0.5	20	0.5
Fish	30	10	100	200	2000	10	ND*	100
Drinking water	1	0.1	0.5	10	5	0.05	0.5	0.05

* ND indicates that no published data are available.

Table 7
Radionuclide contributors to the annual effective dose.

Radionuclide	Dose intake diet urban region (mSv y^{-1})	Dose intake diet rural region (mSv y^{-1})
^{238}U	1.98E-04	1.39E-05
^{234}U	3.02E-04	3.62E-04
^{230}Th	3.65E-04	1.71E-03
^{226}Ra	1.61E-02	4.12E-02
^{210}Pb	1.53E-01	1.63E-01
^{210}Po	2.28E-02	7.19E-02
^{232}Th	3.57E-04	1.09E-03
^{228}Ra	8.18E-02	1.43E-01
^{228}Th	5.62E-03	1.30E-02
^{40}K	1.65E-01*	1.65E-01†
Annual Effective Dose (mSv y^{-1})	4.45E-01	6.00E-01

* Dose value recommended by UNSCEAR – United Nations Scientific Committee on the Effects of Atomic Radiation, Ionizing, 2000 for ^{40}K intake.

the dose contributions of ^{228}Ra and ^{230}Th. The second subgroup (5b), the same as for the urban region samples, by the low dose contributions of all analysed radionuclides, and the third subgroup (5c) by the doses from the radionuclides ^{228}Ra, ^{210}Pb and ^{234}U.

The difference in the dose results found for the urban and rural regions are related to the fact that the activity concentrations in the rural region were higher, with few exceptions, resulting in an effective dose 25.83% higher compared to the urban diet. This is probably related to the fact that the greatest points of radioactive anomalies are located in the plateau and in the rural area it is more common for the food consumed to have been produced on the small properties of the local population.

On average, the human being receives about 2.4 mSv of effective dose each year due to the natural sources of radiation, from which 0.29 mSv of the total is due to the ingestion of food and water, with a variation between 0.2 and 0.8 mSv (UNSCEAR, 2000).

The annual effective dose found for the urban and rural areas of Poços de Caldas are in good agreement with other studies from the same region and also with results from other HBRAs reported in the literature. Silva et al. (2021) calculated an effective dose of 0.5 mSv y^{-1} for the ingestion of beans produced in the Poços de Caldas Plateau. The annual effective dose amongst the fishermen population residing in the Kanyakumari coastal area, due only to the ^{210}Po ingestion of seafood, fish, and edibles, reached 2.24 mSv (Rani et al., 2014). A total of 25 foodstuff samples analysed from some rural districts of Cameroon resulted in a total annual effective dose estimated at 0.70 mSv y^{-1} (Abiama et al., 2012). The calculated doses for Poços de Caldas are higher than that reported for the global average, but in agreement with global range reported by UNSCEAR – United Nations Scientific

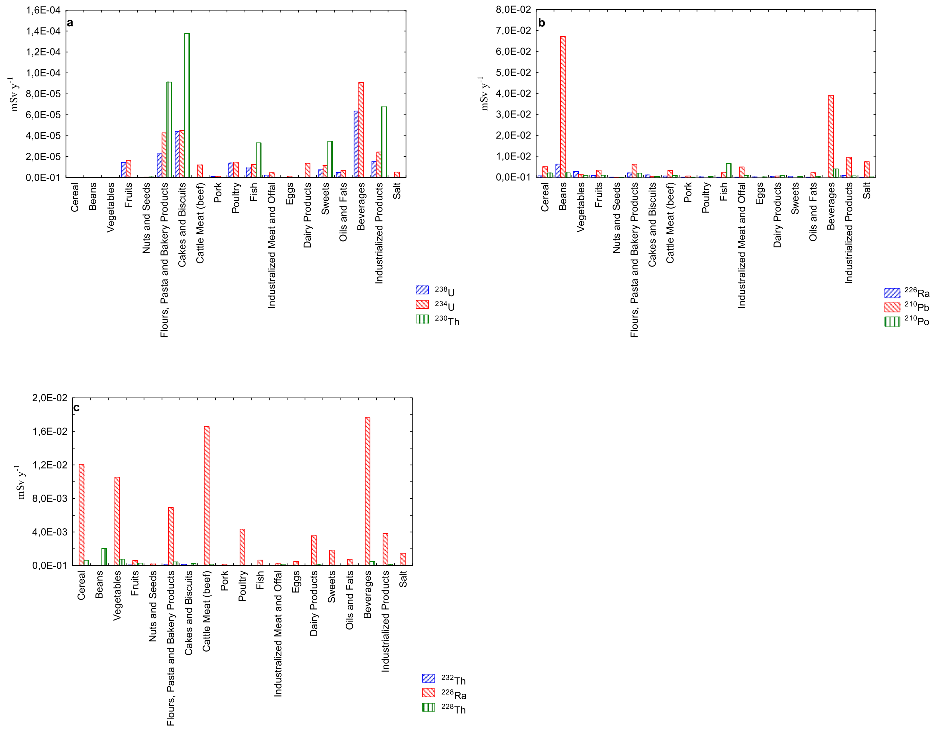

Fig. 2. Effective dose distribution for food groups in the samples from urban regions.

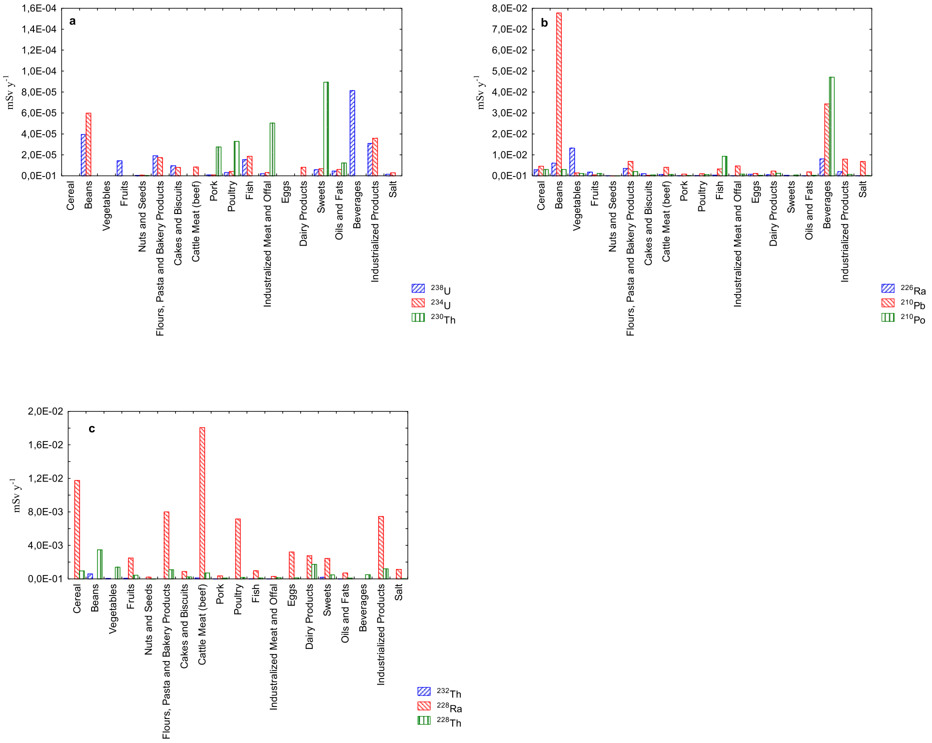

Fig. 3. Effective dose distribution for food groups in the samples from rural regions.

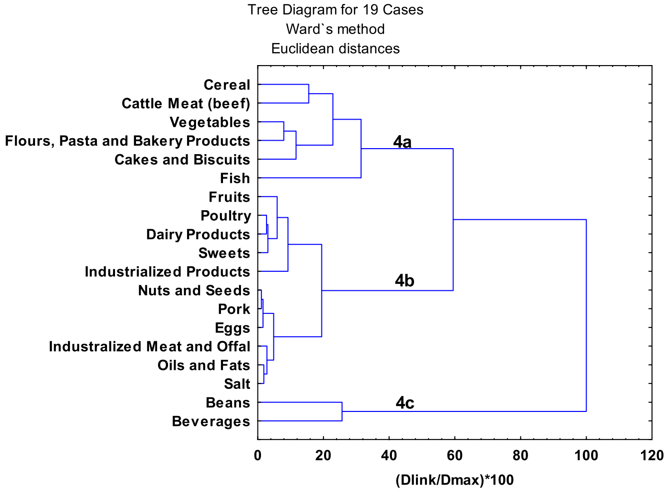

Fig. 4. Dendrogram for the food groups analysed from the urban region, and the three groups formed indicated as 4a, 4b, and 4c.

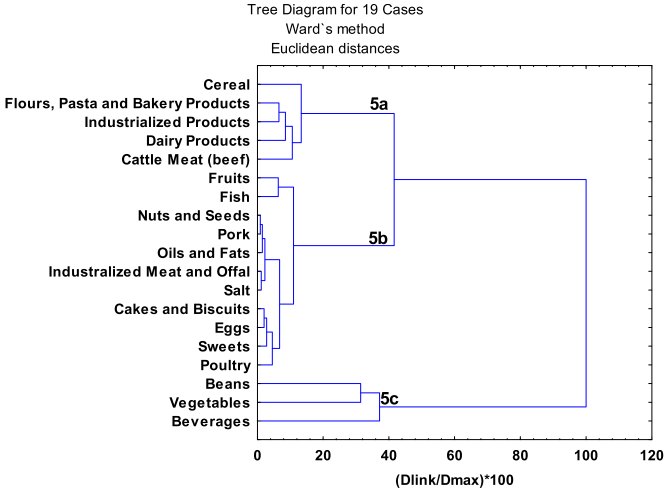

Fig. 5. Dendrogram for the food groups analysed from the rural region, and the three groups formed indicated as 5a, 5b, and 5c.

Committee on the Effects of Atomic Radiation, Ionizing (2000).

3.3. Lifetime cancer risk

The lifetime cancer risk (C_R) index estimates the average total risk for an individual experiencing a radiogenic cancer, whether or not fatal (Giri et al., 2011). According to the guidelines of the United States Environmental Protection Agency (USEPA, 2003), the C_R is negligible if values are lower than 10^{-6} and may need some intervention or remediation for values above 10^{-4}. Based on the annual dose increment of 1 mSv for the general public suggested by ICRP – International Commission on Radiological Protection (1991), the value of the cancer risk factor of 2.5×10^{-3} can be taken as a reference value for the lifetime cancer risk index (Khan and Wesley, 2011). The C_R index is calculated by the sum of the average daily intake of a given radionuclide *i* (ADI_i), in Bq kg^{-1}, the risk coefficient, for mortality, of each radionuclide *i* (RC_i) obtained from data provided by the Health Effect Assessment Summary Tables of United States Environmental Protection Agency (USEPA, 1999) and the life mean duration, considered as 75.2 years for Brazil (Silva et al., 2021), in days, product (equation (2)).

$$C_R = \sum ADI_i \cdot RC_i \cdot 27488 \qquad (2)$$

Considering the constant dose contribution of ^{40}K, a C_R value of 1.6×10^{-3} was obtained for the urban area and a slightly higher value of 2.3×10^{-3} was obtained for the rural area. The main contributors to these values are ^{210}Pb and ^{228}Ra, which contributes 41.6 and 23.9% to the C_R value, respectively, for the urban region. In the rural area, ^{228}Ra is responsible for 31.4% of the C_R and ^{210}Pb contributes 29.3%.

Considering the food types, the main contributions for the C_R comes from beverage and beans in the proportion of 25.3% and 24.1% in the urban area, and 27.0% and 23.8%, in the rural area, respectively.

4. Conclusions

A total diet study was carried out in two regions, urban and rural, of the Poços de Caldas Plateau, considered as a HBRA. In the comparison among the different food groups, higher activity concentrations from the uranium series were found in nuts and seeds for ^{238}U, ^{234}U and ^{226}Ra for both urban and rural areas. For ^{226}Ra higher activity concentrations were found in cakes and biscuits in both areas, and vegetables and eggs in rural areas. Industrialized meat and offal, oil and fats, salt, beans and fish presented high activity concentration for ^{210}Pb in both areas. In the fish and beverage groups, ^{210}Po presented higher activity concentration in samples from the rural area.

For the radionuclides from the thorium series, higher activity concentrations were found in nuts and seeds, salt, and cattle meat for both areas, but overall, they were higher in the rural area. Poultry, eggs and vegetables present high activity concentrations mainly in the rural food samples for ^{228}Ra. For ^{228}Th, nuts and seeds presented high values in the urban samples while the values were higher for vegetables, dairy products and sweets in the rural samples. The activity concentrations of ^{40}K were higher in the rural samples of beans and vegetables.

The higher activity concentrations found in the rural area food groups of local production such as vegetables, beans, beverages, fish, poultry and eggs are indicative of the HBRA contribution to the amount of natural radionuclides present in the food consumed by the Poços de Caldas population, since the local rural production is also sold in the city. Consequently, the effective doses of 0.445 and 0.60 mSv y^{-1} obtained for the urban and rural region, respectively, is relatively higher than the global average (0.29 mSv), but still lower than the 1 mSv y^{-1} recommended dose increment for public individuals. The main food groups responsible for the observed dose in the urban region are beans and beverages, followed by cereals, cattle meat and vegetables, while for the rural region, the main contribution comes from beans, vegetables and beverages, followed by cereals, indicating that the consumption of vegetables and beans from local production should be higher in the rural region.

The cancer risk factors calculated from this data were 1.6×10^{-3} and 2.3×10^{-3} for the urban and rural areas, respectively. These values were below the reference value of 2.5×10^{-3}, although very close to it.

Finally, it should be noted that this study was based on the ingestion of food proportions reported by the IBGE for the Brazilian Southeast region, that does not differentiate between urban and rural areas and this fact may be the main limitation in the statement of the conclusions.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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