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Radioactivity in coffee

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Abstract This research was dedicated to the study of the background levels of ^{210}Po and natural gamma emitters as ^{40}K, ^{214}Pb, ^{214}Bi, ^{228}Ac, ^{212}Pb and ^{212}Bi in coffee powder and in coffee beverage; also the artificial ^{137}Cs was determined. In the coffee powder the mean ^{210}Po activity resulted $7.25 \pm 2.25 \times 10^{-2}$ Bq kg^{-1}. ^{40}K showed a mean activity of 907.4 ± 115.6 Bq kg^{-1}. The mean activity concentration of ^{214}Pb and ^{214}Bi, indicators of ^{226}Ra, given as mean value of the two radionuclides, resulted 10.61 ± 4.02 Bq kg^{-1}. ^{228}Ac, ^{228}Ra indicator, showed a mean activity concentration of 13.73 ± 3.20 Bq kg^{-1}. The mean activity concentration of ^{212}Pb, ^{224}Ra indicator, was 8.28 ± 2.88 Bq kg^{-1}. ^{208}Tl, ^{224}Ra indicator, presented a mean activity concentration of 11.03 ± 4.34 Bq kg^{-1}. In all samples, the artifical ^{137}Cs resulted below the detection limit (2.0 Bq kg^{-1}). The arithmetical mean value of percentage of ^{210}Po extraction in coffee beverage resulted 20.5 ± 6.9. The percentage of transfer of gamma emitters, ^{40}K, ^{214}Pb, ^{214}Bi, ^{228}Ac, ^{212}Pb, ^{208}Tl resulted of 80.0, 33.5, 24.7, 30.0, 35.1 and 53.5 % for ^{40}K, ^{214}Pb, ^{214}Bi, ^{228}Ac, ^{212}Pb and ^{208}Tl respectively.

Keywords Natural radioactivity · ^{137}Cs · Coffee

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Introduction

Coffee, one of the most widely popular non-alcoholic beverage, is a brewed beverage with a strong flavor prepared from the roasted seeds of the coffee plant. The beans are found in coffee cherries, which grow on trees cultivated in over 70 countries, primarily in equatorial Latin America, Southeast Asia, South Asia and Africa. Green (unroasted) coffee is one of the most traded agricultural commodities in the world. Coffee is slightly acidic (5.0–5.1 pH) and can have a stimulating effect on humans due to its caffeine content. It is one of the most-consumed beverages in the world [1].

The annual consumption of coffee varies in different countries of the world. The country has the highest consumption of coffee is Finland with 12 kg per capita, followed by Switzerland 9 kg, Denmark and Sweden 8 kg, Germany 7 kg, France and Italy 6 kg, Spain and the U.S. 5 kg and finally England and Japan with 4 kg.

World Health Organization defined coffee "non nutritive dietary component" so it is not considered a food even though containing certain nutrients, as well as numerous compounds of various type. Coffee may appear to be a simple drink, but in reality it is a highly complex product. It is derived from over 1,500 chemical substances (approximately 850 volatile and 700 soluble). The microroasted coffee, however, provides specific nutrients such as vitamin niacin (essential for many metabolic functions). Coffee also contains potassium (important for the function of blood circulation). Two cups of coffee account for some 10 % of your daily potassium.

Studies on the radiation level and radionuclide concentration in foodstuffs are available in the literature, however, the information on the distribution and enrichment of radionuclides in coffee is very scarce. The aim of this work was to study the natural and artificial radioactivity in coffee powder and in coffee beverage. In fact the presence of radionuclides in these matrixes constitutes the pathway for their migration to human, via food chain.

In our daily lives, we are each exposed to various types of naturally occurring ionizing radiation which is commonly referred to as background radiation. Naturally occurring background radiation comes from a number of source that include terrestrial radiation, cosmic radiation, inhaled radionuclide and internal radionuclide [2].

Any radioactivity present on air or more importantly in the ground and soil may transfer into food grown on it. It happens, however, that some naturally occurring radioactive elements find their way into our body. The most important radionuclide that gives the largest part of the dose to an average person from ingestion are ^{40}K, a primordial radionuclide, and ^{210}Po, a radionuclide of ^{238}U radioactive chain.

^{40}K is a natural radioisotope present in soil and as the element K, an essential plant nutrient, enters in the plant roots via ion channels or specific transporters. The percentage made up by the natural radionuclide ^{40}K is 0.0117 % K is generally abundant in the food [3].

Among the alpha emitters ^{210}Po is estimated to contribute about 7 % of the effective dose equivalent to man from ingested natural internal radiation [4].

This radionuclide and his grandfather ^{210}Pb belong to ^{238}U series. Their presence in the terrestrial environment arises from ^{222}Rn which, once produced, may remain in soil interstitial air spaces, decay within the mineral matrix of soil or be released to the atmosphere. Globally, radon exhalation from soil accounts for about 22 PBq year^{-1} of ^{210}Pb and ^{210}Po [5]. At the present the approximated average atmospheric concentrations of ^{210}Pb and ^{210}Po in northern temperate latitudes are 0.5 and 0.05 mBq m^{-3}, respectively [6]. ^{210}Pb and ^{210}Po return to the earth's surface via both wet and dry deposition. Atmospheric fallout of these decay products result in the contamination of plants and the top layer of soil. Most of the natural radioactivity content in wild leafy plants is ^{210}Po as the result of the direct deposition of ^{222}Rn daughters from atmospheric precipitation and their presence in all terrestrial foodstuffs is inevitable [7, 8]. It is also known that natural levels of ^{210}Pb and ^{210}Po in the environment can locally be increased by anthropogenic activities like phosphate ore processing, coal-fired power stations, coal mining, metal smelting, etc. which produce enhanced levels of ^{210}Pb and ^{210}Po [9].

About 18 % of the average internal dose of the population is due to ingestion of ^{210}Po along with its precursor ^{210}Pb [10]. ^{210}Po, in fact, causes considerable radiation risk even at minimal intake due to its high linear energy transfer (LET). The ^{210}Po toxicity is comparable to ^{239}Pu and about five times greater than ^{226}Ra [11, 12].

It is well known radionuclides are present in the environment either naturally or artificially. ^{137}Cs is an artificial radioelement, released in the past by nuclear weapon testing (1945–1963) and by the Chernobyl accident (1986). ^{137}Cs is mobile in the environment and accumulated in foodstuffs, mainly in animal products [13].

This research was dedicated to the study of the background levels of ^{210}Po and natural gamma emitters as ^{40}K, ^{214}Pb, ^{214}Bi, ^{228}Ac, ^{212}P, ^{212}Bi and the artificial ^{137}Cs in coffee powder and in coffee beverage.

In this work ^{210}Po was determined by alpha spectrometry; gamma spectrometry was used to measure ^{40}K, ^{214}Pb, ^{214}Bi, ^{228}Ac, ^{212}Pb, ^{208}Tl and ^{137}Cs.

Experimental

Samples

Analysis was carried out in the power and in the beverage of 18 Italian different brands of coffee. The powders of coffee, in the vacuum packed, were obtained through normal distribution channels. For some brands, coffee grounds were also analysed.

Coffee beverage (coffee) preparation

Coffee beverage (coffee) preparation can be obtained in several different ways, depending upon how the water is introduced to the coffee grounds. In this work was prepared used a moka pot [14].

The moka pot is also known as the "Italian coffee pot" or the "caffettiera". It consists of two containers which are located between the screw-on filter that contains the coffee powder. Cold water is heated in the container bottom and the generated steam pressure, about one bar (100 kPa, 14.5 psi), forces the boiling water up through coffee powder held in the middle section, separated by a filter mesh from the top section.

Moka pots used from this work made from aluminium with bakelite handles (Fig. 1). The bottom chamber (a) contained 800 ml and basked (b) contained 60 g of coffee power. The beverage obtained by using 800 ml of fresh tap water resulted 660 ml.

Analytical method

The radioisotope determination of the samples was carried out by two different techniques: a) high resolution gamma spectrometry with high purity germanium detector for ^{40}K, ^{214}Pb, ^{214}Bi, ^{228}Ac, ^{212}Pb, ^{208}Tl and ^{137}Cs and b) radiochemical methods for ^{210}Po.

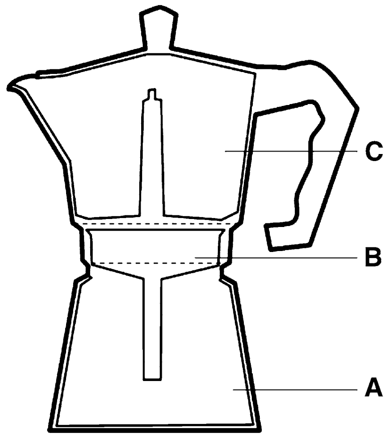

Fig. 1 The bottom chamber (**a**) contains water. When heated, steam pressure pushes the water through a basket containing ground coffee (**b**) into the collecting chamber (**c**)

Gamma spectrometry

It is possible to determine simultaneously many radionuclides by a direct γ-spectrometry of the sample without any specific pre-treatment of this.

The measure of ^{40}K, ^{214}Pb, ^{214}Bi, ^{228}Ac, ^{212}Pb, ^{208}Tl and ^{137}Cs was carried out in all samples of coffee powder and in some samples of coffee grounds. Samples were put and counted in Marinelli beakers for 100,000 s. All the measurements were performed with a HPGe crystal of type "p" in coaxial geometry, with relative efficiency at 1.33 MeV equal to 25 % (measured by placing a point source of ^{60}Co, at 25 cm from the detector); the detectable energy ranges from 0.040 to 2.2 MeV; peak detection efficiencies were calculated automatically through a computer system interfaced with an multichannel analyser. The photo peaks used for ^{228}Ac are 911.0 and 969.0 keV, for ^{214}Pb are 295.0 and 351.9 keV, for ^{214}Bi are 609.3 and 1020.3 keV. The photo peaks used are 238.6 and 593.2 keV for ^{212}Pb and ^{208}Tl respectively. The activity concentration of ^{210}Pb, ^{40}K and ^{137}Cs was found by measuring the activity of their peak at 46.5, 1460.8 and 661.6 keV respectively.

Radiochemical methods

^{210}Po cannot be determined by gamma spectrometry because it is not a gamma emitter, it emits only alpha particles at 5.407 MeV. It is not possible to use the gamma emission of other members of ^{238}U series because a secular equilibrium for all members of the series cannot be assumed. So ^{210}Po was determined through a radiochemical method, which is a destructive technique; it consists in the measurements of the radionuclides after their separation (by extraction chromatography, precipitation, electrodeposition etc.) from the solution coming from the complete dissolution of the sample. The method used in this research to ^{210}Po determination consists in a sample pre-treatment and a polonium source preparation.

Sample pre-treatment

Coffee powder and coffee grounds Fifteen gram of coffee powder and all the sample of coffee grounds were spiked with a know activity of ^{209}Po as a yield tracer and digested using concentrate nitric acid for at least 24 h, hydrogen peroxide was also added to help in oxidizing the organic compounds.

When the solution was clear, the sample was evaporated to dryness; the residue was dissolved in 1 M HCl and the solution was filtered. The dissolution of the powder has been performed several times on the same sample in order to verify the homogeneity of the sample and the repeatability of the test.

Coffee beverage After acidification, the beverage was evaporated and mineralized with concentrate nitric acid and hydrogen peroxide to help in oxidizing the organic compounds. When the solution was clear, the sample was evaporated to near dryness. The residue was dissolved in 1 M HCl and the solution was filtered.

Tap water Fresh, cold water is very important in the preparation of coffee. The water allows the extraction of the substances that characterize the taste of this product. For water, the best temperature of extraction is between 92 and 96 °C; tap water was used for the preparation of coffee. The content of ^{210}Po in tap water was determined after evaporation of 1,000 ml of water (eight samples) and the residue dissolved in 1 M HCl.

Polonium source preparation

Polonium is deposited at 85–90 °C and pH 1.5–2.0 in continuous for 4 h in a silver disk, placed in a syringe and immersed into a 200 mL of 1 M HCl solution coming from the pre-treatment and containing 10 ml of 20 % hydroxylamine hydrochloride and 10 ml of 25 % sodium citrate. No preliminary separation is required and essentially quantitative recoveries are obtained by using standard ^{209}Po tracer [15]. The mean chemical yield resulted 72.9 ± 11.5. The silver disk is then measured by α-spectrometry (^{210}Po alpha particles energy: 5.407 MeV). The measurement is carried out using an alpha spectrometry system with silicon detectors (Canberra, USA) counting the source for 200,000 s. The mean counting efficiency is 31.7 ± 3.1 % and the background is approximately 2×10^{-6} s^{-1} in the energy region of interest.

Table 1 ^{210}Po activity concentration (Bq kg^{-1}), arithmetical mean activity concentration, standard deviation, minimum and maximum values for 18 Italian different brands of coffee powder

Brand	^{210}Po
1	0.0535
2	0.0336
3	0.1110
4	0.0645
5	0.0777
6	0.0495
7	0.0623
8	0.0935
9	0.0548
10	0.0656
11	0.0633
12	0.0757
13	0.1140
14	0.0839
15	0.0661
16	0.1085
17	0.0765
18	0.0513
Mean ± σ	0.0725 ± 0.0225
Maximum	0.1140
Minimum	0.0336

The radio analytical method accuracy was regularly checked through participation in intercomparison exercises organized by the International Atomic Energy Agency (IAEA).

To check the method reproducibility, the ^{210}Po analyses were repeated five times in sample 1. The standard deviation results 13 and 10 % for the powder and the coffee respectively.

Results

Coffee powder

Table 1 reports the ^{210}Po activity concentration found in 18 Italian different brands of coffee powder, the arithmetical mean activity concentration, the standard deviation, the minimum and maximum values. ^{210}Po was always detectable for all samples. The values obtained are accompanied by an uncertainty of 10 % of the activity; they ranged between 0.034 Bq kg^{-1} and 0.114 kg^{-1} with a mean value of 0.073 ± 0.023 Bq kg^{-1}.

Table 2 reports the ^{40}K, ^{214}Pb, ^{214}Bi, ^{228}Ac, ^{212}Pb, ^{208}Tl activity concentration found in 16 Italian different brands of coffee powder, the arithmetical mean activity concentration, the standard deviation, the minimum and maximum values.

The mean ^{40}K activity was 907.4 ± 115.6 Bq kg^{-1} and ranged from 793.8 ± 53.6 to 1274 ± 80.9 Bq kg^{-1}.

The activity concentration of ^{214}Pb and ^{214}Bi, indicators of ^{226}Ra, given as mean value of the two radionuclides, ranged from <3.66 to 19.19 ± 4.55 Bq kg^{-1} with a mean

Table 2 ^{40}K, ^{214}Pb, ^{214}Bi, ^{228}Ac, ^{212}Pb, ^{208}Tl activity concentration (Bq kg^{-1}) in 16 Italian different brands of coffee powder, the arithmetical mean activity concentration, the standard deviation, the minimum and maximum values

Brand	^{40}K	^{214}Pb	^{214}Bi	^{228}Ac	^{212}Pb	^{208}Tl
1	931.1 ± 57.68	6.32 ± 1.3	5.13 ± 1.16	11.7 ± 3.08	5.87 ± 0.99	6.72 ± 1.64
2	927.9 ± 64.82	12.0 ± 2.93	17.9 ± 3.65	14.8 ± 7.54	13.6 ± 2.68	<4.57
4	1274 ± 80.87	17.9 ± 4.37	20.4 ± 4.74	14.7 ± 8.65	8.24 ± 3.32	12.3 ± 5.83
5	1013 ± 69.01	13.5 ± 3.19	18.9 ± 3.74	<4.86	11.2 ± 2.55	13.6 ± 4.09
7	891.1 ± 56.24	8.48 ± 1.38	9.81 ± 1.37	<4.86	6.20 ± 1.00	9.42 ± 1.79
8	793.8 ± 53.56	<3.66	<3.66	<4.86	5.86 ± 1.44	9.56 ± 4.3
9	863.4 ± 54.07	11.2 ± 1.24	10.6 ± 1.27	13.5 ± 3.19	6.85 ± 0.98	6.7 ± 1.47
10	852.6 ± 58.44	7.01 ± 2.46	9.8 ± 2.57	19.3 ± 8.22	11.3 ± 2.19	21.1 ± 8.65
11	843.8 ± 52.48	9.68 ± 2.4	9.82 ± 2.55	15.6 ± 7.07	6.48 ± 1.87	7.15 ± 3.18
12	890.9 ± 55.46	7.88 ± 3.73	<3.66	<4.86	7.94 ± 1.76	13.1 ± 4.93
13	904.7 ± 60.75	6.07 ± 1.18	4.16 ± 1.25	16.5 ± 4.32	4.95 ± 0.96	7.75 ± 1.79
14	950.5 ± 59.43	8.37 ± 2.31	14.3 ± 2.81	12.1 ± 5.75	10.1 ± 1.95	14.8 ± 4.94
15	828.1 ± 55.86	7.23 ± 2.07	10.9 ± 2.31	<4.86	9.98 ± 1.89	<4.57
16	811.2 ± 55.04	12.1 ± 2.56	11.0 ± 2.61	7.87 ± 3.8	6.21 ± 1.79	<4.57
17	796.3 ± 54.49	7.03 ± 2.51	12.2 ± 2.93	<4.86	13.0 ± 2.2	14.4 ± 6.14
18	945.9 ± 59.78	8.59 ± 1.24	8.38 ± 1.24	11.2 ± 2.88	4.64 ± 1.00	6.81 ± 1.74
Mean ± σ	907.4 ± 115.6	9.56 ± 3.95	11.7 ± 4.78	13.7 ± 3.20	8.28 ± 2.88	11.0 ± 5.90
Minimum	793.8 ± 53.56	<3.66	<3.66	<4.86	4.64	<4.57
Maximum	1275 ± 80.87	17.9 ± 4.37	20.4 ± 4.74	19.3 ± 8.22	13.6 ± 2.68	21.1 ± 8.65

Table 3 Comparison between ^{210}Po activity (Bq) in coffee beverage (coffee, 660 ml) and ^{210}Po concentration in coffee beverage without water contribution

Sample	Coffee	Coffee–water contribution
1	1.30×10^{-3}	1.06×10^{-3}
2	1.64×10^{-3}	1.40×10^{-3}
3	1.87×10^{-3}	1.63×10^{-3}
4	1.10×10^{-3}	8.63×10^{-4}
5	9.68×10^{-4}	7.31×10^{-4}
6	1.23×10^{-3}	9.93×10^{-4}
7	9.84×10^{-4}	7.47×10^{-4}
8	8.67×10^{-4}	6.30×10^{-4}
9	1.23×10^{-3}	9.93×10^{-4}
10	7.91×10^{-4}	5.54×10^{-4}
11	1.17×10^{-3}	9.33×10^{-4}
12	8.02×10^{-4}	5.65×10^{-4}
13	1.82×10^{-3}	1.58×10^{-3}
14	1.28×10^{-3}	1.04×10^{-3}
15	1.03×10^{-3}	7.93×10^{-4}
16	4.72×10^{-3}	4.48×10^{-3}
17	7.24×10^{-4}	4.87×10^{-4}
18	6.75×10^{-4}	4.38×10^{-4}
Mean ± σ	$1.34 \times 10^{-3} \pm 9.12 \times 10^{-4}$	$1.11 \times 10^{-3} \pm 9.12 \times 10^{-4}$
Minimum	6.75×10^{-4}	4.38×10^{-4}
Maximum	4.72×10^{-3}	4.48×10^{-3}

Table 4 Percentage of ^{210}Po transferred from coffee powered (60 g) to the coffee beverage (660 ml)

Brand	% of transfer
1	33
2	20
3	25
4	22
5	16
6	33
7	20
8	11
9	30
10	14
11	25
12	12
13	23
14	21
15	20
16	20
17	11
18	14
Mean ± σ	20.5 ± 6.9
Maximum	33
Minimum	11

Table 5 Activity (Bq) in coffee powder and in coffee grounds and percentage of transfer from coffee powered (60 g) to the coffee beverage (660 ml) for ^{40}K, ^{214}Pb, ^{214}Bi, ^{228}Ac, ^{212}Pb, ^{208}Tl

Radionuclide	Brand	Coffee powder	Coffee grounds	% of transfer	Mean (%)
^{40}K					
	12	53.45	12.07	78	
	14	49.69	8.77	82	80.0
	17	47.78	12.35	74	
	18	56.75	7.75	86	
^{214}Pb					
	12	0.47	0.50	–	
	14	0.43	0.36	16	33.5
	17	0.42	0.43	–	
	18	0.55	0.27	51	
^{214}Bi					
	12	–	–	–	
	14	0.65	0.50	23	
	17	0.73	0.40	45	24.7
	18	0.50	0.47	6	
^{228}Ac					
	12	–	–	–	
	14	–	–	–	30.0
	17	–	–	–	
	18	0.67	0.47	30	
^{212}Pb					
	12	0.48	0.43	10	
	14	0.60	0.43	28	35.1
	17	0.78	0.26	67	
	18	0.30	0.33	0	
^{228}Tl					
	12	0.78	0.39	50	
	14	–	–	–	53.5
	17	0.86	0.37	57	
	18	0.41	0.45	–	

value of 10.61 ± 4.02 Bq kg^{-1}. This low activity concentration indicates low level of ^{226}Ra, notwithstanding the measurements were not done at radioactive equilibrium between ^{226}Ra and ^{214}Pb–^{214}Bi.

The activity concentration of ^{228}Ac, ^{228}Ra indicator, ranged from <4.86 to 19.3 ± 8.22 Bq kg^{-1} with a mean value of 13.7 ± 3.20 Bq kg^{-1}.

The activity concentration of ^{212}Pb, ^{224}Ra indicator, ranged from 4.64 ± 1.00 to 13.61 ± 2.68 Bq kg^{-1} with a mean value of 8.28 ± 2.88 Bq kg^{-1}.

The activity concentration of ^{208}Tl, ^{224}Ra indicator, ranged from <4.56 to 21.14 ± 8.65 Bq kg^{-1} with a mean value of 11.03 ± 4.34 Bq kg^{-1}.

In all samples, the artificial ^{137}Cs resulted below the detection limit (2.0 Bq kg^{-1}).

Coffee beverage

Table 3 reports the ^{210}Po activity in 660 ml of coffee beverage; the ^{210}Po activity concentration resulted $2.03 \times 10^{-3} \pm 1.02$ 10^{-3} Bq l^{-1}. In the same table, ^{210}Po activity in coffee beverage after subtraction of tap water contribution was reported; the water contribution resulted of 17 %.

Radionuclide extraction percentage in coffee beverage

The determination of a radionuclide extraction percentage in beverage was done on 18 samples for ^{210}Po and on four samples for the radionuclides gamma emitters.

The radionuclide extraction percentage was calculated by the ratio between the radionuclide activity obtaining subtracting the activity found in the coffee grounds to that found in the powder and the total quantity of the activity radionuclide found in the powder. The arithmetical mean value of percentage of ^{210}Po extraction in coffee beverage resulted 20.5 ± 6.9 % (Table 4). The percentage of transfer of gamma emitters, ^{40}K, ^{214}Pb, ^{214}Bi, ^{228}Ac, ^{212}Pb, ^{208}Tl from coffee powered (60 g) to the coffee beverage (660 ml) was reported in Table 5. The percentage of transfer resulted of 80.0, 33.5, 24.7, 30.0, 35.1 and 53.5 % for ^{40}K, ^{214}Pb, ^{214}Bi, ^{228}Ac, ^{212}Pb and ^{208}Tl respectively.

Conclusions

Coffee may appear to be a simple drink, but in reality it is a highly complex product. Natural and artificial radioactivity has been measured in foodstuffs but there are very few data for this beverage. The aim of this research was to determine the activity of ^{210}Po and natural gamma emitters as ^{40}K, ^{214}Pb, ^{214}Bi, ^{228}Ac, ^{212}Pb and ^{212}Bi in 18 brands of coffee powder and in coffee.

^{40}K showed a mean activity of 907.4 ± 115.6 Bq kg^{-1}. ^{210}Po activity resulted $7.25 \times 10^{-2} \pm 2.25$ 10^{-2} Bq kg^{-1}. The mean activity concentration of ^{214}Pb and ^{214}Bi, indicators of ^{226}Ra, given as mean value of the two radionuclides, resulted 10.61 ± 4.02 Bq kg^{-1}. ^{228}Ac, ^{228}Ra indicator, showed a mean activity concentration of 13.73 ± 3.20 Bq kg^{-1}. The mean activity concentration of ^{212}Pb, ^{224}Ra indicator, was 8.28 ± 2.88 Bq kg^{-1}. ^{208}Tl, ^{224}Ra indicator, presented a mean activity concentration of 11.03 ± 4.34 Bq kg^{-1}. In all samples, the artificial ^{137}Cs resulted below the detection limit (2.0 Bq kg^{-1}).

The data obtained from in this research provide information on the activity concentration of natural radionuclides and increase the databases on natural radioactivity.

As the presence of radionuclides in coffee beans constitutes one of the pathways for their migration to human, via food chain, the coffee quality must strictly controlled and the study of radionuclide concentration in such matrix results to have a great significance.

In terms of the coffee market, it is very important to determine the radioactive values of these commodities for customers. The results obtained give also a useful information to carry out a dose assessment due to ingestion of coffee beverage.

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