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STUDY ON NATURAL RADIONUCLIDE ACTIVITIES IN MEAT SAMPLES CONSUMED IN SÃO PAULO CITY, BRAZIL

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ABSTRACT

Consumption of food is usually the most important route by which natural and artificial radionuclides can enter the human body. An assessment of radionuclide levels in different foods and diets is therefore important to estimate the intake of these radionuclides by man. The contamination by radionuclides can occur via the food chain (soil, root, plant and animal), with emphasis to the long half-life radionuclides, which can also have their transfer through the animal meat. The inclusion of meat in human nutrition is important because it is an excellent source of high quality protein, nutrient related to construction and cell regeneration. This work aims the determination of natural radionuclides (^{234}U, ^{235}U, ^{238}U, ^{228}Th, ^{230}Th, ^{232}Th, ^{226}Ra, ^{228}Ra, and ^{210}Pb) in meat samples. Five groups of samples were analyzed, such as cattle meat (beef), fish, pork, poultry, and processed meat, after radiochemical separation followed by alpha or alpha beta spectrophotometry, and total count quantification. The determination of these radionuclides is very important because they are products of the natural decay series of ^{238}U and ^{232}Th, being easily found in meat samples.

Keywords: meat, natural radionuclides, radiochemical separation.

1. INTRODUCTION

Most of the radiation received by humans comes from natural sources, either by external or internal exposure. The external exposure is due to the extraterrestrial radiation and natural radionuclides in the environment while the internal exposure to radionuclides present in the human body, as a result of incorporation via inhalation or ingestion of food and water [1].

The primary routes man exposure to radionuclides present in the ecosystem are: (1) the radiation gamma originating from decay products of radon gas present in the soil, rocks and suspended particles in the atmospheric air; (2) the retention of these radionuclides by plants that are ingested by animals who later transfer these radionuclides to man through the meat, milk and derivatives; (3) dissolution of radionuclides in the water of rivers, seas and oceans contaminating the marine flora and fauna and waters used for drinking, cooking and for irrigation; (4) ^{226}Ra emanation for the atmosphere and inhalation of various other radionuclides resulting from the decay of the element itself; (5) resuspension of solid

particles of radionuclides that are inhaled and transferred to the ecosystem entering the food chain and consequently being available to the human organism [2].

The food consumption is one of the main entry routes of radionuclides in the human body. On a global scale, the incorporation of radionuclides in biosphere is due to fallout. These elements present in the atmosphere reach plants, soil and water and can contaminate the environment and food. The main sources of radionuclides in food diet are milk and meat, especially for long half-life radionuclides [3].

When radionuclides in meat are determined using the photon spectrometer system, the most of the naturally occurring radionuclides are found to have specific activities below the detectable limit [4].

This study has as objective the determination of natural radionuclide of long half-life, ^{234}U, ^{235}U, ^{238}U, ^{228}Th, ^{230}Th, ^{232}Th, ^{226}Ra, ^{228}Ra, and ^{210}Pb, in meat samples. The samples were subdivided into five groups: cattle meat, fish, pork, poultry, and processed meat. The determination of these radionuclides is very important, because they are natural series decay products of ^{238}U and ^{232}Th.

2. MATERIALS AND METHODS

2.1 Food Group Preparation

The following meat groups were analyzed: cattle meat, fish, pork, poultry and processed meat.

The composition and the preparation process of each meat group were described in more details in Avegliano *et al.* [5]. These group samples are part of the List Food of the 2nd Brazilian Total Diet Study. The Table 1 shows the food items of each meat group.

Table l: Food items and daily consumption of meat groups

Meat group	Food items	Daily consumption per capita (g)
Cattle meat (beef)	Rump	67.10
Fish	sardine	11.80
	codfish	2.69
	sardine in oil	0.41
Pork	slender leg	9.70
Poultry	chicken	33.00
Processed meat	sausage	5.74
	cooked sausage	3.60
	bovine liver	1.91
	mortadella	1.58
	ham	1.46

The meat samples were prepared table-ready, which means that the food was prepared following the normal preparation habits of population. The preparation includes discarding non edible portions (bones, fat, etc.) and cooking. No seasonings, oils or other condiments were added in the preparation.

After the table-ready preparation, similar samples were mixed to form the groups. They were then homogenized, freeze-dried and grounded to a fine powder.

2.2 Experimental Procedures

Approximately 10 g of food sample freeze-dried were burnt in order to obtain ash at 450 °C for 24 hours in a muffle furnace. After that, the ash samples were dissolved with three concentrate acids (nitric, perchloric, and hydrofluoric). The final solution was evaporated and the salts dissolved with 8 M nitric acid. The solution was divided into two parts in order to determine all radioisotopes.

2.3 Ra and Pb Determination

These radioisotopes were purified by radiochemical separation, with Pb^{2+} (20 $mg.mL^{-1}$) and Ba^{2+} (20 $mg.mL^{-1}$) used as carriers to determine loss during the analysis. The radionuclide was coprecipitated as $Ba(Ra)SO_4$ and $PbSO_4$ using 3 M sulfuric acid, solution of 1 M citric acid with 0.1% phenol and concentrated ammonium hydroxide at pH 6.5 using methyl orange as indicator.

After that, the precipitate was dissolved with 2g of EDTA and 5 mL of concentrated ammonium hydroxide. The ^{226}Ra and ^{228}Ra were selectively coprecipitated with addition of 5 mL of 25 mg/mL ammonium sulphate and concentrated glacial acetic acid. The supernatant was saved to ^{210}Pb determination.

The Ra precipitate was filtered through a 0.45 µm Millipore membrane. The samples were stored for 30 days to await the secular equilibrium between ^{226}Ra and ^{222}Rn, and ^{228}Ra and ^{228}Ac.

The ^{210}Pb contained in the supernatant was precipitated using 1mL of 1 M Na_2S. The precipitate was dissolved with 5 M HNO_3, evaporated to dryness and the salts dissolved with deionized water. The lead was finally precipitated in the form of $PbCrO_4$ with addition of 40% ammonium acetate and solution of 30% Na_2CrO_4.

The precipitate formed was filtered through a 0.45 µm Millipore membrane. The samples were stored for 30 days to wait the secular equilibrium between ^{210}Pb and ^{210}Bi.

The chemical recovery of Ra and Pb were obtained by gravimetry.

The ^{226}Ra, ^{228}Ra and ^{210}Pb were quantified using an Ultra Low Level Alpha and Beta total counting with a gaseous flow proportional detector model S5-XLB Tennelec from Canberra Industries during 120 minutes.

2.4 U and Th Determination

A tracer was added in the other fraction of the sample solution containing uranium and thorium ^{229}Th and ^{232}U in order to determine the chemical recuperation.

Radiochemical Th separation was carried out using Dowex 1x2 anionic exchange resin pre-conditioned with 8 M nitric acid. The sample in nitric media was percolated through anionic column Dowex 1x2 resin. Thorium was retained and the uranium followed to the effluent. Thorium was eluted with concentrated hydrochloric acid. The eluted solution was dried on a hot plate and the salts dissolved with the electroplating solution at pH 2.3. The electroplating solution was prepared using concentrated sulfuric acid, 0.3 M sodium sulfate and deionized water.

The effluent containing U was dried, the salts dissolved with 3M HNO_3, and the solution was percolated through a chromatography column UTEVA pre-conditioned with 3 M HNO_3. The effluent was discarded. After that, 9 M HCl was added through the column to modify the medium, the effluent was discarded, and U was then eluted with 0.01 M HCl. The effluent was dried on a hot plate and the salts dissolved with 3 M sulfuric acid and 0.8 M ammonium sulfate.

The radioisotopes were electrodeposited on polished silver plates using an electrical current of 1.0 A to thorium and 1.2 A to uranium during 60 minutes.

The U and Th isotopes were analyzed in an Alpha Analyst spectrometer with 12 PIPS (Passivated Implanted Planar Silicon) detectors (counting efficiency 18%), and Genie 2000/Alpha Analyst spectroscopy systems, from Canberra Industries during 200,000 seconds.

Alpha particles energies 5.41 MeV for ^{228}Th, 4.90 MeV for the ^{229}Th tracer, 4.67 MeV for ^{230}Th, and 4.01 MeV for ^{232}Th were used to quantify thorium.

Alpha particles energies 4.31 MeV for the ^{232}U tracer, 4.74 MeV for ^{234}U, 4.47 MeV for ^{235}U, and 4.19 MeV for ^{238}U were used to quantify uranium.

3. RESULTS AND DISCUSSION

The methodology applied in this study was validated through analysis of Reference material IAEA Soil 327. The results obtained showed that the methods applied to determine natural radioisotopes after radiochemical separation and counting of Total Alpha and Beta emitters were reliable and accurate. Relative error values were less than 3% for ^{230}Th and less than 17% for ^{210}Pb [6].

The chemical recovery of Ra isotopes determination was between 77 to 92% and between 50 to 78% of Pb was recovered.

The chemical recovery of thorium analysis varied from 85 to 100%. The results obtained for ^{230}Th and ^{232}Th activities for freeze-dried food samples were below the detection limit, being below 0.17 Bq.kg^{-1}for ^{230}Th, and below 0.12 Bq.kg^{-1} for ^{232}Th.

The chemical recovery of the uranium analysis varied from 75 to 100%. The results obtained for uranium isotopes activity for all freeze-dried food samples were below the detection limit (0.26 $Bq.kg^{-1}$) for ^{235}U.

Table 2 shows the results obtained for determinations of ^{234}U, ^{238}U, ^{228}Th, ^{226}Ra, ^{228}Ra, and ^{210}Pb activities in the freeze dried meat group samples.

Table 2. Results of the determination in the freeze-dried meat samples.

Sample	^{234}U ($Bq.kg^{-1}$)	^{238}U ($Bq.kg^{-1}$)	^{228}Th ($Bq.kg^{-1}$)	^{226}Ra ($Bq.kg^{-1}$)	^{228}Ra ($Bq.kg^{-1}$)	^{210}Pb ($Bq.kg^{-1}$)
Cattle meat	< 0.14	< 0.14	1.20 ± 0.10	5.00 ± 1.40	< 2.00	9.70 ± 1.60
Fish	0.84 ± 0,10	< 0.14	1.40 ± 0.12	1.12 ± 0.15	< 2.00	31.78 ± 7.62
Pork	1.62 ± 0.12	0.21 ± 0.03	1.77 ± 0.14	0.68 ± 0.05	< 2.00	< 4.00
Poultry	< 0.14	< 0.14	0.90 ± 0.10	4.50 ± 1.30	2.10 ± 0.60	< 4.00
Processed Meat	< 0.14	< 0.14	0.80 ± 0.10	5.20 ± 1.40	4.90 ± 0.50	< 4.00

Table 3 shows the results obtained for the determinations of ^{234}U, ^{238}U, ^{228}Th, ^{226}Ra, ^{228}Ra, and ^{210}Pb activities in the table-ready meat group samples which took into consideration the water loss during the freeze-dried process.

Table 3. Results of the determinations in the table-ready meat samples.

Sample	^{234}U ($Bq.kg^{-1}$)	^{238}U ($Bq.kg^{-1}$)	^{228}Th ($Bq.kg^{-1}$)	^{226}Ra ($Bq.kg^{-1}$)	^{228}Ra ($Bq.kg^{-1}$)	^{210}Pb ($Bq.kg^{-1}$)
Cattle Meat	< 0.06	< 0.06	0.56 ± 0.05	2.32 ± 0.65	< 0.93	4.50 ± 0.74
Fish	0.33 ± 0.04	< 0.06	0.55 ± 0.05	0.44 ± 0.06	< 1.35	12.59 ± 3.02
Pork	1.09 ± 0.08	0.14 ± 0.02	1.19 ± 0.09	0.46 ± 0.03	< 0.79	< 2.70
Poultry	< 0.05	< 0.05	0.31 ± 0.03	1.54 ± 0.44	0.72 ± 0.21	< 1.37
Processed Meat	< 0.07	< 0.07	0.38 ± 0.05	2.48 ± 0.67	2.34 ± 0.24	< 1.91

Akinloye *et al.* [7] obtained mean activity of the natural radionuclides (^{226}Ra, and ^{228}Ra) in three types of meat in nature (goat meat, beef and pork) from stock animals within the Obafemi Awolowo University (OAU) environment in Nigeria. They measured these radionuclides using a high-purity germanium detector. The specific activities of ^{226}Ra in the meat samples ranged from 1.11 to 5.83 $Bq.kg^{-1}$ with a mean of 3.10 ± 1.52 $Bq.kg^{-1}$. ^{228}Ra was not detectable in both beef and pork but had a range of 1.53 – 1.63 $Bq.kg^{-1}$ with a mean of 1.58 ± 0.30 $Bq.kg^{-1}$ in goat meat. The mean activity values of ^{226}Ra and ^{228}Ra for the poultry were 2.59 ± 0.48 and 0.78 ± 0.13 $Bq.kg^{-1}$, respectively. In the 2nd Brazilian Total Study Diet, the detection limits obtained for these radionuclides were lower because the applied quantification methodology by alpha beta total count after radiochemical separation is more sensitive than the high-purity germanium detector.

Table 4 presents daily intakes of ^{234}U, ^{238}U, ^{228}Th, ^{226}Ra, ^{228}Ra, and ^{210}Pb in meats samples. The daily consumption per capita for each food group was considered for these results.

Table 4. Daily intakes of the natural radionuclides.

Sample	Daily consumption (kg)	^{234}U (Bq.kg^{-1})	^{238}U (Bq.kg^{-1})	^{228}Th (Bq.kg^{-1})	^{226}Ra (Bq.kg^{-1})	^{228}Ra (Bq.kg^{-1})	^{210}Pb (Bq.kg^{-1})
Cattle Meat	0.0671	$< 4.36\ 10^{-3}$	$< 4.36\ 10^{-3}$	$3.73\ 10^{-2}$	$1.56\ 10^{-1}$	$< 6.22\ 10^{-2}$	$3.02\ 10^{-1}$
Fish	0.0149	$4.94\ 10^{-3}$	$< 8.26\ 10^{-4}$	$8.23\ 10^{-3}$	$5.59\ 10^{-3}$	$< 1.18\ 10^{-2}$	$1.88\ 10^{-1}$
Pork	0.0097	$1.06\ 10^{-2}$	$1.40\ 10^{-3}$	$1.16\ 10^{-2}$	$4.43\ 10^{-3}$	$< 1.31\ 10^{-2}$	$< 2.62\ 10^{-2}$
Poultry	0.0330	$< 1.58\ 10^{-3}$	$< 1.58\ 10^{-3}$	$1.01\ 10^{-2}$	$5.07\ 10^{-2}$	$2.37\ 10^{-2}$	$< 4.51\ 10^{-2}$
Processed Meat	0.0143	$< 9.55\ 10^{-4}$	$< 9.55\ 10^{-4}$	$5.46\ 10^{-3}$	$3.55\ 10^{-2}$	$3.34\ 10^{-2}$	$< 2.73\ 10^{-2}$

The results obtained for the daily radionuclide intake of the various meat types showed that those of ^{226}Ra ranged from 0.4 to 8.1 mBq.d^{-1}with a mean of 4.9 ± 0.4 mBq.d^{-1}. Since ^{228}Ra was not detectable in beef and pork its daily intake could not be estimated. However, a mean value of intake of 2.2 ± 0.4 mBq.d^{-1} was obtained for goat meat. The mean daily intakes of ^{226}Ra and ^{228}Ra for the two types of poultry were 6.15 ± 0.70 and 1.9 ± 0.3 mBq.d^{-1}, respectively [7].

The daily intakes of natural long-lived radionuclides obtained in this study are in good agreement with the ones referred by UNSCEAR [8], showing that the total intake of the inhabitants from São Paulo city in the lower range.

4. CONCLUSION

This study determined radionuclides from the natural series, ^{234}U, ^{235}U, ^{238}U, ^{228}Th, ^{230}Th, ^{232}Th, ^{226}Ra, ^{228}Ra, and ^{210}Pb in meat samples, showing low radioactivity concentrations. The daily intakes of naturally occuring long-lived radionuclides are in good agreement with the ones referred by UNSCEAR, showing that the total intake.

The radiochemical separation followed by alpha spectrometry or alpha beta total count were demonstrated to be accurate and sensitive methods for determinations in comparasion with other methods.

5. ACKNOWLEDGEMENTS

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