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# PRELIMINARY RESULTS OF NATURAL RADIOACTIVITY IN FLOURS COMMERCIALIZED IN THE CITY SÃO PAULO BY HIGH RESOLUTION GAMMA RAY SPECTROMETRY

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## ABSTRACT

In the present work, the activity concentrations of natural radionuclides $^{226}$Ra, $^{232}$Th and $^{40}$K were analyzed in 32 samples of wheat, corn, manioc and rice flours commonly used by residents of the city of São Paulo, Brazil. All samples were dried, homogenized and tightly sealed in standard 100 mL HDPE flasks with plan screw cap and bubble spigot and measured for 150 ks by gamma spectrometry with an Extended Range Coaxial germanium detector (Canberra GX4020 detector), after approximately 4 weeks storage, in order to ensure secular equilibrium. All spectra were analyzed with the InterWinner 6.0 software. The $^{226}$Ra activity concentration was determined from the weighted average concentrations of $^{214}$Pb and $^{214}$Bi. The $^{232}$Th activity concentration was determined from the weighted average concentrations of $^{228}$Ac, $^{212}$Pb and $^{212}$Bi and the $^{40}$K one from its single gamma transition. The $^{226}$Ra, $^{232}$Th and $^{40}$K activity concentrations results ranged, respectively, from 0.97 Bq/kg ± 0.42 Bq/kg to 4.75 Bq/kg ± 0.82 Bq/kg, from 8.07 Bq/kg ± 2.36 Bq/kg to 19.02 Bq/kg ± 3.56 Bq/kg and from 25.22 Bq/kg ± 4.38 Bq/kg to 153.5 Bq/kg ± 17.02 Bq/kg.

## 1. INTRODUCTION

Humans are continuously and unavoidably exposed to natural sources of ionizing radiation and natural radioactivity has the greatest contribution to the effective dose received by mankind. The major terrestrial contribution to internal exposure come from the long-lived alpha and beta emitters from the $^{238}$U and $^{232}$Th natural decay series and from the single occurring radionuclide $^{40}$K [1, 2, 3].

One of the most relevant contributions comes from the dose per food intake. The concentrations of natural radionuclides in food vary significantly according different levels of background radiation and variety of these foods, climate and agricultural culture conditions. Since most of the foods consumed are cultivated on soil, the radionuclides present are metabolically incorporated into the plants and consequently ingested, increasing the contribution to the dose per intake [1, 4].

Previous studies [5, 6, 7] show dose data by ingestion of foods widely used in Brazilian cuisine, such as flours used basically as a source of food. With significant consumption of these flours as raw material for the production of several foods, it becomes necessary studies that present relevant data when it comes to the dose by ingestion and the activity concentration of the radionuclides possibly present in these flours.

Other relevance for the analysis of these flours is given to large consumption as a staple in the world diet the flours are used as a means of intervention in order to eradicate iron and folic acid deficiency anemia, which is an epidemiological problem in several countries, affecting the whole population and the maternal/infant groups. The Ministry of Health - MS following the recommendations of the World Health Organization - WHO, through DRC No. 344, determined the mandatory fortification of wheat and corn flours with iron and folic acid in Brazil in 2002. The implementation of fortification of wheat flour and corn is a simple, effective and low-cost strategy as a supply of vitamins and minerals in the diet, supplying this deficiency in large population groups [8, 9].

In the present work, activity concentrations of the natural radionuclides $^{226}$Ra, $^{232}$Th and $^{40}$K in 32 samples of several flours marketed in the city of São Paulo, consequently the most consumed in the southeastern region of Brazil were assessed.

## 2. MATERIALS AND METHODS

Initially, 32 samples of different brands and producers, commonly available in the local markets were selected, namely wheat, corn, manioc and rice flours. The classification of samples they are summarized in table. 1.

Table 1. Classification of wheat samples (w), corn (c), manioc (m) and rice (r) according to the normative instruction of the Ministry of Agriculture, Livestock and Supply [10].

| Number of samples according to their classification | | | |
|---|---|---|---|
| Sample | Type 1 | Type 2 | Description |
| Wheat | 17 | 1 | 3 Integral<br>15 Traditional |
| Corn | 8 | - | 3 flocked<br>5 fubá |
| Rice | 3 | - | 1 flocked<br>2 traditional |
| Manioc | 3 | - | 2 fine<br>1 thick |

### 2.1 Samples preparation and counting methodology

The samples were dried, homogenized and hermetically sealed in standard 100 mL HDPE flasks with plan screw cap and bubble spigot in order to ensure secular equilibrium per a period of approximately 4 weeks before counting, in order to allow the secular equilibrium in the $^{238}U$ and $^{232}Th$ series [11, 12].

All samples were measured by high resolution gamma spectrometry [11] with a Canberra extended range hyperpure germanium (HPGe) detector (GX4020), with associated electronics and a 920-8 ORTEC EG&G Spectrum Master 4k multichannel analyzer and an acquisition time of 150 ks. The efficiency and background radiation curves were obtained using, respectively, a multi-element standard aqueous radioactive solution sample and an ultra-pure water sample, in the same geometry as the flour samples. The efficiency and background curves are verified at least two times every year since 1997, by proficiency tests of the National Intercomparison Program (PNI - Programa Nacional de Intercomparação, in portuguese) carry out by the Institute of Radioprotection and Dosimetry (Instituto de Radioproteção e Dosimetria, in Portuguese) [13]. Usually in gamma spectrometry analysis of solid samples, the self attenuation component is assessed in order to allow correct estimation of the data [14]. In the case of the flour samples, the average apparent density was found as 0.84 $g.cm^{-3}$ and therefore required no correction for the self attenuation contribution, once the density of the standard aqueous radioactive solution is approximately 1.0 $g.cm^{-3}$.

## 2.2 Samples Analysis

All flour spectra were analyzed with the InterWinner 6.0 software [15].

The activity concentration of a single transition was calculated as [16, 17]:

$$A_i(E_{\gamma i}) = \frac{C_{\gamma i}}{\varepsilon_{\gamma i}.\mathrm{I}_{\gamma i}.t_l.m} \tag{1}$$

Where:

$A(E_{\gamma i})$ = activity of the considered gamma transition of the isotope X in the sample (Bq/kg);
$C_{\gamma i}$ = net area for the gamma transition with energy ($E_{\gamma i}$) emitted by X;
$\varepsilon_{\gamma i}$ = detector efficiency for the considered gamma transition;
$I_{\gamma i}$ = probability of emission of the gamma transition with energy (E);
$t_l$ = counting live time (s);
m = sample mass (kg);

The activity of $^{40}K$ was calculated through its single gamma transition of 1460.83 keV, as in Eq. (1). The activity of $^{226}Ra$ was determined by the weighted mean of the $^{214}Pb$ and $^{214}Bi$ gamma transitions, as in Eq. (2) and the activity of $^{232}Th$ by the weighted mean of the $^{228}Ac$, $^{212}Pb$ and $^{212}Bi$ [12, 15].

$$C_{^{226}Ra} = \frac{\dfrac{C_{^{214}Pb}}{\sigma^2_{^{214}Pb}} + \dfrac{C_{^{214}Bi}}{\sigma^2_{^{214}Bi}}}{\dfrac{1}{\sigma^2_{^{214}Pb}} + \dfrac{1}{\sigma^2_{^{214}Bi}}} \qquad (2)$$

Where:

$C_{^{226}Ra}$ : $^{226}$Ra activity concentration (Bq/kg);
$C_{^{214}Pb}$ : $^{214}$Pb activity concentration (Bq/kg);
$\sigma_{^{214}Pb}$ : Uncertainty of $^{214}$Pb activity concentration (Bq/kg);
$C_{^{214}Bi}$ : Uncertainty of $^{214}$Bi activity concentration (Bq/kg);
$\sigma_{^{214}Bi}$ : Uncertainty of $^{214}$Bi activity concentration (Bq/kg).

$$C_{^{232}Th} = \frac{\dfrac{C_{^{212}Pb}}{\sigma^2_{^{212}Pb}} + \dfrac{C_{^{212}Bi}}{\sigma^2_{^{212}Bi}} + \dfrac{C_{^{228}Ac}}{\sigma^2_{^{228}Ac}}}{\dfrac{1}{\sigma^2_{^{212}Pb}} + \dfrac{1}{\sigma^2_{^{212}Bi}} + \dfrac{1}{\sigma^2_{^{228}Ac}}} \qquad (3)$$

Where:

$C_{^{232}Th}$ : $^{232}$Th activity concentration (Bq/kg);
$C_{^{212}Pb}$ : $^{212}$Pb activity concentration (Bq/kg);
$\sigma_{^{212}Pb}$ : Uncertainty of $^{212}$Pb activity concentration (Bq/kg);
$C_{^{212}Bi}$ : Uncertainty of $^{212}$Bi activity concentration (Bq/kg);
$\sigma_{^{212}Bi}$ : Uncertainty of $^{212}$Bi activity concentration (Bq/kg);
$C_{^{228}Ac}$ : Uncertainty of $^{228}$Ac activity concentration (Bq/kg);
$\sigma_{^{228}Ac}$ : Uncertainty of $^{228}$Ac activity concentration (Bq/kg).

## 3 RESULTS AND CONCLUSIONS

Activities concentrations values, for all measured samples, lies in the range from 0.97 Bq/kg ± 0.42 Bq/kg to 4.75 Bq/kg ± 0.82 Bq/kg for $^{226}$Ra, 8.07 Bq/kg ± 2.36 Bq/kg to 19.02 Bq/kg ± 3.56 Bq/kg for $^{232}$Th and 153.5 Bq/kg ± 17.02 Bq/kg for $^{40}$K. All results are summarized in Fig. 1.

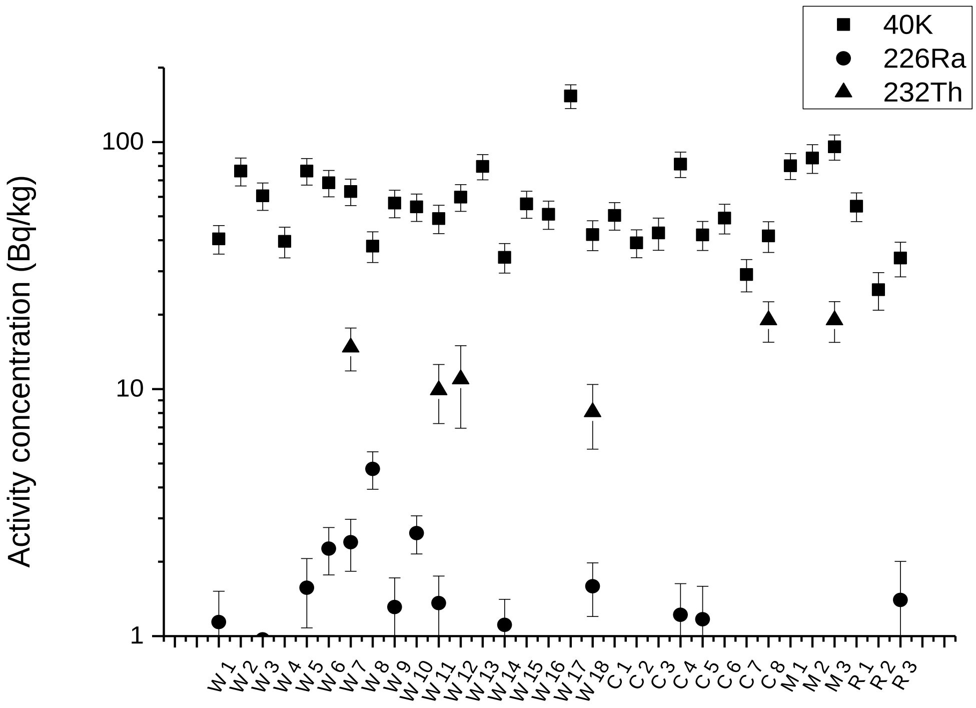


Fig. 1 Natural radionuclides activity concentration values, assessed for 32 different samples of flours. Sample type are labelled as W for wheat, C for corn, M for manioc and R for rice.

The highest values of activity concentration were obtained for $^{40}$K, where the highest value corresponding to the wheat flour sample w17. Only few samples presented activity concentration values for $^{226}$Ra and $^{232}$Th for samples wheat flour w8 and manioc flour m3, respectively.

It is observed that all the samples present activities concentration of greater for $^{40}$K. The $^{232}$Th of activity concentrations are higher than the activity concentrations of $^{226}$Ra. The activity concentration values obtained for all the radionuclides considered in all analyzed samples show very close values with some exceptions. Probably, these variations occur according to the different levels of background radiation, climate and agricultural conditions in which these foods are grown [1].

Further, the activity concentrations results will be used to estimate the effective dose by ingestion to the human consumers and compared to the recommended dose limits of UNSCEAR and ICRP (International Commission on Radiological Protection) of 140 $\mu$Sv.a$^{-1}$ and 1 mSv.a$^{-1}$ respectively, for general public by ingestion of radioactive elements present in food [2, 18].

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