

# DETERMINATION OF GROSS ALPHA, GROSS BETA, AND NATURAL RADIONUCLIDES ( $^{210}\text{Po}$ , $^{210}\text{Pb}$ , $^{238}\text{U}$ , $^{232}\text{Th}$ AND $^{40}\text{K}$ ) ACTIVITY CONCENTRATIONS IN BREAD AND THEIR CONTRIBUTION TO THE EFFECTIVE DOSE

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**Abstract.** In this paper the gross alpha, gross beta and natural radionuclides ( $^{210}\text{Po}$ ,  $^{210}\text{Pb}$ ,  $^{238}\text{U}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$ ) activities were determined in bread samples collected during 2013–2015 from Galati, Braila and Vrancea counties, South-Eastern Romania. The concentrations of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  were determined after self deposition on nickel disc by gross alpha measurements. The concentrations of  $^{238}\text{U}$  and  $^{232}\text{Th}$  were calculated after the determination of the concentration of  $^{nat}\text{U}$  and  $^{nat}\text{Th}$  through spectrophotometric measurements of arsenazo III- $\text{U}^{4+}$  complex and arsenazo III- $\text{Th}^{4+}$  complex, respectively. The concentrations of  $^{40}\text{K}$  were measured by gamma-spectrometry using a NaI(Tl) detector. The gross alpha and gross beta activity in bread samples ranged from 0.006 to 0.180 Bq kg<sup>-1</sup>, and from 21.3 to 45.7 Bq kg<sup>-1</sup>, respectively. The radionuclide concentrations (fresh weight) in bread ranged in the intervals 0.014–0.031 Bq kg<sup>-1</sup> for  $^{210}\text{Po}$ , 0.006–0.080 Bq kg<sup>-1</sup> for  $^{210}\text{Pb}$ , 0.009–0.040 Bq kg<sup>-1</sup> for  $^{238}\text{U}$ , 0.001–0.021 Bq kg<sup>-1</sup> for  $^{232}\text{Th}$ , and 10.4–31.4 Bq kg<sup>-1</sup> for  $^{40}\text{K}$ . The annual effective dose obtained ranged between 0.023  $\mu\text{Sv y}^{-1}$  (for  $^{232}\text{Th}$ ) and 19.572  $\mu\text{Sv y}^{-1}$  (for  $^{40}\text{K}$ ).

**Key words:** bread, natural radionuclides,  $^{210}\text{Po}$ ,  $^{210}\text{Pb}$ ,  $^{238}\text{U}$ ,  $^{232}\text{Th}$ ,  $^{40}\text{K}$ , effective dose.

## 1. INTRODUCTION

Natural and artificial radionuclides usually enter the human body by the most important pathway which is the consumption of food. In order to estimate the intake of the radionuclides, it is important to make an assessment of their level in different diets, food items [1–6], and water [7–9].

In the global estimate, the contribution to the annual effective dose from natural sources is  $2.4 \text{ mSv y}^{-1}$ . Of this,  $1.2 \text{ mSv y}^{-1}$  is due to radon,  $0.5 \text{ mSv y}^{-1}$  to terrestrial gamma rays,  $0.4 \text{ mSv y}^{-1}$  to cosmic rays and  $0.3 \text{ mSv y}^{-1}$  to ingestion of food and drinking water [10].

Bread is an important foodstuff for people and it represents an essential part of the food pyramid's base [11]. *World Health Organization* (WHO) recommendation is to eat bread several times a day. A number of European countries recommend a daily bread intake of about 250 g, which corresponds to 4–8 slices, depending on national food habits [12].

The components of bread are: wheat flour, water, salt and yeast. The presence of natural radionuclides in grains and, implicit, in wheat flour, is due to the existence of the primordial radionuclides originating from earth. Isotopes of natural decay series uranium and thorium and their decay product are found in soil, vegetation and also in water [13]. The natural radionuclides from soil come from the natural decay series (uranium-238, thorium-232, and uranium-235) and from the contribution of  $^{40}\text{K}$ . Soil-to-plant and plant-to-human body are important pathways for the ingestion of radionuclides [14].  $^{210}\text{Po}$ , a radionuclide of the  $^{238}\text{U}$  series, with a high specific activity of  $166 \text{ TBq g}^{-1}$ , is the most radiotoxic alpha emitter. The internal exposure to radiation caused by alpha emitters is more dangerous than the external exposure. The reasons for this are the relatively high energy of alpha particles, of about 5.3 MeV, and their very short range in tissues.  $^{210}\text{Po}$  is concentrated in the soft tissue such as liver and kidney [15]. In the same series,  $^{210}\text{Pb}$  is among the most radiotoxic beta emitters, its half-life is 22.17 years and it has a high conversion factor among the beta emitters. Lead is accumulated in bone, brain, liver and kidney [10], thorium in human lungs, liver and skeleton tissues, uranium in lungs and kidneys, and potassium in muscles [16].

Similar to other countries, Romania has a radioactivity monitoring system of food through the Ionizing Radiation Hygiene Laboratories and Regional Public Health Centres [17]. The *Ionizing Radiation Hygiene Laboratory* (IRHL) from Galati, Romania, implements the National Public Health Programme in Romania regarding the monitoring of radioactivity of drinking water and foodstuff according with the Euratom requirements.

To the best of our knowledge, the information regarding the natural radionuclides in bread is very scarce in literature and no published data on their concentrations is currently available for Romania.

The aim of this study was two-fold: 1) to determine the concentrations of gross alpha and gross beta activity and of naturally occurring radionuclides  $^{210}\text{Po}$ ,  $^{210}\text{Pb}$ ,  $^{238}\text{U}$ ,  $^{232}\text{Th}$ , and  $^{40}\text{K}$  in bread samples collected during 2013–2015 from Galati, Braila and Vrancea counties, South-Eastern Romania, and 2) to determine the radiation doses received by adult people from radionuclide intake due to bread consumption. This work is a continuation of the previous study performed in IRHL

Galati, Romania, for the assessment of the annual effective dose due to intake of drinking water [8] and food supplements [6] by adult people.

## 2. MATERIAL AND METHODS

### 2.1. SAMPLE PREPARATION

The samples of white bread were obtained in the period 2013–2015 from various markets of Galati, Braila and Vrancea, three counties located in the South-Eastern region of Romania and prepared for analysis in the *Ionizing Radiation Hygiene Laboratory* (IRHL) at Galati, Romania. The mass of the each bread sample used for the determination of all the radionuclides from this study was about 3 kg. Each bread sample was collected from the same lot. Firstly, the bread sample was ashed to 125°C. From this residue, about 30 grams were used for the determination of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  concentrations. Secondly, the other rest of the residue calcined up to 450°C was used for the determination of  $^{40}\text{K}$  concentration by gamma-spectrometry. After that, the same ash sample, which was used for gamma-spectrometry, it was also utilized for the measurement of gross alpha and gross beta activity and the determination of  $^{\text{nat}}\text{U}$  and  $^{\text{nat}}\text{Th}$ , followed by calculation of  $^{238}\text{U}$  and  $^{232}\text{Th}$  concentrations. All the samples were analysed in duplicate for quality assessment.

According with ISO/IEC 17025:2005, there is a constant preoccupation for quality assurance of results. Moreover, IRHL Galati, Romania, is participating every year to various interlaboratory comparisons for water samples, one of the exercise being described by Jobbágy *et al.* (2014) [18] and Sobiech-Matura *et al.* (2015) [19].

### 2.2. DETERMINATION OF $^{210}\text{Po}$ AND $^{210}\text{Pb}$ CONCENTRATION

The residue resulted from drying to 125°C an amount of about 30 grams was mineralized with  $\text{HNO}_3$  (65%) and  $\text{HCl}$  (37%), then with  $\text{HCl}$  (37%) and  $\text{H}_2\text{O}_2$  (30%). After that, ascorbic acid and hydroxylamine hydrochloride was added and the solution was used for self deposition onto nickel disc. The self deposition was performed at 90°C for at least 4 hours. The gross alpha activity of the nickel disc was measured with the aid of the MPC-2000-DP system (Protean Instruments Corporation), calibrated at Horia Hulubei National R&D Institute for Physics and Nuclear Engineering (IFIN-HH), Bucharest, Romania. In this study, each measurement lasted 400 minutes.

The concentration of  $^{210}\text{Po}$  was calculated with the formula:

$$C_{^{210}\text{Po}} = \frac{(R_{disc} - R_0) \times m}{\varepsilon \times \eta \times M \times m_1} \text{ (Bq kg}^{-1}\text{)}, \quad (1)$$

where:  $C_{^{210}\text{Po}}$  is the concentration of the  $^{210}\text{Po}$  (Bq kg<sup>-1</sup>),  $R_{disc}$  is the rate of measurement of self deposited nickel disc (counts s<sup>-1</sup>),  $R_0$  is the rate of measurement for background (counts s<sup>-1</sup>),  $m$  is the mass of the residue resulted after ashing up to 200 °C (grams),  $\varepsilon$  is efficiency of the detector of MPC-2000-DP system,  $\eta$  is radiochemical recovery,  $M$  is the value of the fresh bread mass (kg), and  $m_1$  is the mass of the residue which was taken for radiochemical separation (grams) – about 30 grams.

The rest of the solution was stored for three months and after that the procedure of self deposition on the nickel disc was repeated. The nickel disc was measured for gross alpha activity. The  $^{210}\text{Pb}$  activity was determined through its daughter,  $^{210}\text{Po}$ , using the formula:

$$C_{^{210}\text{Po}} = \frac{(R_{disc} - R_0) \times m}{\varepsilon \times \eta \times M \times m_1 \times 0.37} \text{ (Bq kg}^{-1}\text{)} \quad (2)$$

where:  $C_{^{210}\text{Po}}$  is the concentration of the  $^{210}\text{Pb}$  (Bq kg<sup>-1</sup>),  $R_{disc}$  is the rate of measurement of nickel disc after the second self deposited (counts s<sup>-1</sup>),  $R_0$  is the rate of measurement for background (counts s<sup>-1</sup>) and 0.37 is the correction factor due to secular equilibrium between  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  after three months.

In this study the measurements were performed using gross alpha activities for  $^{210}\text{Po}$  and  $^{210}\text{Pb}$ , hence the radiochemical recovery and the analysis of  $^{210}\text{Po}$  or  $^{210}\text{Pb}$  are not simultaneous. Therefore, for the chemical recovery it was assumed the value 100%. For this reason, the results for concentrations of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  from this work might be higher.

### 2.3. DETERMINATION OF GROSS ALPHA AND GROSS BETA ACTIVITY

For the general screening the gross alpha and gross beta activity was measured using the ash which resulted after calcination of the sample up to 450 °C. Approximately 50 mg ash sample was transferred on a steel planchet and it was measured for 100 minutes using the MPC-2000-DP system, ten times per sample, as described by Pintilie *et al.* (2016) [8].

The gross alpha and gross beta activity was calculated using the formula:

$$C_{\alpha,\beta} = \frac{(R_{sample} - R_0) \times m \times a}{\varepsilon \times M \times m_2 \times a_1} \text{ (Bq kg}^{-1}\text{)} \quad (3)$$

where:  $C_{\alpha,\beta}$  is the gross alpha/gross beta activity (Bq kg<sup>-1</sup>),  $R_{sample}$  is the rate of measurement of the ash transferred onto the planchet (counts s<sup>-1</sup>),  $R_0$  is the rate of

alpha/beta background (count s<sup>-1</sup>),  $m$  (grams) is the total resulted residue from the fresh sample with mass  $M$  which was ashed to 125 °C,  $a$  (grams) is the total ash resulted from  $m$  residue,  $M$  (kg) is the fresh weight initially processed in analysis,  $\varepsilon$  is efficiency of the detector,  $m_2$  (grams) is the mass of the residue taken for burning to 450 °C,  $a_1$  (grams) is the mass of the ash transferred onto the planchet.

#### 2.4. DETERMINATION OF <sup>40</sup>K CONCENTRATION

The concentration of <sup>40</sup>K was determined using a gamma-ray spectrometer with NaI(Tl) detector coupled with a multichannel analyzer and the gamma spectra were processed with the aid of Maestro-32 software. The ash which was resulted through burning at 450 °C, was measured in the same geometry used for calibration. The ash was placed on the detector into a Sarpagan type box. Acquisition of the gamma spectrum was performed for 7200 seconds. The concentration of <sup>40</sup>K was determined using the gamma-ray peak of 1460 keV.

The formula which was used to calculate the concentration of <sup>40</sup>K is given below:

$$C_{^{40}\text{K}} = \frac{A_{net} \times m \times a}{\varepsilon \times M \times I_{^{40}\text{K}} \times m_2 \times a_2 \times t} \quad (\text{Bq kg}^{-1}) \quad (4)$$

where:  $C_{^{40}\text{K}}$  is the concentration of <sup>40</sup>K (Bq kg<sup>-1</sup>),  $A_{net}$  is the number of counts for the net peak area, corrected for the background of the gamma peak (at 1460 keV),  $m$  (grams) is the total resulted residue from the fresh sample with mass  $M$  which was ashed to 125°C,  $m_2$  (grams) is the mass of the residue taken for burning to 450°C,  $M$  (kg) is the fresh weight initially processed in the analysis,  $\varepsilon$  is the percentage of energy efficiency for <sup>40</sup>K (for the spectrometer used in this work,  $\varepsilon = (7.40 \pm 0.44) \times 10^{-3}$ , imp s<sup>-1</sup> Bq<sup>-1</sup>),  $I_{^{40}\text{K}}$  is the percentage of gamma-emission probability of <sup>40</sup>K,  $a$  is the total ash resulted from  $m$  residue through calcining to 450°C (grams),  $a_2$  is the mass of ash which is measured by gamma spectrometry (grams),  $t$  is the counting time. The gamma spectrometer was calibrated using a mixed source SEG 8-471 (<sup>60</sup>Co, <sup>137</sup>Cs, <sup>241</sup>Am), produced at IFIN-HH.

#### 2.5. DETERMINATION OF <sup>238</sup>U AND <sup>232</sup>Th CONCENTRATION

The concentration of the <sup>nat</sup>U and <sup>nat</sup>Th was determined through radiochemical separation and purification on a column with Dowex resin. The ash resulted after calcination at 450°C for acidic extraction of the cations was used. Acidic extraction was performed with HNO<sub>3</sub> (65%) and H<sub>2</sub>O<sub>2</sub> (30%) 1:1. After that, the solution was evaporated until a residue form. The residue was dissolved with

HCl 8N. Then, uranium was separated from thorium on a column with Dowex resin-Cl<sup>-</sup> form. Purification of the uranium and thorium on the column with Dowex resin-NO<sub>3</sub><sup>-</sup> form was performed. After that, the eluate containing uranium and thorium was measured at the wavelength of 665 nm, through Arsenazo III-U<sup>4+</sup> and Arsenazo III-Th<sup>4+</sup>, respectively, using a Perkin Elmer Lambda-11 UV-VIS spectrophotometer. U<sub>3</sub>O<sub>8</sub> and Th(NO<sub>3</sub>)<sub>5</sub> were used in order to determine the radiochemical recovery.

The concentration of uranium was calculated with the formula:

$$C_{natU} = \frac{c_{natU} \times 25.35 \times 10^{-3} \times m \times a}{\varepsilon \times M \times m_2 \times a_2 \times \eta_{natU}} \text{ (Bq kg}^{-1}\text{)}, \quad (5)$$

where:  $C_{natU}$  – is the concentration of the natural uranium (Bq kg<sup>-1</sup>),  $25.35 \times 10^{-3}$  represents the conversion coefficient of the concentration in µg to Bq activity for uranium,  $c_{natU}$  is the concentration of the uranium (in µg) in the  $a_2$  grams of ash taken for the determination, which was determined from the calibration curve equation for uranium:

$$E = c_{natU} \times p + q, \quad (6)$$

where:  $E$  is the extinction given by the spectrophotometer,  $p$  and  $q$  are coefficients of the calibration curve,  $m$  is the total resulted residue from the fresh sample with mass  $M$  which was ashed to 125°C (grams),  $m_2$  is the mass of the residue taken for burning to 450°C (grams),  $a$  is the total ash resulted from  $m$  residue through calcining at 450°C (grams),  $a_2$  is the mass of ash which was used for acidic extraction (grams), and  $\eta_{natU}$  is the radiochemical recovery of uranium.

Similarly, the concentration of thorium is calculated with the formula:

$$C_{natTh} = \frac{c_{natTh} \times 0.041 \times m \times a}{\varepsilon \times M \times m_2 \times a_2 \times \eta_{natTh}} \text{ (Bq kg}^{-1}\text{)} \quad (7)$$

where:  $C_{natTh}$  is the concentration of the natural thorium (Bq kg<sup>-1</sup>), 0.041 represents the conversion coefficient of the concentration in µg to Bq activity for thorium,  $c_{natU}$  is the concentration of thorium (in µg) in the  $a_2$  grams ash taken for determination, which was determined from the calibration curve for thorium.

After the determination of natural uranium and thorium concentrations, the specific activities of <sup>238</sup>U and <sup>232</sup>Th were calculated, taking into account that the natural isotopic abundance of <sup>238</sup>U from <sup>nat</sup>U is 99.27%, and of <sup>232</sup>Th for natural thorium is 100%.

### 3. RESULTS AND DISCUSSION

#### 3.1. ACTIVITY CONCENTRATION OF NATURAL RADIONUCLIDES IN BREAD

The values of the activity obtained in this work for gross alpha, gross beta,  $^{210}\text{Po}$ ,  $^{210}\text{Pb}$ ,  $^{238}\text{U}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  in the bread samples from Galati (GL), Braila (BR) and Vrancea (VN) counties in Romania are shown in Table 1. The samples labeled SB1-SB3 were collected in 2013, SB4-SB6 in 2014 and SB7-SB9 in 2015.

Table 1

Gross alpha, gross beta and radionuclide activity concentrations and average (Ave.) (in  $\text{Bq kg}^{-1}$ ) in bread samples from Galati (GL), Braila (BR) and Vrancea (VN), Romania, collected during 2013–2015

| Sample code | Location  | $\Lambda_{\alpha}$<br>( $\text{Bq kg}^{-1}$ ) | $\Lambda_{\beta}$<br>( $\text{Bq kg}^{-1}$ ) | $\Lambda_{^{210}\text{Po}}$<br>( $\text{Bq kg}^{-1}$ ) | $\Lambda_{^{210}\text{Pb}}$<br>( $\text{Bq kg}^{-1}$ ) | $\Lambda_{^{238}\text{U}}$<br>( $\text{Bq kg}^{-1}$ ) | $\Lambda_{^{232}\text{Th}}$<br>( $\text{Bq kg}^{-1}$ ) | $\Lambda_{^{40}\text{K}}$<br>( $\text{Bq kg}^{-1}$ ) |
|-------------|-----------|---|--|--|--|---|--|--|
| SB1         | GL        | 0.006±0.001                                   | 25.4±6.3                                     | 0.028±0.008  | 0.080±0.024  | 0.009±0.002   | 0.001±0.0003   | 21.5±3.2   |
| SB2         | BR        | 0.006±0.001                                   | 21.3±5.32                                    | 0.028±0.008  | 0.006±0.002  | 0.012±0.003   | 0.001±0.0003   | 10.4±1.5   |
| SB3         | VN        | 0.006±0.001                                   | 35.9±1.7                                     | 0.014±0.004  | 0.010±0.003  | 0.012±0.003   | 0.009±0.002  | 24.5±3.6   |
| SB4         | GL        | 0.180±0.050                                   | 21.4±5.3                                     | 0.021±0.006  | 0.006±0.002  | 0.032±0.009   | 0.021±0.006  | 21.16±3.1  |
| SB5         | BR        | 0.006±0.001                                   | 45.7±11.4                                    | 0.019±0.006  | 0.007±0.002  | 0.040±0.012   | 0.003±0.0009   | 25.2±3.7   |
| SB6         | VN        | 0.006±0.001                                   | 39.4±9.8                                     | 0.031±0.009  | 0.010±0.003  | 0.030±0.009   | 0.003±0.0009   | 23.4±3.5   |
| SB7         | GL        | 0.026±0.007                                   | 29.3±7.32                                    | 0.019±0.006  | 0.014±0.004  | 0.037±0.011   | 0.019±0.005  | 31.4±4.7   |
| SB8         | BR        | 0.032±0.009                                   | 31.4±7.8                                     | 0.022±0.007  | 0.013±0.004  | 0.028±0.008   | 0.015±0.004  | 29.4±4.4   |
| SB9         | VN        | 0.015±0.004                                   | 30.4±7.6                                     | 0.021±0.006  | 0.010±0.003  | 0.019±0.005   | 0.009±0.002  | 27.5±4.1   |
| Ave.        | All sites | 0.031±0.008                                   | 31.15±7.5                                    | 0.023±0.008  | 0.017±0.005  | 0.024±0.007   | 0.009±0.002  | 23.8±3.5   |

From the data presented in the Table 1, it can be seen that the lowest concentration of the natural radionuclides in bread samples was found for thorium, while the highest concentration was found for  $^{40}\text{K}$ .

The gross alpha activity was smaller than  $1 \text{ Bq kg}^{-1}$  for all the bread samples. The values of the gross alpha activities are much lower than the gross beta activities of the investigated bread samples. No correlation was found between gross alpha activity and the concentrations of alpha emitters. Also, no correlation was found between gross beta activity and the concentrations of beta emitters.

The highest values of the gross alpha activity and the concentration of  $^{210}\text{Po}$ , the most toxic alpha emitter, were found in SB4/2014 and SB6/2014 samples, respectively. There is no correlation between gross alpha activity and the concentration of  $^{210}\text{Po}$ .

The lowest value of the gross alpha activity, gross beta activity and specific activity of  $^{210}\text{Po}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  was found in the SB3/2013 bread sample from Vrancea County.

The ratio between the gross beta activity and the concentration of  $^{40}\text{K}$  ranged from 0.93 to 2.04.

The ranges and/or averages of concentrations of the radionuclides in bread investigated in this study, together with those reported from different regions of the world in bread and other related ingredient items, are shown in Table 2.

Table 2

The concentrations of  $^{210}\text{Po}$ ,  $^{210}\text{Pb}$ ,  $^{238}\text{U}$ ,  $^{232}\text{Th}$ , and  $^{40}\text{K}$  ( $\text{Bq kg}^{-1}$ ) in bread samples and other related ingredient items from different regions of the world

| Sample type | Country/ Report | R/ Ave.   | $\Lambda_{210\text{Po}}$ ( $\text{Bq kg}^{-1}$ ) | $\Lambda_{210\text{Pb}}$ ( $\text{Bq kg}^{-1}$ ) | $\Lambda_{238\text{U}}$ ( $\text{Bq kg}^{-1}$ ) | $\Lambda_{232\text{Th}}$ ( $\text{Bq kg}^{-1}$ ) | $\Lambda_{40\text{K}}$ ( $\text{Bq kg}^{-1}$ ) | Ref.      |
|-------------|-----------------|-----------|--|--|---|--|--|-----------|
| wheat       | Spain           | R         | –  | –  | –   | –  | 67.0–122.6                                     | [20]      |
| wheat       | Poland          | R         | –  | –  | –   | –  | 127.9–145.1                                    | [4]       |
| grains      | South India     | Ave.      | –  | –  | –   | –  | 482.7±19.2                                     | [21]      |
| wheat flour | Iraq            | R<br>Ave. | –  | –  | 1.086–12.532<br>6.6025                          | 0.126–4.298<br>1.9465                            | 41.84–264.73<br>133.097                        | [22]      |
| wheat flour | Finland         | R         | –  | 0.058–0.030                                      | –   | –  | –  | [23]      |
| white bread | Saudi Arabia    | R<br>Ave. | –  | –  | –   | <5.6–26.0<br>16.4±2.3                            | 203–297<br>240±22                              | [24]      |
| wheat grain | Pakistan        | R<br>Ave. | –  | –  | –   | 1.0–1.5<br>1.3±0.2                               | 95.7–146.9<br>122.8±23.1                       | [25]      |
| flours      | Italy           | R<br>Ave. | 0.020–0.114<br>0.061±0.026                       | –  | –   | –  | –  | [2]       |
| grain       | UNSCEAR         | R         | 0.020–0.360                                      | 0.049–0.059                                      | 0.006–0.085                                     | 0.0016–0.033                                     | –  | [10]      |
| white bread | Romania         | R<br>Ave. | 0.014–0.031<br>0.023±0.008                       | 0.006–0.08<br>0.017±0.005                        | 0.009–0.04<br>0.024±0.007                       | 0.001–0.021<br>0.009±0.002                       | 10.4–31.4<br>23.83±3.57                        | This work |

R = Range; Ave. = Average.

### 3.2. EFFECTIVE DOSE EQUIVALENT DUE TO INGESTION OF BREAD

Taking into consideration the statistical data regarding the annual consumption of bread, the annual effective dose for adults was calculated and shown in Table 3. According to the data from the National Statistical Institute, the annual consumption of bread in Romania for the years 2013, 2014 and 2015 was  $102 \text{ kg y}^{-1}$ ,  $99.99 \text{ kg y}^{-1}$ , and  $100.53 \text{ kg y}^{-1}$ , respectively ([www.insse.ro](http://www.insse.ro)) [26].



The annual effective dose is calculated with the formula:

$$D = \Sigma(C_X \times BI \times CF), \quad (8)$$

where:  $C_X$  is the concentration of radionuclide  $X$  ( $X = {}^{210}\text{Po}, {}^{210}\text{Pb}, {}^{238}\text{U}, {}^{232}\text{Th}, {}^{40}\text{K}$ ) ( $\text{Bq kg}^{-1}$ ),  $BI$  is the annual bread consumption ( $\text{kg year}^{-1}$ ),  $CF$  is effective dose conversion factor for adults ( $\text{Sv Bq}^{-1}$ ). For the calculations the following dose conversion factors for adults were used:  $1.2 \times 10^{-6} \text{ Sv Bq}^{-1}$  for  ${}^{210}\text{Po}$ ,  $6.9 \times 10^{-7} \text{ Sv Bq}^{-1}$  for  ${}^{210}\text{Pb}$ ,  $4.5 \times 10^{-8} \text{ Sv Bq}^{-1}$  for  ${}^{238}\text{U}$ ,  $2.3 \times 10^{-7} \text{ Sv Bq}^{-1}$  for  ${}^{232}\text{Th}$  and  $6.9 \times 10^{-9} \text{ Sv Bq}^{-1}$  for  ${}^{40}\text{K}$  [10].

Table 3

Effective dose due to ingestion of natural radionuclides from bread consumed per year in Romania in 2013, 2014, 2015 ( $\mu\text{Sv y}^{-1}$ )

| Sample code | Annual consumption (kg) | Effective dose due to ingestion of:           |   |  |   |   | Total effective dose (adults) ( $\mu\text{Sv y}^{-1}$ ) |
|-------------|-------------------------|---|---|--|---|---|---|
|             |                         | ${}^{210}\text{Po}$ ( $\mu\text{Sv y}^{-1}$ ) | ${}^{210}\text{Pb}$ ( $\mu\text{Sv y}^{-1}$ ) | ${}^{238}\text{U}$ ( $\mu\text{Sv y}^{-1}$ ) | ${}^{232}\text{Th}$ ( $\mu\text{Sv y}^{-1}$ ) | ${}^{40}\text{K}$ ( $\mu\text{Sv y}^{-1}$ ) |   |
| SB1         | 102                     | 3.427±1.028                                   | 5.630±1.689                                   | 0.041±0.010                                  | 0.023±0.006                                   | 13.596±2.039                                | 22.719±4.773  |
| SB2         | 102                     | 3.427±1.028                                   | 0.422±0.126                                   | 0.055±0.014                                  | 0.023±0.006                                   | 6.576±0.986                                 | 10.505±2.161  |
| SB3         | 102                     | 1.714±0.514                                   | 0.703±0.211                                   | 0.055±0.014                                  | 0.211±0.052                                   | 15.493±2.324                                | 18.177±3.115  |
| SB4         | 99.99                   | 2.520±0.756                                   | 0.413±0.124                                   | 0.144±0.036                                  | 0.483±0.120                                   | 13.118±1.967                                | 16.679±3.004  |
| SB5         | 99.99                   | 2.280±0.684                                   | 0.482±0.144                                   | 0.180±0.045                                  | 0.069±0.017                                   | 15.623±2.343                                | 18.635±3.234  |
| SB6         | 99.99                   | 3.720±1.116                                   | 0.689±0.206                                   | 0.135±0.034                                  | 0.069±0.017                                   | 14.507±2.176                                | 19.121±3.550  |
| SB7         | 100.53                  | 2.292±0.688                                   | 0.971±0.291                                   | 0.167±0.042                                  | 0.439±0.109                                   | 19.572±2.935                                | 23.442±4.066  |
| SB8         | 100.53                  | 2.654±0.769                                   | 0.901±0.270                                   | 0.127±0.032                                  | 0.346±0.086                                   | 18.325±2.748                                | 22.355±3.934  |
| SB9         | 100.53                  | 2.534±0.760                                   | 0.693±0.208                                   | 0.086±0.021                                  | 0.208±0.052                                   | 17.141±2.571                                | 20.662±3.612  |

The highest value of the total effective dose presented in Table 3 was found for the sample SB7/2015 ( $19.572 \pm 2.935 \mu\text{Sv y}^{-1}$ ), in which case the contribution of  ${}^{40}\text{K}$  was the highest. The lowest value for the annual effective dose ( $0.023 \pm 0.006 \mu\text{Sv y}^{-1}$ ) was calculated for the sample SB2/2013. In this case the gross alpha activity, the gross beta activity, the concentration of  ${}^{210}\text{Pb}$ , and the concentration of  ${}^{232}\text{Th}$  were the lowest.

The annual effective dose due to the ingestion of natural radionuclides from bread allows the assessment of the health risk for adults. The lowest effective dose calculated for 1 kg of bread was due to ingestion of  ${}^{232}\text{Th}$  ( $0.023 \pm 0.057 \text{ nSv kg}^{-1}$ ), while the highest effective dose to ingestion of  ${}^{40}\text{K}$  ( $194.68 \pm 29.20 \text{ nSv kg}^{-1}$ ).

#### 4. CONCLUSIONS

Bread is a basic staple of our diet and the knowledge of radiological implications on the population which consumes this food is very important for human health.

The radiochemical separations for the determination of the concentrations of the  $^{210}\text{Po}$ ,  $^{210}\text{Pb}$ ,  $^{238}\text{U}$  and  $^{232}\text{Th}$ , and gamma spectrometry measurements for the determination of the concentration of  $^{40}\text{K}$  were carried out in samples of white bread from Romanian markets in Galati, Braila and Vrancea counties.

The average values of the concentrations of  $^{210}\text{Po}$ ,  $^{210}\text{Pb}$ ,  $^{238}\text{U}$ ,  $^{232}\text{Th}$ , and  $^{40}\text{K}$  natural radionuclides in the bread samples were the following:  $0.022\pm 0.006$ ,  $0.017\pm 0.005$ ,  $0.024\pm 0.006$ ,  $0.009\pm 0.002$ , and  $23.828\pm 3.574$  Bq  $\text{kg}^{-1}$ , respectively. The average of the annual effective doses due to ingestion of natural radionuclides from bread was calculated as  $19.14\pm 3.49$   $\mu\text{Sv y}^{-1}$ , which is lower than the recommended value for ingested dose (100  $\mu\text{Sv y}^{-1}$ ) according to the current EU legislation.

It is the first study performed in Romania regarding the naturally occurring radionuclides concentration in bread and the results will serve as a base in further investigations and statistical assessments carried out by National Institute of Public Health.

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