GROSS ALPHA, GROSS BETA AND ^{40}K ACTIVITIES AND DAILY EFFECTIVE DOSE DUE TO NATURAL RADIONUCLIDES FROM FOOD SUPPLEMENTS

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Abstract. The fast rhythm of everyday life has led to habit of people to consume food supplements. The consumption of food supplements represents a pathway for the ingestion of natural radionuclides. For the radiological risk assessment studies, 14 samples of food supplements have been analysed in this work for gross alpha and gross beta activities, as well as for concentrations of potassium-40. The concentrations of potassium-40 radioisotope were calculated on the basis of the concentrations of total potassium, determined by sequential *High-Resolution Continuum Source Atomic Absorption Spectroscopy* (HR-CS AAS) using the flame technique. The investigations revealed values for gross alpha activities, gross beta activities and the concentrations of ^{40}K in the ranges of 0.25 to 30.92 mBq g^{-1}, 2.18 to 769.59 mBq g^{-1} and <LOD (*limit of detection*) to 651.52 mBq g^{-1}, respectively. The maximum values were obtained for a supplement based on lucerne (alfalfa) extract, while the minimum ones were found for a supplement based on fish oil. For comparison purpose, 5 samples of frequently used drugs were also analysed. The corresponding values found for the investigated drugs are lower than those obtained for food supplements. High correlation coefficients have been found in all the studied samples between the gross alpha activity and gross beta activity (0.646), and between the gross beta activity and concentration of potassium-40 (0.778). The effective dose due to ingestion of natural radionuclides from food supplements varied as following: from 1.54 to 97.41 nSv per daily dose, assigned to ^{210}Po, and from 20.12 to 1393.92 nSv per daily dose, assigned to ^{228}Ra. The effective dose due to the ingestion of ^{40}K from food supplements ranged from 0 to 10.60 nSv per daily dose. The obtained values of effective doses were compared with the daily dose values recommended by the current EU legislation and *World Health Organization* (WHO).

Key words: food supplements, drugs, ^{40}K concentration, gross alpha/beta activities, daily effective dose.

1. INTRODUCTION

The consumption of food supplements and drugs became a necessity in people's lives. On one hand, food supplements are used for increasing the neuropsychological and physical capacities and for coping with stress; on the other hand, drugs are used for health improvements. People consume the food supplements for a few reasons: to balance the diet, relatively low cost, accessibility (the procurement without the medical prescription), natural origin and as alternative medicines without side effects [1]. Also, the medicinal plants are consumed for their therapeutic effect. Furthermore, it is estimated that 25% of the prescribed drugs worldwide are of plant origin, and 121 active substances are used in them [2].

Provision of European Directive 2002/46/CE [3] were transposed in Romanian national legislation through the Order no. 1069/2007 by norms regarding the food supplements [4].

Due to the demands of modern life, people tend to have an unbalanced diet that fails to bring them all the nutrients which they need. Food supplements are products whose purpose is to supplement the normal diet, being concentrated sources of nutrients or other substances with a nutritional or physiological effect, alone or in combination, marketed in dose form [4].

From the definitions of *Food and Drug Administration* (FDA) – an agency within the U.S. Department of Health and Human Services – these are called "dietary supplements" [5]. Food science defines the food supplements as specific products used to ensure a balanced diet, by facilitating the intake of those nutrients which are not present in the unbalanced or inadequate practiced diet. Of all the food supplements, multivitamins and preparations containing, along with vitamins, multiminerals are most consumed and a lot of multivitamin – multimineral dietary supplement products with various compositions are available aiming to compensate the possible deficiencies in human intake of these elements in diet [6].

Drug is any substance (human, animal, vegetal or chemical origin) or combination of substances which may be used or administered to humans or to restoring, correcting or modifying physiological functions by exerting a pharmacological, immunological or metabolic action, or to making a medical diagnosis [7]. The most used drugs are: cardiovascular, antibiotics, psycho-stimulants, analgesic drugs, and those used in the treatment of disorders of the gastro-intestinal tract.

The extraction of active principles from natural sources, in order to obtain the food supplements, is carried out along with the transfer of natural radionuclides in the final product. The natural radionuclides from natural sources used to obtain food supplements come from the natural decay chains ^{238}U, ^{232}Th, ^{235}U, and from the contribution of ^{40}K. Therefore, the consumption of food supplements along with the consumption of food and drinking water [8, 9] is a pathway to ingestion of natural radionuclides. The *United Nations Scientific Committee on the Effects of Atomic Radiation* (UNSCEAR) estimated that the exposure of the population to ionizing radiation from natural sources is 2.4 mSv y^{-1}: 0.3 mSv y^{-1} is due to ingestion of food and drinking water, 0.4 mSv y^{-1} to cosmic rays, 0.5 mSv y^{-1} to terrestrial gamma rays and 1.2 mSv y^{-1} to radon [10].

There are very scarce data in the literature about the radioactive content of food supplements. The aim of the present work was to determine the gross alpha and gross beta activities and the concentration of ^{40}K from food supplements and to assess the effective dose received by people through this intake.

2. EXPERIMENTAL

2.1. STUDIED AREA AND SAMPLING

Two types of samples were used for this study. Fourteen samples of food supplements, called "natural supplements", and five drugs have been analyzed. All the investigated food supplements and drugs are notified and approved by Ministry of Health of Romania. The samples was collected from various pharmacies of Galati County, Romania, during November 2015. There were selected food supplements of natural provenance in order to determine the gross alpha and gross beta activity and the concentration of ^{40}K, as a preliminary screening of natural radioactivity from these. Also, five drugs of synthetic origin were analysed in order to compare their results with results obtained for food supplements.

The samples were dried up to 105 ^{o}C and then calcined, by gradually increasing the temperature up to 450 ^{o}C to reduce the volume of the sample and to remove the organic matter. The resulted ashes were used to determine the gross alpha activities, gross beta activities, and the specific activities of ^{40}K.

2.2. MEASUREMENT OF GROSS ALPHA AND GROSS BETA ACTIVITY

For gross alpha and gross beta measurements, about 50 mg ash was uniformly distributed onto stainless planchet with the aid of a few drops of acetone, then this was dried at room temperature.

The gross alpha and gross beta activity was measured using the low background MPC-2000-DP counting system with a ZnS dual phosphor detector, calibrated at Horia Hulubei National Institute of R&D in Physics and Nuclear Engineering (IFIN-HH), Magurele, Romania. For current calibration, at Ionizing Radiation Laboratory, Department of Public Health Galati, a ^{241}Am reference source (serial: 2830) and a $^{90}Sr/^{90}Y$ reference source (serial: 9891) are used. Each sample was measured ten times, for 100 minutes.

In order to calculate the gross alpha and gross beta activity from food supplements and drugs, it was used the formula:

$$A_{\alpha/\beta} = \frac{(R-R_0)\times m}{m_1\times \varepsilon \times M} \quad (Bq\ g^{-1}) \tag{1}$$

where $A_{\alpha/\beta}$ is the gross alpha/beta activity of the measured sample (Bq g^{-1}), fresh weight, R is the rate of alpha or beta measurement for the sample (counts s^{-1}), R_0 is the rate of alpha, beta, respectively, measured for background (counts s^{-1}), m is the quantity of the ash resulted from the calcination of the fresh sample (g), m_1 is the weight of residue transferred to a stainless steel planchet for measurements (g), ε is the efficiency of the detector, and M is the weight of the fresh sample (g).

2.3. MEASUREMENT OF THE SPECIFIC ACTIVITY OF ^{40}K

The mass of the ash used for the determination of the concentration of ^{40}K was about 0.15 g. This ash was solved with a mixture of HCl (37%) and HNO_3 (65 %) at 200 °C for 1 hour, then the resulted solution was evaporated near dryness. After that, the residue was dissolved with HNO_3 (65%). The resulting solutions were transferred to a 50-mL volumetric flask and diluted to volume using high purity water (18.2 MΩ·cm electrical resistivity at 25 °C).

The concentration of total potassium was determined by the new sequential High-Resolution Continuum Source Atomic Absorption Spectroscopy (HR-CS AAS) at INPOLDE laboratories at Dunarea de Jos University of Galati, using a ContrAA700 (Analytik Jena, Germany) spectrometer equipped with a 300 W xenon short-arc lamp as continuum radiation source, through the flame atomization technique. HR-CS AAS technique is considered to be a novel, simple, fast, reliable and feasible approach to analyze essential elements in food products [6].

The type of used atomization flame was oxidant air/acetylene with the following parameters: fuel flow rate 80 L h^{-1} and burner height of 8 mm.

Each sample was analysed in duplicate for total potassium content. Quantification was performed using the linear calibration technique. Measurements were carried out at a wavelength of 766.4908 nm (main line, with relative sensitivity of 100%), using a spectral range of 200 nm and a wavelength integrated absorbance over the center pixel (CP) corresponding to CP ± 2 (5 pixels) for signal evaluation. The ASpect CS 2.1.0.0 software was employed to control the system.

A linear calibration curve of absorbance *versus* concentration of potassium was prepared from stock standard solution for AAS, TraceCERT, 1000 mg/L in 2% HNO_3 Suprapur grade (Sigma-Aldrich). The correlation coefficient (R^2) obtained in the linear least square fit of the calibration points was 0.9981, the value of LOD (limit of detection) was 0.0170 ppm, and the relative standard deviation (RSD) was 0.1–1.1 %.

The concentration of ^{40}K from food supplements and drugs was calculated with the formula:

$$A_{40K} = \frac{c_K \times m \times 0.0307}{m_2 \times M} \ (\mathrm{Bq\ g^{-1}}), \tag{2}$$

where A_{40K} is the specific activity of ^{40}K measured sample (Bq g^{-1}) fresh weight, c_K is the concentration of total potassium in the sample (mg), m is the quantity of the ash resulted from to calcine of the fresh sample (g), 0.0307 is specific activity of potassium-40 (Bq mg^{-1}), m_2 is the weight of ash took for the measurement using Atomic Absorption Spectroscopy, and M is the fresh mass of the sample (food supplements or drugs).

3. RESULTS AND DISCUSSIONS

3.1. GROSS ALPHA, GROSS BETA AND ^{40}K CONCENTRATION ACTIVITY

The samples, labeled FS1-FS14 and D1-D5, are food supplements and drugs, respectively. Gross alpha and gross beta activities, and concentrations of ^{40}K, in the food supplement and drugs determined in this study are presented in Table 1. The results are associated with the expanded uncertainty for coverage factor k = 2.

The gross alpha activity from food supplements and drugs ranged from (0.251±0.060) to (30.924±7.422) mBq g^{-1}, and from (0.370±0.089) to (13.165±3.160) mBq g^{-1}, respectively. The gross beta activity from food supplements and drugs ranged from (2.184±0.328) to (769.592±115.439) mBq g^{-1}, and from (10.581±1.587) to (83.719±12.558) mBq g^{-1}, respectively.

The maximum values for the concentration of ^{40}K were found in the food supplements as being 651.524±58.637 mBq g^{-1}, while in the drug samples set the concentrations of ^{40}K were below 0.279±0.024 mBq g^{-1}.

For all the studied samples the gross beta activity is higher than gross alpha activity, as also found for water samples in a previous study [8]. The analyses showed that the concentrations of ^{40}K were under the *limit of detection* (LOD) for three samples, one of food supplements and two of drugs.

There are very scarce data in the literature about the radioactive content of food supplements. Most food supplements are made based on the extracts from natural sources. The values for gross alpha and gross beta activities from some medicinal plants commonly used in Romania ranged from 3.20 to 10.75 Bq kg^{-1} and from 214.50 to 429.40 Bq kg^{-1}, respectively, as found by Oprea and co-workers (2011), [11]. Jevremovic *et al.* (2011) analyzed 18 different samples of medicinal herbs from the Serbian market, reporting values of ^{40}K concentration in the range 126.0–1243.7 Bq kg^{-1} [12].

Table 1

Gross alpha, gross beta, and ^{40}K concentration activities, A (mBq g^{-1}), fresh weight, in the investigated food supplements and drugs from Galati, Romania, 2015

Sample code	The base substance	A alpha	A beta	A ^{40}K
FS1	choline, lecithin extract	8.755±2.101	84.141±12.621	81.515±7.353
FS2	vitamins, minerals extract	1.461±0.351	126.368±18.955	239.228±21.530
FS3	lucerne extract	30.924±7.422	769.592±115.439	651.551±58.637
FS4	plants extract	6.154±1.477	376.699±56.505	101.562±9.136
FS5	extract from marine algae	7.246±1.739	240.589±36.088	412.229±37.100
FS6	fish oil	0.251±0.060	2.184±0.328	0.061±0.005
FS7	enzimes extract	1.840±0.442	104.904±15.736	80.333±7.230
FS8	C vitamin extract from rosehip	5.100±1.224	85.800±12.870	1.884±0.170
FS9	sylibium extract	5.460±1.311	28.328±4.249	19.160±1.724
FS10	vitamins extract	0.453±0.109	361.674±54.251	5.663±0.341
FS11	minerals extract	1.540±0.370	99.334±14.900	176.855±15.916
FS12	pancreatic enzymes extract	25.509±6.122	169.077±25.362	37.929±3.415
FS13	mint oil extract	2.67±0.640	37.304±5.595	<LOD
FS14	L-arginine extract	0.578±0.139	24.818±3.723	0.226±0.020
D1	acetylsalicylic acid	13.165±3.160	83.719±12.558	0.033±0.003
D2	carbamazepine	4.235±1.016	10.581±1.587	0.275±0.025
D3	pentoxifylline	1.635±0.392	18.499±2.775	0.279±0.024
D4	midodrine	4.250±1.020	61.280±9.192	<LOD
D5	dexketoprofen	0.370±0.089	49.200±7.380	<LOD

The highest gross beta activity and ^{40}K concentration were found for the same sample, FS3. The explanation would be that this food supplement is based on lucerne (alfalfa), and the high concentration of potassium recommended this supplement to be used for its alkalinizing properties, which is specified in the prospectus. Also, the concentration of potassium for FS3, which is 21.22 mg g^{-1} fresh weight (data not shown), is similar to the results found by Mielmann *et al.* (2017) [13]. The lowest gross alpha activity and gross beta activity were found in the sample FS6, based on fish oil, probably due to the low capacity of binding the natural radionuclides in oil extraction.

As regards the drug samples set, the highest gross alpha activity and gross beta activity were found in the drug sample D5. In this set, the highest concentration of potassium was found in sample D3.

There are high correlation coefficients between gross alpha activity and gross beta activity (0.646), and between the values of gross beta activity and concentration of potassium (0.778) from all the studied samples.

Table 2

The daily effective dose due to the ingestion of natural radionuclides from food supplements and drugs from Galati, Romania, 2015

Sample code	The daily effective dose due to ingestion of radionuclides:		
	^{210}Po (nSv per daily dose)	^{228}Ra (nSv per daily dose)	^{40}K (nSv × 10^{-3} per daily dose)
FS1	11.031±2.521	60.960±9.144	530±47
FS2	1.965±0.421	97.744±14.662	1662±149
FS3	97.409±8.906	1393.924±209.089	10603±954
FS4	12.474±1.772	439.009±65.851	1064±95
FS5	22.903±2.087	437.261±65.589	6732±605
FS6	4.017±0.072	20.122±3.018	5.0±0.4
FS7	3.478±0.530	114.005±17.101	784±71
FS8	9.180±1.469	88.803±13.320	18±2
FS9	6.753±1.573	20.144±3.022	122±11
FS10	2.309±0.131	1059.063±158.859	149±8
FS11	3.925±0.444	145.538±21.831	2328±210
FS12	62.474±7.347	238.097±35.715	480±43
FS13	12.496±0.769	100.385±15.058	–
FS14	1.540±0.167	38.016±5.702	3.0±0.3
D1	35.569±3.791	130.061±19.509	0.5±0.04
D2	1.906±1.220	2.738±0.411	0.6±0.05
D3	4.067±0.471	26.457±3.969	3.5±0.3
D4	2.272±1.224	18.837±2.826	–
D5	0.358±0.107	27.376±4.106	–

3.2. EQUIVALENT EFFECTIVE DOSE

Based on the data obtained from the analyses, the daily effective doses due to intake of natural supplements and drugs were calculated. In order to calculate the exposure of people to ionizing radiation through the ingestion of natural radionuclides from food supplements and drugs, the gross alpha activity and the gross beta activity were assumed to be from the contribution of ^{210}Po and ^{228}Ra, respectively. These radionuclides have the highest dose conversion factor and are the most radiotoxic alpha and beta emitters. Also, the daily effective dose due to ingestion of ^{40}K was assessed.

The daily effective dose due to ingestion of radionuclides from food supplements and drugs are presented in Table 2. In order to calculate the daily effective dose there were used the following dose conversion factors published by the World Health Organization [14]: 1.2 × 10^{-6} Sv Bq^{-1} for ^{210}Po, 6.9 × 10^{-7} Sv Bq^{-1} for ^{228}Ra, and 6.2 × 10^{-9} Sv Bq^{-1} for ^{40}K. The daily intake of natural supplements and drugs was considered accordingly to their specifications.

The daily effective dose for all the samples investigated ranged as following: from (0.358±0.107) to (97.409±8.906) nSv per daily dose, assigned to ^{210}Po, from (2.738±0.411) to (1393.924±209.089) nSv per daily dose, assigned to ^{228}Ra, and the level of ^{40}K was below (10.604±0.954) nSv per daily dose.

The values for the effective dose assigned to ^{210}Po are below the WHO recommended reference daily effective dose of 273 nSv day^{-1}, while the results for the daily effective dose assigned to the ^{228}Ra revealed values higher than the recommended value of the effective daily dose for the samples FS3, FS4, FS5 and FS10. However, for sustaining this statement, determinations of ^{228}Ra from this sample should be performed. Regarding the effective doses due to the ingestion of ^{40}K from food supplements, these are below the daily recommended value. The highest effective dose assigned to ^{40}K was found in FS3, as expected, because in this sample it was found the highest concentration of ^{40}K.

4. CONCLUSIONS

This study was carried out in order to evaluate the gross alpha and gross beta activities and the concentration of ^{40}K in several food supplements and drugs. Also, the daily effective doses due to ingestion of natural radionuclides through intake of food supplements and drugs were estimated to assess the radiological health risk for adults. The values for daily effective dose calculated to assign gross alpha activity as being due to ^{210}Po are lower than the recommended value. Starting from the assumption that the entire gross beta activity is due to ^{228}Ra radionuclide, values for daily effective dose higher to the WHO recommended dose were obtained for four food supplements. Whereas the consumption of the food supplements and drugs occurs for defined time period, for a good estimation of the effective dose due to ingestion of radionuclides from food supplements it is necessary to determine at least the concentrations of the radionuclides with the highest dose conversion factor, namely ^{210}Po and ^{228}Ra, which are the most radiotoxic natural isotopes. This represents the subject of future studies.

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