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DETERMINATION OF THE ACTIVITY CONCENTRATION OF ^{137}Cs AND ^{40}K IN BLUEBERRY-BASED PRODUCTS CONSUMED IN SERBIA

Borjana Vranješ[1*], Velibor Andrić[2], Mila Vranješ[2], Jelena Ajtić[1], Branislava Mitrović[1]

[1]Faculty of Veterinary Medicine, University of Belgrade, Belgrade, Serbia
[2]Vinča Institute of Nuclear Science, University of Belgrade, Belgrade, Serbia

Abstract. *The aim of the study is the investigation of the activity concentration of ^{137}Cs and ^{40}K in blueberry-based products that are available on the market in the Republic of Serbia. Samples were bought in stores during September 2017 and in total, ten packaged juices, two jams, two sweets and a fresh wild blueberry were measured. The activity concentrations of ^{137}Cs in blueberry-based juices, jams and sweets varied from <MDA to 4.1 Bq/kg, <MDA to 21 Bq/kg and 0.6 Bq/kg to 28 Bq/kg, respectively. The average activity concentration of ^{137}Cs in fresh wild blueberry was 4.1 Bq/kg. In Serbia, the recommended activity concentration of ^{137}Cs in juices and sweets is 15 Bq/kg and 150 Bq/kg in fresh blueberries. The tested samples of juices, jams, fresh wild blueberry and one of the sweets meet the set criteria for ^{137}Cs while one sweets sample exceeds the limit. The activity concentrations of ^{40}K in juices, jams and sweets varied from 3.5 to 55 Bq/kg, 13.9 to 19.2 Bq/kg and 17.2 to 227 Bq/kg, respectively. The average activity concentration of ^{40}K in fresh wild blueberry was 32 Bq/kg. With the obtained result the annual effective dose equivalent due to ingestion of blueberry-based products for adults was calculated, and for ^{137}Cs in blueberry-based juices, jams, sweets and fresh wild blueberry varied from 0.2 to 2.5 μSv, 2.8 μSv, 0.4 to 17.0 μSv and 2.5 μSv, respectively. The annual effective dose equivalent for ^{40}K in blueberry-based juices, jams, sweets and fresh wild blueberry varied from 1.0 to 16.0 μSv, 1.2 μSv, 5.0 to 66.0 μSv, and 9.3 μSv, respectively.*

Key words: *^{137}Cs, ^{40}K, blueberries, juices, jams, sweets*

1. INTRODUCTION

The 1986 nuclear accident in the Chernobyl power plant left a great impact on the world through a massive contamination of the environment by artificial radionuclides [1]. The total deposition of ^{137}Cs in Serbia after the accident was estimated to 5 kBq/m^2. Thirty years later, ^{137}Cs is still present in the natural ecosystems and it can be seen as a secondary source of contamination of plants and their fruits [2]. Blueberries, forest edible berries, are abundant in vitamins, minerals and other elements beneficial to humans. Blueberries have an extraordinary nutritional and pharmaceutical value and they are considered to be today's "super fruits" [3]. Forest blueberries are among the foods that contain post-Chernobyl ^{137}Cs. Ingestion of foods, beverages and water that contain radioactive elements such as ^{137}Cs are the main contributors to the internal exposure to radiation [4].

In the summer of 2017, Laboratory of the Department of Radiobiology with Radiation Hygiene, at the Faculty of Veterinary Medicine, University of Belgrade, received a sample of concentrated blueberry juice. In this sample, activity concentration of ^{137}Cs was 53 Bq/l. Activity concentrations for radionuclides are given in the Official Gazette of the Republic of Serbia (2018), and the permitted limit for ^{137}Cs in alcoholic and non-alcoholic beverages is 15 Bq/l. The investigated sample was not allowed on the market, and it raised a question of safety of blueberry-based products in Serbia.

The aim of this paper is to investigate the activity concentration of ^{137}Cs and naturally occurring ^{40}K, and to calculate the annual effective dose equivalent which an adult person can receive from ingestion of blueberry-based products on the market in Serbia.

2. MATERIALS AND METHODS

Activity concentrations of ^{137}Cs and ^{40}K were determined in blueberry-based products available on the market in Belgrade, the capital of the Republic of Serbia. Samples, 15 in total, were bought in stores during September 2017. The samples were weighed and then packaged– juices and one fresh wild blueberry in 1 l Marinelli beakers, two jams and two sweets in 450 ml Marinelli beakers.

A content of the investigated radionuclides in the samples was determined by gamma spectrometry on a High Purity Germanium detector (Ortec, USA) with a relative efficiency of 30 % and the energy resolution of 1.85 keV (1332.5 keV ^{60}Co). The detector efficiency calibration was performed for different geometries and

* vranjesb@vet.bg.ac.rs

different matrices, in accordance with the measured sample type.

We used commercially available standards with mixed radionuclides:

- ^{241}Am, ^{133}Ba, ^{109}Cd, ^{139}Ce, ^{57}Co, ^{60}Co, ^{137}Cs, ^{54}Mn, ^{113}Sn, ^{85}Sr, ^{88}Y, dispersed in silicone resin in Marinelli beaker, density (0.98±0.01) g/cm^3, volume 1 l; and

- ^{241}Am, ^{109}Cd, ^{139}Ce, ^{57}Co, ^{60}Co, ^{137}Cs, ^{113}Sn, ^{85}Sr, ^{88}Y, dispersed in silicone resin in Marinelli beaker, density (1.22±0.01) g/cm^3, volume 1 l.

- ^{241}Am, ^{109}Cd, ^{139}Ce, ^{57}Co, ^{60}Co, ^{137}Cs, ^{113}Sn, ^{85}Sr, ^{88}Y, dispersed in silicone resin in Marinelli beaker, density (0.98±0.01) g/cm^3, volume 450 ml.

The counting time for the samples, as well as for the background, was 60,000 s. The MDA value for radiocaesium is < 0.1 Bq/kg.

The activities of ^{40}K and ^{137}Cs were derived from 1460.8 keV and 661.66 keV gamma lines, respectively.

The analysis of the measured gamma spectra was performed by a software program GAMMA VISION® 32 (Ortec, USA).

The average annual effective dose equivalent that an individual receives due to the radionuclides ingestion by consumption of contaminated blueberry-based products was calculated using the following formula [5]:

$$E_{ingp} = C_{p,i} \cdot H_p \cdot DF_{ing} \qquad (1)$$

where: E_{ingp} is the annual effective dose from the consumption of nuclide i in foodstuff p (Sv/a), $C_{p,i}$ is the concentration of radionuclide i in foodstuff p at the time of consumption (Bq/kg), H_p is the consumption rate for foodstuff p (kg/a), DF_{ing} is the dose coefficient for the ingestion of radionuclide i (Sv/Bq), and its value for ^{137}Cs is 1.3×10^{-8} Sv/ Bq and for ^{40}K is 6.2×10^{-9} [6].

The calculated annual intake of juices, based on the data given in [7], was 0.128 l per day. Since no data for annual intake of jams and sweets are given in the bulletin report [7], calculations for average annual effective dose equivalent ware made using the weight of a standardized jam packing for hotels which is 0.028 kg, and assuming this as a minimal daily intake of jams and sweets.

3. Results and Discussion

Activity concentrations of ^{137}Cs and ^{40}K in the collected samples are shown in Table 1. Samples are grouped according to their blueberry percentage.

Potassium-40 was detected in all the collected samples. Its activity concentration was 3.5-55 Bq/kg for juices, 13.9-19.2 Bq/kg for jams, 17.2-227 Bq/kg for sweets and 32 Bq/kg for the fresh wild blueberry sample.

Article 12 of the Rulebook on limits of radionuclide content in drinking water, foodstuffs, feeding stuffs, medicines, general use products, construction materials and other goods that are put on market [5] states that the limits of the content of ^{137}Cs in non-alcoholic beverages and sweets is 15 Bq/l or 15 Bq/kg, and for fresh blueberries is 150 Bq/kg. Our results for the ^{137}Cs activity in juices ranged from minimal detectable activity (MDA) to 4.1 Bq/kg and in a fresh wild blueberry it was 4.1 Bq/kg, making these products safe for consumption as they did not exceed the given limit. For comparison, results presented by [8] for bottled fruit juices in Nigeria, also showed no presence of ^{137}Cs.

Table 1. Specific activities (Bq/kg) of ^{137}Cs and ^{40}K in blueberry-based samples

Sample	^{137}Cs (Bq/kg)	^{40}K (Bq/kg)
Juices		
Blueberry 25%	3.9 ± 0.1	17.8 ± 0.8
Blueberry 25.5%	< MDA	13.5 ± 0.5
Blueberry 26%	< MDA	24 ± 1
Blueberry 5%	< MDA	19.7 ± 0.7
Blueberry 12%	0.8 ± 0.1	10.1 ± 0.7
Blueberry 6.2%	0.4 ± 0.1	14.3 ± 0.6
Blueberry 0.5%	< MDA	3.5 ± 0.3
Blueberry 20%	0.9 ± 0.1	55 ± 1
Blueberry 3.9%	0.9 ± 0.1	44 ± 3
Blueberry syrup 100%	4.1 ± 0.1	7.1 ± 0.4
Jams		
Blueberry 55%	21 ± 1	19.2 ± 0.8
Blueberry 25.5%	< MDA	13.9 ± 0.8
Sweets		
Blueberry 100%	0.6 ± 0.1	17.2 ± 0.7
Blueberry dessert 7%	28 ± 1	227 ± 7
Fresh wild blueberry	4.1 ± 0.2	32 ± 1

The activity concentration of ^{137}Cs in one sweet was 28 Bq/kg which is 1.9 times higher than the allowed limit making this product unsuitable for consumption.

The two samples of jams showed different results: the measured activity concentration of caesium-137 was below the MDA, but the other sample showed the activity concentration of 21 Bq/kg. A particular issue here is that jams are classified under sweets so the investigated sample exceeds the limit given by the Rulebook.

Based on the obtained result for the activity concentrations of ^{137}Cs and ^{40}K, the average annual effective dose equivalent for adults was calculated and presented in Table 2.

The values calculated for the average annual effective dose equivalent due to ingestion of this blueberry-based products for both ^{137}Cs and ^{40}K, for an individual adult person were below the recommended level of 100 μSv (0.1 mSv) given in Article 5 of the Rulebook on limits of radionuclide content in drinking water, foodstuffs, feeding stuffs, medicines, general use products, construction materials and other goods that are put on market [5], making all of the products safe for consumption.

If we assume that all the measured ^{137}Cs activity concentration derives from the blueberries used for making the products, then we can calculate, using a simple proportion, the activity of ^{137}Cs in fresh blueberries. The comparison of the ^{137}Cs activity concentration in products and blueberries used for their production is given in Table 3. Products that are declared as 100% blueberry and the ones that have ^{137}Cs activity below MDA were excluded from this calculation.

Table 2. Annual effective dose equivalent (μSv) from the ingestion of ^{137}Cs and ^{40}K for an adult person who consumes 0.128 l of juice and 0.028 kg of jams and sweets

Sample	^{137}Cs (μSv)	^{40}K(μSv)
Juices		
Blueberry 25%	2.4	5.2
Blueberry 25.5%	/	3.9
Blueberry 26%	/	7.0
Blueberry 5%	/	5.7
Blueberry 12%	0.5	2.9
Blueberry 6.2%	0.2	4.1
Blueberry 0.5%	/	1.0
Blueberry 20%	0.9	16.0
Blueberry 3.9%	0.5	12.7
Blueberry syrup 100%	2.5	2.1
Jams		
Blueberry 55%	2.8	1.2
Sweets		
Blueberry 100%	0.4	5.0
Blueberry dessert 7%	17.0	66.0
Fresh wild blueberry	2.5	9.3

Table 3. Specific activities of ^{137}Cs in products and used blueberries

Sample	Measured activity of ^{137}Cs in products (Bq/kg)	Calculated activity of ^{137}Cs in fresh blueberries (Bq/kg)
Juices		
Blueberry 25%	3.9 ± 0.1	15.6
Blueberry 12%	0.8 ± 0.1	6.7
Blueberry 6.2%	0.4 ± 0.1	6.5
Blueberry 20%	0.9 ± 0.1	4.5
Blueberry 3.9%	0.9 ± 0.1	23.1
Jams		
Blueberry 55%	21 ± 1	38.2
Sweets		
Blueberry dessert 7%	28 ± 1	400

Calculated activities show that all the hypothetical blueberries used for making the sampled juices were safe for consumption. The activity concentration of ^{137}Cs in hypothetical blueberries used in producing the debatable jam is 38.2 Bq/kg making them safe for consumption. The importance of investigating the radioactivity modification during culinary preparation of forest berries has already been emphasized and a processing factor for cooking jam found to be 0.5 [9], which corresponds to our results. Hypothetical blueberries used in producing the sweet exceeded the given limit which would make them unsafe for consumption. The calculated activities concentration of ^{137}Cs in fresh blueberries is in good agreement with [4].

4. Conclusion

Gamma-spectrometric analysis of blueberry-based products can indicate whether contaminated blueberries are used in food processing, and give an insight into the ^{137}Cs burden of blueberries. It can also show how different methods of processing can reduce or increase the activity of ^{137}Cs in finished products.

Since jams are classified under sweets, their ^{137}Cs content limit is 15 Bq/kg. On the other hand, the limit for fresh blueberries is 10 times higher (150 Bq/kg). Knowing that the processing factor for cooking jam is 0.5 it can be concluded that the limits for fresh blueberries should not be over 30 Bq/kg. Calculated average annual effective dose equivalent showed that even the products that exceeded the activity concentration limit for ^{137}Cs are safe for consumption. In our opinion, it is important to introduce changes in the Rulebook, either to lower the limit for ^{137}Cs activity in fresh blueberries, or to categories jams differently. Furthermore, we would like to highlight the necessity to perform radioactivity analyses of different products to determine their safety for consumption.

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