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DETERMINATION OF RADIOCAESIUM IN BLUEBERRIES

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Abstract

In forest ecosystems, radionuclides are deposited in surface organic layers of the trees and other plants like grass, berries moss and lichen. From artificial radionuclides, ^{137}Cs is one of the most important fission product and it is of particular concern in the natural environment due to a long half–life ($T_{1/2}$ = 30 years), easy migration in the tropic chains and great bioavailability. Because of that, this artificial radionuclide is presents in environment especially in food samples, even 20 years after Chernobyl accident.

This paper presents the results of measurement the activity concentration of ^{137}Cs in blueberries (42 samples) for the period August 2011 to December 2012, which were imported in Serbia from different countries. Measurements were performed in Radiation and Environmental Protection Department in the Vinča Institute of Nuclear Sciences. Concentrations of ^{137}Cs were determined by gamma spectrometry using a HPGe detector.

The obtained results show that the activity concentration of ^{137}Cs in blueberries ranged from MDC (minimum detectable concentration) to 404 Bq kg^{-1}. Recommended level of activity concentration for ^{137}Cs in blueberries in Serbia is 150 Bq kg^{-1} (Official Gazette of the Republic of Serbia, 2011). Out of tested samples, 79 % of blueberries met the defined criteria of radiological safety.

Based on the obtained results for activity concentration of ^{137}Cs, the annual effective dose due to ingestion of blueberries for adults was calculated.

Keywords: Radiocaesium, blueberries, gamma spectrometry, annual effective dose.

Introduction

Radiation and Environmental Protection Department in the Vinča Institute of Nuclear Sciences performs regular control of radioactivity of imported goods (foodstuffs, fertilizers, building materials, items of general use, etc). In Serbia, based on the regulation (Official Gazette of the Republic of Serbia, 2011) limit values of activity concentrations of certain radionuclides are defined for different materials (drinking water, foodstuffs, feeding stuffs, drugs, items of general use, building materials and other goods to be placed on the market). Among others, for imported blueberries and other berries which are subject to control, limit value for the activity concentration of ^{137}Cs is 150 Bq kg^{-1}.

Blueberries are among the most popular fruits for home and market gardening. Today, blueberries are grown commercially also in South America, Australia, New Zealand, Asia, South Africa and Europe (Strik, 2005; Wach, 2008). Almost 10% of the world total blueberries cultivated area is located in Europe where Poland and Germany are more important producers (Delian et al., 2010). Blueberries require acid soils, and a soil pH between 4.5 and 5.2 is ideal. However, blueberries can be productive on places where the pH is as high as 6.0 (Strik, 2006). Adequate soil for the growth of blueberry is sour and moderately moist. Blueberries are growing in the area of deciduous and beech forests.

^{137}Cs, which can be found in blueberries, is a fission product of concern due to its long physical half–life of 30.2 years and its high bioavailability. ^{137}Cs is water–soluble and can rapidly enter the biological cycles and accumulate in terrestrial ecosystems, behaving in a very similar way to potassium. As a result, it is mainly deposited in soil and vegetation (Bourcier et al., 2010).

The atmospheric nuclear weapon tests carried out in the 1950's and 1960's and up to 1980 led to a global contamination with fallout of radionuclides, and in particular of ^{137}Cs, especially in the northern hemisphere. Additional contamination in Europe occurred after the Chernobyl accident in 1986 (Bourcier et al., 2010). ^{137}Cs is produced during fission of nuclear fuel, as a result of nuclear weapons testing, operation of nuclear reactors, reprocessing of spent fuel and reactor accidents. This causes radionuclide release into the environment which can enter the human food chain. An additional small risk does exist from production of radioisotope sources for medical and industrial uses (Carpenter et al., 1995).

Forests are effective trapping systems for fallout radionuclides, which persist in the forest ecosystem for much longer than in cultivated agricultural land. In general, the consumption of forest products may contribute to the increase the internal human dose, which is caused by intake of radionuclides via ingestion. For example, blueberries and mushrooms are dominant and they are commonly consumed (Vaaramaa et al., 2009).

After deposition of fallout and interception by the above–ground parts of the plant, radionuclides in soluble form can be retained, absorbed and translocated to fruits as well as to other parts of the plant. Such processes are important not only for short–term releases, but also play a role during the following years in perennials, especially (re–)translocation within the plant, but also along with the process of resuspension. Contamination of fruits following wet or dry deposition can be directly on the exposed fruit surface, or initially on any of the exposed plant surfaces followed by absorption and translocation to fruit (Carini and Bengtsson, 2001).

One of the factors in the assessment of risk to human health, is the risk of ionizing radiation which can be monitored via the annual effective dose. In assessing the annual effective dose which the population received, one of the elements is the dose which is received via ingestion.

The aim of this study was to calculate the annual effective dose which the population can received due to ingestion of 150 g fruits per day (IAEA, 1999), if we assume that all of ingested fruit are blueberries.

Materials and methods

The samples of blueberries imported from different countries were investigated. The preparation of samples included weighing and placing into the 500 cm^3 Marinelli beakers (IAEA, 1989). Since the purpose of measurement is screening, the counting of the samples was conducted immediately after preparation, without drying. The total of 42 samples of blueberries for the period of 17 months were analyzed.

Samples were measured using a high purity germanium detector (HPGe) with relative efficiencies of 18 % and 20 % and energy resolution of 1.8 keV for the 1332 keV ^{60}Co peak. Calibration of detectors for measurement of food samples was performed using of silicone resin matrix in geometry of the plastic Marinelli beaker of 500 cm^3, (Czech Metrological Institute, Praha, 9031−OL−208/08, type ERX) spiked with a series of radionuclides (^{241}Am, ^{109}Cd, ^{139}Ce, ^{57}Co, ^{60}Co, ^{203}Hg, ^{88}Y, ^{113}Sn, ^{85}Sr i ^{137}Cs) with total activity of 40.624 kBq on the day April 15, 2008.

Counting time interval was 3600 s. The spectra were analyzed using the program GENIE 2000. The activity of ^{137}Cs was determined from its 661 keV γ−energy.

The accuracy and reproducibility of gamma spectrometry systems were verified on a periodic basis – every week. Total background count rate without a source is monitored to verify that the detector and shield have not been contaminated by radioactive materials. Energy calibration is checked in whole region before applying usual quality control (QC) procedure for gamma spectrometry measurement. The total activity of calibration source is used to check the efficiency calibration and the general operating parameters of the gamma spectrometry system (source positioning, contamination, library values, and energy calibration). The detector – shield background, detector efficiency, peak shape, and peak drift are measured and verified if they are within the warning and acceptance limits. For that purpose ^{60}Co and ^{133}Ba sources were used.

The activity concentration of ^{137}Cs in the samples was calculated using the equation:

$$A = \frac{N}{t \times P_{\gamma} \times E_f \times m} \qquad (1)$$

where N is count of the sample corrected on background, t – counting time (s), P_{γ} – probability of gamma decay (%), E_f – full energy peak efficiency (%), m – the mass (kg) of the sample.

Minimum detectable concentration (MDC) was calculated by the equation (2):

$$MDC = \frac{LLD}{t \times P_{\gamma} \times E_f \times m} \qquad (2)$$

where LLD is the detection limit, $LLD = 2.71 + 4.65\sqrt{B}$, where B is count of the background. The combined measurement uncertainty of the results was calculated at the 95% level of confidence (k = 2).

A possible risk of radioactivity for human health is expressed by the individual annual effective dose due to ingestion (E_{ing}). A contribution to the individual annual effective dose to an adult from blueberry consumption may be calculated, according to (IAEA, 2001) using equation (3):

$$E_{ing} = H \times A \times DF_{ing} \qquad (3)$$

where E_{ing} is the individual annual effective dose due to ingestion (Sv), H – the annual intake of blueberries (kg per person) which in our case is 54.75 kg, A – the activity concentration of ^{137}Cs (Bq kg^{-1}) and DF_{ing} – the dose conversion factor for ingestion of ^{137}Cs defined as the dose received by an adult per unit intake of radioactivity and its value is 1.3×10^{-8} Sv Bq^{-1} (IAEA 1999, Kalač 2001).

Results and discussion

The results of measurements of ^{137}Cs in blueberries, which were imported in Serbia from different countries are presented in *Table 1*. The activity concentration of ^{137}Cs in Bq kg^{-1} ranged between MDC and 404. Recommended level of activity concentartion for ^{137}Cs in Serbia is 150 Bq kg^{-1} (Official Gazette of the Republic of Serbia, 2011). Based on the results presented in *Table 1*, it can be seen that out of measured samples, 79 % of blueberries met the defined criteria of radiological safety, while 21 % of the samples do not met the criteria. As can be seen from *Table 1*, all the samples which do not met the criteria for radiological safety were imported from Ukraine.

Also, based on the obtained results for the activity concentration of ^{137}Cs, the annual effective dose for adults was calculated (assuming that adults eat 150 g of fruit per day (IAEA, 1999), in this case blueberries) and presented in *Table 1*.The annual effective dose values due to the ingestion of ^{137}Cs from blueberries were ranged from 0.9 to 287.5 μSv. The values of the annual effective dose due to ingestion blueberries for all samples that met the criteria of radiological safety are below the recommended reference level of 100 μSv (Official Gazette of the Republic of Serbia, 2011), so they are not dangerous to human health.

Table 1. The activity concentration of ^{137}Cs in blueberries and the annual effective dose for age group (> 17).

№	**Country of import**	**^{137}Cs (Bq kg^{-1})**	**Annual effective dose (μSv)**
1.	Republic of Macedonia	< 1	/
2.	Montenegro	< 1	/
3.	Republic of Macedonia	< 1	/
4.	Republic of Macedonia	< 2	/
5.	Republic of Macedonia	< 2	/

6.	Russia	3.8 ± 1.8	2.7
7.	Republic of Macedonia	3.5 ± 1.3	2.5
8.	Bosnia and Herzegovina	< 2	/
9.	Republic of Macedonia	< 2	/
10.	Ukraine	112 ± 12	79.7
11.	Ukraine	83 ± 9	59.1
12.	Russia	< 1.3	/
13.	Ukraine	276 ± 25	196.4
14.	Ukraine	46 ± 6	32.7
15.	Ukraine	60 ± 7	42.7
16.	Republic of Macedonia	1.3 ± 0.2	0.9
17.	Ukraine	121 ± 12	86.1
18.	Ukraine	71 ± 8	50.5
19.	Ukraine	160 ± 15	113.9
20.	Republic of Macedonia	< 1.4	/
21.	Russia	3.3 ± 1.0	2.3
22.	Republic of Macedonia	< 1.5	/
23.	Ukraine	5 ± 2	3.6
24.	Russia	2.1 ± 1.0	1.5
25.	Republic of Macedonia	< 1	/
26.	Montenegro	< 1	/
27.	Republic of Macedonia	2.0 ± 0.7	1.4
28.	Montenegro	7.1 ± 2.0	5.1
29.	Ukraine	206 ± 19	146.6
30.	Ukraine	20 ± 3	14.2
31.	Ukraine	93 ± 10	66.2
32.	Republic of Macedonia	< 1	/
33.	Ukraine	266 ± 24	189.3
34.	Ukraine	325 ± 30	231.3
35.	Montenegro	2.4 ± 1.1	1.7
36.	Ukraine	71 ± 7	50.5
37.	Ukraine	63 ± 7	44.8
38.	Ukraine	404 ± 36	287.5
39.	Ukraine	395 ± 34	281.1
40.	Ukraine	230 ± 21	163.7
41.	Ukraine	264 ± 23	187.9
42.	Montenegro	5 ± 2	3.6

Conclusion

42 samples of blueberries imported from different countries in Serbia, were investigated in order to determine the activity concentration of ^{137}Cs in them. The obtained results for the activity concentration of ^{137}Cs ranged between MDC and 404 Bq kg^{-1} and the obtained values for the annual effective dose ranged between 0.9−287.5 μSv.

Considering the fact that 21 % samples of blueberries expressed higher values of the activity concentration of ^{137}Cs, as well as the annual effective dose than the recommended limit value, there is a need for regular control blueberries which are imported.

All samples which do not met the criteria for radiological safety were imported from Ukraine and apparently this is consequence of the nuclear accident at Chernobyl.

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