A TWENTY-YEAR RADIOCONTAMINATION STUDY OF EDIBLE FUNGI OF SERBIA
Activity Levels of ^{137}Cs and Estimated Dose to the Population

by

Ana A. ČUČULOVIĆ [1*], ***Jelena N. STANOJKOVIĆ*** [1], ***and Dragan S. VESELINOVIĆ*** [2]

[1] INEP – Institute for the Application of Nuclear Energy, University of Belgrade, Belgrade, Serbia
[2] Faculty of Physical Chemistry, University of Belgrade, Belgrade, Serbia

Scientific paper
https://doi.org/10.2298/NTRP2002165C

Penny bun (*Boletus edulis*) and chanterelle (*Cantharellus cibarius*) are fungi used for human consumption in several forms: fresh, thermally treated, or conserved (salted, marinated, pasteurized, dried). Random samples were collected on the territory of the Republic of Serbia in the period 1999-2018 intended for export to the EU. The presence of ^{137}Cs was noted in each individual fungi sample. The higher activity level of ^{137}Cs in fresh penny bun samples was noted in 2001 (25.9 Bqkg^{-1}), while in fresh chanterelle samples it was noted in 2000 (18.6 Bqkg^{-1}). Marinating the fungi reduced activity levels of ^{137}Cs in relation to the fresh fungi activity, for penny bun from 1.6 to 12.6 times for a year, while for chanterelle from 0.8 to 14.7 times for a year. Drying the fungi increases activity levels of ^{137}Cs: for penny bun from 0.04 to 0.29 times for a year, while for chanterelle from 0.03 to 1.94 times for a year. The values of annual effective radiation doses were much lower than the recommended values for the population (1 mSv).

Key words: Boletus edulis, Cantharellus cibarius, ^{137}Cs, annual effective dose

INTRODUCTION

Fungi are the most widespread organisms on Earth according to the number of species and represent a special domain, the eukaryote domain [1-3]. Fungi share plant hormones with plants, while with animals they share the chitin cell wall, melanin pigment, and enzymes present in mitochondria [4]. Fungi are tolerant of distinctly low or high temperatures, drying, and antifungal substances. Their growth is influenced by many external factors: availability of organic matter, the concentration of hydrogen ions (pH value), temperature, and humidity. The chemical composition of fungi is complex, it varies and depends on many factors: soil and climatic conditions, a season of growth, and maturation. The content of organic compounds and mineral salts in fungi directly influences their nutritive and biological value [5-8]. Fungi have nutritive value as they contain: vitamins, amino acids, proteins, fats, mineral, and aromatic substances, carbohydrates, lipids, organic acids, and enzymes. Fresh edible fungi contain up to 95 % water, while the remaining is dry matter. Proteins are the most significant component of fungi participating with 1-4 % in fresh, and 10-45 % in dry fungi.

In the sixties of the last century research showed that fungi are good bioindicators and bio monitors of pollution of the environment with polluting substances (heavy metals, radionuclides, pesticides) and responsible for their retention in organic layers of woodland soil. Fungi have an essential role in the decomposition of organic matter. Research in the period before and after the Chernobyl accident (26.4.1986, Ukraine) proved that they are good bioindicators of radio pollution [9-13].

According to UNSCEAR data, the radioactive cloud that formed after the Chernobyl accident included the territory of SFRY in two waves, and pollution of the territory with radionuclides was not homogenous. Levels of total contamination by Chernobyl radioactive fallout in Yugoslavia were 0.08-6.40 GyL^{-1}, Eastern Serbia 1.41-2.56 GyL^{-1}, South Serbia 0.45-1.40 GyL^{-1}, and Central Serbia 0.08-0.44 GyL^{-1}. It was estimated that during 1986 about 2.4 % of the totally released radionuclides (without inert gases) were deposited on the territory of Yugoslavia: 1.3 10^{18} Bq ^{131}I, 3.0 10^{17} Bq ^{133}I, 8.9 10^{16} Bq ^{137}Cs and 2.0 10^{16} Bq ^{134}Cs [14].

Dry or wet fallout is responsible for ^{137}Cs one of the potentially most dangerous radionuclides reaching

* Corresponding author; e-mail: anas@inep.co.rs

organisms. It is absorbed by ionic exchange, physical and chemical sorption from the environment. The physical half-life ^{137}Cs is long (30.2 years), while its physicochemical characteristics are such that it actively participates in the human and animal food chain through plants. It bonds with tissue, cells, and molecules like a protein replacing the natural element potassium. It is the chemical and biochemical homologue of potassium and follows its metabolism in organisms. No organ is critical for ^{137}Cs. It is an organotropic radionuclide, completely soluble in body fluids, which distributes uniformly in the organism [15]. Fungi accumulate ^{137}Cs from the substrate on which they grow, instead of potassium, that besides phosphorous and carbon is one of the most significant elements for fungi. They do not have the capability of differentiating between chemically close elements and deposit them on different parts in the body [15,16]. Absorption of ^{137}Cs by fungi is influenced by many factors (species, age, fungi nutrition way, mycelium depth, atmospheric fallout, climate, soil characteristics, altitude, application of artificial fertilizers, physicochemical composition, and distribution and availability of radionuclides in the soil) [17].

Immediately after the Chernobyl accident, high levels of ^{137}Cs from 6680 Bqkg^{-1} (Finland) to 131000 Bqkg^{-1} (Russia) were measured [18-21]. Research has shown that different fungi species absorb ^{137}Cs differently [22]. The *Boletu*s species show a great affinity for ^{137}Cs, while the *Cantharellus* species accumulate moderate levels of ^{137}Cs [5, 23]. Different fungi species were polluted by radiocesium on the territory of SFRY in a different way, thus black morel (*Morchella conica*) and black trumpet (*Craterellus cornucopioides*) were polluted more with ^{137}Cs than penny bun and chanterelle [24]. Research has shown that activity levels of ^{137}Cs in fungi decrease with time [25-27].

On average 3000 tons of fungi are collected yearly on the territory of the Republic of Serbia and about 60 % are sold to countries in the European Union [28-31]. According to EU regulations, the activity levels of ^{137}Cs for fungi to be exported to EU countries need to be lower than 600 Bqkg^{-1} [32]. Due to refined tastes, nutritional and medicinal values, fungi consumption in many countries in Central and Eastern Europe is increasing [33-35]. Fungi for human consumption can be used in several forms: fresh, thermally treated, or conserved (salted, marinated, pasteurized, dried), and as such, they are exported from Serbia. The annual average of fresh fungi consumption per inhabitant varies in different countries ranges from negligible amounts to more than 58.4 kg per year [36]. Data on the annual average fungi consumption per capita does not exist in Serbia. In Serbia penny bun (*Boletus edulis*) and chanterelle (*Cantharellus cibarius*) are collected the most and are considered of the best quality for human consumption. Penny bun grows in deciduous and conifer woods, but also on forest meadows from May to October. Chanterelle grows in dark deciduous and conifer woods from June to October. *Boletus edulis* and *Cantharellus cibarius* are the best known and most indicated fungi for radiometric controls of soil and environment [5].

Due to their ability to absorb ^{137}Cs from the soil and concentrate it in their body [36-38], fungi are unavoidable factors of radioecological investigation of the ecosystem. Monitoring of the environment using fungi is significant for the evaluation of the radiation load of the population of a certain area. High activity levels of ^{137}Cs measured in edible fungi species indicate a potential direct danger for people consuming them. This danger can grow indirectly through consuming venison and game that use fungi in their nutrition [36].

The purpose of this paper is to present activity levels of ^{137}Cs in fungi collected on the territory of Serbia in the period 1999-2018 in order to evaluate if they represent a danger to human health when observed as a complete product consumed by the population.

MATERIALS AND METHODS

In the period from 1999 to 2018 in the Institute for the Application of Nuclear Energy – INEP samples of 2287 penny bun and chanterelle were analysed in total, as follows: 457 of fresh, 222 of frozen, 272 of marinated and 537 of dry penny bun, and also 530 of fresh, 14 of frozen, 140 of marinated and 115 of dry chanterelle. Samples were collected on the territory of Serbia randomly and were intended for export. Activity levels of ^{137}Cs in samples were determined using the standard gamma spectroscopic method. Samples were cleaned from visible dirt (soil, grass), homogenized, weighed, and measured in Marinelli vessels of 0.5 and 1.0 dm^3 volume. The samples were analysed using the standard gamma spectroscopic method [39]. Calibration of the detector for Marinelli geometry with 0.5 and 1.0 dm^3 volume containers, was performed using the calibration standard in the same geometry, containing a mixture of radionuclides. Energy calibration and efficiency calibration of the gamma-spectrometer were performed before measurements using a calibration source containing a mixture of radionuclides with known activity. Detector calibration was performed using three different radioactive reference materials in the Marinelli geometry: Silicone resin (Czech Metrological Institute Praha (CMI Praha), Cert. No. 1035-SE-40517-17, Type MBSS 2 (^{241}Am, ^{109}Cd, ^{139}Ce, ^{57}Co, ^{60}Co, ^{137}Cs, ^{113}Sn, ^{85}Sr, ^{88}Y, ^{51}Cr), 490.0 g, density: 0.98   0.01 gcm^3, vol. 500.0   5.0 cm^3 ref. date 1.9.2017); silicone resin CMI Praha, Cert. No. 1035-SE-40661/14, Type MBSS 2 (^{241}Am, ^{109}Cd, ^{139}Ce, ^{57}Co, ^{60}Co, ^{137}Cs, ^{113}Sn, ^{85}Sr, ^{88}Y, ^{51}Cr), 490.0 g, density: 0.98   0.01 g/cm^3, vol. 500.0   5.0 cm^3, ref. date 1.10.2014; silicone resin CMI Praha, Cert. No. 9031-OL-420/10, Type MBSS 2 (^{241}Am, ^{109}Cd, ^{139}Ce, ^{57}Co, ^{60}Co, ^{137}Cs, ^{113}Sn, ^{85}Sr, ^{88}Y, ^{203}Hg), 985.0g, density: 0.985 0.01 gcm^{-3}, vol. 1000.0 10.0 cm^3, ref. date 16.8.2010; and Vegetation (Inst. Radiological Protection, Belgrade: QAP 9709, 23.12.2002).

The measurement time of one sample was 3600-60000 seconds. The relative error of sample preparation and measurement was up to 10 %. The activity of ^{137}Cs was determined using the -line at the energy of 661.6 keV-a. Gamma spectroscopic measurements were until 2002 performed on a scintillation NaI detector ORTEC-CANBERRA efficiency 8.7 % and resolution 6.7 % for ^{137}Cs at 661.6 keV, and from 2002 on an HPGe ORTEC/Ametek detector with 8192 channels, resolution 1.65 keV and relative efficiency of 34 % at 1.33 MeV for ^{60}Co. The spectra were analyzed using the Gamma Vision 32 software package [40].

The annual effective dose, D (Sv), for the population due to ingestion from consuming fresh fungi is evaluated based on activity levels of ^{137}Cs, $A_{^{137}\mathrm{Cs}}$ (Bqkg^{-1} fresh mass), coefficient of dose conversion, DC (SvBq^{-1}), and annually amount of consumed fresh fungi, m (kg)

$$D \quad A_{^{137}\mathrm{Cs}} \quad DC \quad m \qquad (1)$$

The radiation dose conversion coefficient includes calculations that contain aspects of human physiology, radiation physics, time, and spatial deposition of absorbed energy formed due to consuming products contaminated with radionuclides and for ^{137}Cs it is 1.3 10^{-8} SvBq^{-1} [41, 42].

RESULTS AND DISCUSSION

The average yearly activity levels of ^{137}Cs are shown in tabs. 1 and 2 for penny bun and chanterelle samples, fresh, frozen and marinated collected in the period 1999-2018 in Serbia, including standard deviation, minimal and maximal levels of ^{137}Cs (Bqkg^{-1}), and the number of samples.

The presence of ^{137}Cs was noted in all investigated samples of penny bun and chanterelle. Analysis of the data shown in tabs. 1 and 2 enabled the conclusion that the average activity levels of ^{137}Cs in the period from 1999-2018 in fresh penny bun and chanterelle were: 3.6-25.9 Bqkg^{-1}, *i. e.* 3.0-18.6 Bqkg^{-1}. The average activity levels of ^{137}Cs (Bqkg^{-1}) in penny bun were: 4.1-16.9 (frozen), 0.9-7.4 (marinated), and 45.9-103 (dry), while in chanterelle they were: 7.0-35.0 (frozen), 0.6-15.4 (marinated), and 41.6-312 (dry). Considering

Table 1. Average activity levels of ^{137}Cs standard deviation (sd), minimal (min), and maximal (max) values of ^{137}Cs [Bqkg^{-1}], number of samples (N.s.) of penny bun: fresh, frozen, marinated, and dry collected on the territory of Serbia in the period 1999-2018

Year	Penny bun							
	Fresh		Frozen		Marinated		Dried	
	Average activity sd [Bqkg^{-1}] (N.s.)	Min-max [Bqkg^{-1}]	Average activity sd [Bqkg^{-1}] (N.s.)	Min-max [Bqkg^{-1}]	Average activity sd [Bqkg^{-1}] (N.s.)	Min-max [Bqkg^{-1}]	Average activity sd [Bqkg^{-1}] (N.s.)	Min-max [Bqkg^{-1}]
1999.	20.5 17.4 (42)	3.0-69.0	16.9 9.8 (11)	5.0-38.0	4.8 2.7 (8)	2.0-10.0	100 93.7 (3)	20.0-503
2000.	12.2 9.2 (55)	5.0-47.0	13.8 7.0 (15)	5.0-23.0	5.2 1.9 (24)	3.0-9.0	96.1 85.7 (55)	15.0-318
2001.	25.9 26.2 (53)	5.0-116	15.8 14.2 (19)	5.0-62.0	7.4 5.9 (25)	3.0-22.0	88.8 86.2 (43)	15.0-298
2002.	11.8 11.8 (52)	3.0-53.0	12.6 8.2 (29)	2.0-30.0	5.9 3.4 (31)	1.0-16.0	91.7 83.3 (50)	10.0-297
2003.	18.5 21.7 (36)	2.0-117	12.1 7.3 (19)	2.0-27.0	4.5 2.9 (16)	1.0-10.0	63.7 35.5 (43)	4.0-150
2004.	14.7 15.6 (32)	2.0-70.0	8.8 2.5 (10)	6.0-15.0	3.3 3.8 (9)	1.0-13.0	103 148 (33)	10.0-877
2005.	17.0 14.5 (26)	2.0-71.0	11.8 6.9 (12)	2.0-22.0	4.2 2.4 (5)	2.0-8.0	70.9 52.1 (27)	11.0-265
2006.	11.5 13.2 (22)	2.0-43.0	11.7 7.6 (7)	5.0-24.0	1.7 0.5 (12)	0.6-2.0	87.0 47.4 (29)	6.0-189
2007.	7.0 9.6 (25)	0.8-51.0	10.0 7.7 (7)	2.3-22.0	1.7 1.0 (18)	0.5-5.0	70.0 54.1 (27)	11.0-245
2008.	11.3 12.6 (27)	1.3-50.0	11.8 9.0 (4)	3.7-23.0	0.9 0.6 (10)	0.3-2.1	59.6 30.3 (17)	11.0-127
2009.	12.4 12.5 (14)	1.5-42.0	7.9 11.5 (8)	0.2-32.0	2.3 2.4 (7)	0.4-6.7	79.4 52.6 (24)	12.0-242
2010.	12.5 8.1 (13)	1.5-26.0	7.9 7.9 (16)	2.5-27.0	2.7 3.2 (25)	0.2-11.0	56.9 47.5 (22)	4.6-240
2011.	7.7 4.5 (5)	1.3-14.0	4.7 4.2 (7)	1.0-12.0	3.5 3.6 (13)	0.4-9.2	50.4 42.5 (21)	11.0-165
2012.	5.3 2.0 (8)	3.9-9.0	11.9 8.2 (12)	4.9-29.0	1.9 2.5 (8)	0.3-7.6	45.9 23.7 (20)	11.0-93.0
2013.	3.6 2.3 (7)	0.5-7.5	12.0 9.8 (7)	2.0-24.0	2.2 2.4 (14)	0.2-6.1	87.5 112 (18)	7.5-383
2014.	3.6 2.8 (5)	0.4-5.9	12.9 10.9 (7)	2.5-31.0	1.0 1.0 (15)	0.2-4.0	48.3 30.8 (16)	9.0-103
2015.	5.3 7.9 (7)	1.0-23.0	4.1 2.5 (5)	1.8-8.2	1.2 1.6 (7)	0.2-4.3	56.4 36.0 (21)	15.0-145
2016.	7.3 5.4 (11)	0.3-18.0	10.0 8.1 (11)	0.8-28.0	1.7 2.1 (11)	0.2-7.3	52.9 44.0 (33)	3.5-212
2017.	5.0 2.0 (6)	4.0-8.0	8.8 7.0 (7)	1.0-18.0	1.4 1.1 (8)	0.4-3.5	63.1 49.4 (13)	20.0-190
2018.	4.7 3.4 (11)	1.5-12.0	2.2 1.6 (9)	0.2-5.3	0.5 0.2 (6)	0.2-0.7	45.7 52.0 (22)	12.0-256
Total N.s.	457		222		272		537	

Table 2. Average activity levels of ^{137}Cs, standard deviation (sd), minimal (min) and maximal (max) values of ^{137}Cs [Bqkg^{-1}], number of samples (N.s.) of chantarelle: fresh, frozen, marinated, and dry collected on the territory of Serbia in the period 1999-2018

	Chantarelle							
Year	Fresh		Frozen		Marinated		Dried	
	Average activity sd [Bqkg^{-1}] (N.s.)	Min-max [Bqkg^{-1}]	Average activity sd [Bqkg^{-1}] (N.s.)	Min-max [Bqkg^{-1}]	Average activity sd [Bqkg^{-1}] (N.s.)	Min-max [Bqkg^{-1}]	Average activity sd [Bqkg^{-1}] (N.s.)	Min-max [Bqkg^{-1}]
1999.	13.6 12.7 (18)	5.0-55.0	7.0 (1)	---	3.1 0.6 (10)	2.0-4.0	122 62.5 (5)	56.0-197
2000.	18.6 14.6 (55)	6.0-90.0	---	---	6.2 3.9 (21)	2.0-15.0	101 71.1 (6)	54.0-204
2001.	11.8 9.7 (51)	5.0-69.0	16.7 2.9 (3)	15.0-20.0	15.4 19.6 (17)	4.0-56.0	312 148 (6)	174-489
2002.	16.4 20.4 (73)	4.0-96.0	---	---	5.6 1.4 (44)	4.0-9.0	98.0 (1)	---
2003.	14.4 15.2 (52)	2.0-80.0	10.0 (1)	---	4.7 0.8 (6)	4.0-6.0	198 (1)	---
2004.	8.9 13.5 (41)	1.0-85.0	32.0 (1)	---	3.3 1.5 (4)	2.0-5.0	90.3 54.8 (4)	50.0-166
2005.	8.7 10.3 (31)	2.0-42.0	15.0 0 (2)	15.0	2.0 (1)	---	280 37.1 (7)	208-315
2006.	10.6 12.3 (34)	2.0-67.0	35.0 (1)	---	3.4 3.5 (9)	1.0-10.0	88.9 9.8 (5)	81.0-102
2007.	10.2 8.5 (34)	1.4-43.0	27.0 0 (2)	27.0	2.8 2.6 (5)	1.2-7.5	91.9 39.2 (9)	46.0-167
2008.	8.8 8.2 (17)	0.6-23.0	---	---	0.6 (1)	---	144 177 (5)	55.0-459
2009.	8.1 6.1 (24)	1.2-22.0	---	---	2.9 0.9 (5)	1.7-4.1	69.4 34.7 (5)	43.0-130
2010.	13.0 18.7 (24)	1.4-26.0	---	---	1.0 0 (2)	1.0	54.6 32.6 (5)	18.0-97.0
2011.	9.2 7.1 (15)	2.0-27.0	---	---	0.8 (1)	---	41.6 13.2 (7)	25.0-57.0
2012.	4.1 2.0 (3)	2.4-6.3	---	---	---	---	81.8 41.2 (4)	43.0-131
2013.	4.4 2.6 (4)	2.0-8.0	---	---	---	---	76.0 (1)	---
2014.	4.0 3.6 (8)	0.6-11.0	---	---	---	---	74.0 42.6 (7)	16.0-124
2015.	8.3 6.4 (9)	0.5-20.0	---	---	1.0 0.9 (3)	0.4-2.0	74.3 51.8 (8)	40.0-190
2016.	3.0 6.0 (9)	0.8-6.0	32.0 (1)	---	2.1 2.4 (3)	0.6-4.9	94.2 118 (15)	0.6-465
2017.	4.9 4.0 (18)	0.7-18.0	---	---	2.7 2.7 (4)	0.3-6.5	191 188 (6)	28.0-487
2018.	2.6 2.4 (10)	0.7-8.0	4.7 4.7 (2)	1.4-8.0	1.6 1.5 (4)	0.4-3.7	65.0 28.2 (8)	26.0-119
Total N.s.	530		14		140		115	

that in each individual sample ^{137}Cs originating from Chernobyl was noted, this means that pollution of ^{137}Cs originating from Chernobyl has expanded to the whole territory of Serbia.

Analysis of the data shown in tabs. 1 and 2 shows that the higher average activity level of ^{137}Cs in samples of fresh penny bun was noted in 2001 (25.9 Bqkg^{-1}), while in samples of fresh chanterelle in 2000 (18.6 Bqkg^{-1}). The higher activity level of ^{137}Cs in fresh penny bun was measured in 2003 (117 Bqkg^{-1}), while in fresh chanterelle in 2002 (96.0 Bqkg^{-1}).

Penny bun and chanterelle are exported from Serbia frozen. Analysis of the data number of samples (N.s.), presented in tabs. 1 and 2, shows that chanterelle is exported in a much smaller amount than penny bun. The higher average activity level of ^{137}Cs in samples of frozen penny bun was noted in 1999 (16.9 Bqkg^{-1}). The higher activity level of ^{137}Cs in frozen penny bun was measured in 2001 (62.0 Bqkg^{-1}). Freezing fungi, penny bun, and chanterelle resulted in a difference in average activity levels between frozen and fresh fungi from 0.28 to 2.14 times, *i. e.* from 0.09 to 1.94 times.

Fungi are most often consumed marinated (cleaned and washed fungi are blanched, strained, and covered with a marinating solution containing 12-18 % salt). Analysis of data presented in tabs. 1 and 2 shows that the higher activity levels of ^{137}Cs were noted in 2001 in marinated fungi, in penny bun the average value was 7.4 Bqkg^{-1} (3.0-22.0 Bqkg^{-1}) and in chanterelle 15.4 Bqkg^{-1} (4.0-56.0 Bqkg^{-1}). These results indicate that marinating fungi reduced activity levels of ^{137}Cs in comparison with the values determined for fresh fungi, for penny bun from 1.6 to 12.6 times for a year, while for chanterelle from 0.8 to 14.7 times for a year, as a consequence of ^{137}Cs extraction by the marinating solution. These results agree with the research results of other authors [36, 43].

Drying is the oldest and simplest method for conserving fungi, by removing water from them. The dry to fresh mass ratio is 0.08 [44]. Analysis of the data given in tabs. 1 and 2 shows that a higher average activity level of ^{137}Cs in samples of dried penny bun was noted in 2004-103 Bqkg^{-1} (10-877 Bqkg^{-1}), while for chanterelle it was noted in 2001-312 Bqkg^{-1} (174-489 Bqkg^{-1}). Higher levels of activity of ^{137}Cs in dried samples of both fungi types compared to other samples are due to a loss of water, *i. e.* a calculation per mass unit, that leads to an increase in relation to fresh, non-hydrated fungi, for penny bun from 0.04 to 0.29 times for a year, while for chanterelle it was from 0.03 to 1.94 times for a year. Activity levels of ^{137}Cs increase due to the drying process applied to the fungi.

Different preparation procedures of dry fungi for human consumption can reduce the activity levels of ^{137}Cs in them [36, 44].

Analysis of the results presented in tabs. 1 and 2 shows that maximal activity levels of ^{137}Cs in investigated samples of penny bun and chanterelle were lower than 600 Bqkg^{-1}, except in dry fungi collected in 2004, so they could be exported from Serbia to countries in the EU [32].

The above data show that activity levels are reduced in time as a result of radioactive decay of ^{137}Cs on one hand, and sorption-desorption processes on the other. Besides, it leads to the reduction of ^{137}Cs content in fungi. Sorption-desorption processes occur between the fungi and the soil on which it grows, *i. e*. water from fallout and water from the soil. The presented results correspond to the investigation results of other authors [28].

Based on data on activity levels of ^{137}Cs in fresh fungi (penny bun and chanterelle) shown in tabs. 1 and 2, eq. (1) can be used to calculate the annual effective dose, D (μSv), ingested by a person consuming 1 kg of fresh fungi. The annual effective dose, D (μSv), ingested by a person consuming 1 kg of penny bun or chanterelle is shown in tab. 3, including annual effective doses for minimal, D_{min}, and maximal values D_{max}, of activity levels of ^{137}Cs in fungi. From tab. 3 it follows that in 2001 the annual effective dose was the highest if the population consumed 1 kg of fresh penny bun (0.337 μSv), *i. e*. in 2000 if the population consumed 1 kg of fresh chanterelle (0.242 μSv). D_{max} was noted in 2003 for a population consuming 1 kg of penny bun (1.521 μSv), and in 2002 for a population consuming 1 kg of chanterelle (1.248 μSv). Due to the reduction in the ^{137}Cs content in fungi with time, due to decay and reduction of the content in the soil, their possible inclusion in nutrition increases. The presented results show that if an inhabitant of Serbia were to exceed the value of 1 mSv, they would need to consume in a year: 6.6 kg penny bun from 2003 *i. e*. 8.0 kg of chanterelle from 2002 or 29.7 kg penny bun or 41.3 kg chanterelle from 2018. Our research also shows that an adult could consume about 13 kg of fungi per year with an activity level of ^{137}Cs 600 Bqkg^{-1} and not exceed the yearly recommended value of 1 mSv. It follows that the values of the annual effective doses are much lower than the doses recommended for the population (1 mSv) [32], so the effects on the population health are negligible concerning the total annual effective dose from natural sources of ionizing radiation, that is on average 2.4 mSv worldwide [45, 46].

CONCLUSION

Penny bun (*Boletus edulis*) and chanterelle (*Cantharellus cibarius*) are used for human consumption in several forms: fresh, thermally treated, or conserved and as such are exported from Serbia. The presence of ^{137}Cs was determined in each individual sample of penny bun and chanterelle and this is the reason why these fungi are unavoidable factors for radioecological monitoring of the ecosystem. High ac-

Table 3. Annual effective dose, D, [μSv] ingested by a person consuming 1 kg of fresh penny bun or chanterelle, annual effective dose for minimal, D_{min}, and for maximal values, D_{max}, of activity levels of ^{137}Cs in fungi

Year	Penny bun			Chanterelle		
	D	D_{min}	D_{max}	D	D_{min}	D_{max}
	[μSv]					
1999.	0.267	0.039	0.897	0.177	0.065	0.715
2000.	0.156	0.065	0.611	0.242	0.078	1.170
2001.	0.337	0.065	1.501	0.153	0.065	0.897
2002.	0.153	0.039	0.689	0.213	0.052	1.248
2003.	0.241	0.026	1.521	0.187	0.026	1.040
2004.	0.191	0.026	0.910	0.116	0.013	1.105
2005.	0.221	0.026	0.923	0.113	0.026	0.546
2006.	0.150	0.026	0.559	0.138	0.026	0.871
2007.	0.091	0.010	0.663	0.133	0.018	0.559
2008.	0.147	0.017	0.650	0.114	0.008	0.299
2009.	0.161	0.020	0.546	0.105	0.016	0.286
2010.	0.163	0.020	0.338	0.169	0.018	0.338
2011.	0.100	0.017	0.182	0.120	0.026	0.351
2012.	0.069	0.051	0.117	0.053	0.031	0.082
2013.	0.047	0.007	0.098	0.057	0.026	0.104
2014.	0.047	0.005	0.072	0.052	0.008	0.143
2015.	0.069	0.013	0.299	0.108	0.007	0.260
2016.	0.095	0.004	0.234	0.039	0.010	0.078
2017.	0.065	0.052	0.104	0.064	0.009	0.234
2018.	0.061	0.020	0.156	0.034	0.009	0.104

tivity levels of ^{137}Cs measured in penny bun and chanterelle indicate a potential direct danger for people consuming them. The results obtained indicate that in time the activity levels of ^{137}Cs reduced in the investigated fungi. The determined values of annual effective radiation doses were much lower than the values recommended for the population (1 mSv).

ACKNOWLEDGMENT

This work was supported by the Ministry of Education, Science and Technological Development of the Republic of Serbia, Contract on realization and financing of scientific research projects 451-03-68/2020-14 from 16.01.2020.

AUTHORS' CONTRIBUTIONS

A. A. Čučulović, J. N. Stanojković, and D. S. Veselinović analysed the samples, interpreted the results, and discussed them in reference to literature data. All the authors contributed to the manuscript preparation and approved its final version.

REFERENCES

[1] Stevanović, V., *et al*., Biological Diversity of Yugoslavia, FM for Development, Sci. and Env. & Ecolibri-Bionet, Belgrade, 1997

[2] Muntanola-Cvetković, M., *General Mycology* (in Serbian), Scientific Book, Belgrade, 1980

[3] Blackwell, M., The fungi: 1, 2, 3…5.1 Million Species?, *Am. J. Bot., 98* (2011), 3, pp. 426-438

[4] Dai, Y. C., Hymenochaetaceae (Basidiomycota) in China, *Fungal Divers., 45* (2010), 1, pp. 131-343

[5] Kalač, P., Trace Element Contents in European Species of Wild Growing Edible Mushrooms: A Review for the Period 2000-2009, *FOOD Chem., 122* (2010), 1, pp. 2-15

[6] Sanchez, C., Modern Aspects of Mushroom Culture Technology, *Appl. Microbiol. Biotechnol., 64* (2004), 6, pp. 756-762

[7] Sanchez, C., Reactive Oxygen Species and Antioxidant Properties from Mushrooms, *Synth. Syst. Biotechnol, 2* (2017), 1, pp. 13-22

[8] Patel, S., Goyal, A., Recent Developments in Mushrooms as Anticancer Therapeutics: A Review, 3 *Biotech., 2* (2012), 1, pp. 1-15

[9] Garibova, L. V., *In the Kingdom of Mushrooms* (in Russian), Forestry, Moscow, 1981

[10] Linkov, I., *et al*., Fungi Contaminated by Radionuclides: Critical Review of Approaches to Modeling, *Proceedings*, IRPA-10, Hiroshima, Japan, P-4b-255, 2000, pp. 1-10

[11] Cooper, E. L., *et al*., Radioactivity in Food and Total Diet Samples Collected in Selected Settlements in the USSR, *J. Environ. Radioact., 17* (1994), 1, pp. 147-158

[12] Persson, Ch., *et al*., The Chernobyl Accident – A Meteorological Analysis of How Radionuclides Reached and Were Deposited in Sweden, *Ambio, 16* (1987), 1, pp. 20-31

[13] Gudiksen, P. H., *et al*., Chernobyl Source Term, Atmospheric Dispersion and Dose Estimation, *Health Phys., 57* (1989), 5, pp. 697-706

[14] ***, Levels of Radioactive Contamination of the Human Environment and Radiation Exposure of the Population of Yugoslavia in 1986 Due to the Accident at the Chernobyl Nuclear Power Plant (in Serbian), Federal Committee for Labor, Health and Social Welfare, Belgrade, 1987

[15] Nichols, A. L., Hunt, E., Nuclear Data Table, in The Radiochemical Manual, (Longworth, G, ed), Howell, UK, 1998

[16] Kammerer, L., *et al*., Uptake of Radiocaesium by Different Species of Mushrooms, *J. Environ. Radioact., 23* (1994), 2, pp. 135-150

[17] Sugiyama, H., *et al*., Radiocesium Concentrations in Wild Mushrooms and Characteristic of Cesium Accumulation by the Edible Mushrooms (Pleurotus Ostreatus), *J. Health Sci., 46* (2000), 5, pp. 370-375

[18] ***, IAEA, Environmental Consequences of the Chernobyl Accident and their Remediation: Twenty Years of Experience, Report of the Chernobyl Forum Expert Group "Environment", Radiological Assessment Report Series, IAEA, Vienna, 2006

[19] Rantavaara, A., Radioactivity of Vegetables and Mushrooms in Finland After Chernobyl Accident in 1986: Supplement 4 to Annual Report STUK-A55 Helsinki: Finnish Centre for Radiation and Nuclear Safety, 1987

[20] Elstner, E. F., *et al*., Natural and Chernobyl-Caused Radioactivity in Mushrooms, Mosses, and Soil-Samples of Defined Biotops in SW Bavaria, *Oecologia, 73* (1987), 4, pp. 553-558

[21] Yoshida, S., *et al*., Radiocesium Concentrations in Mushrooms Collected in Japan, *J. Environ. Radioact., 22* (1994), 2, pp. 141-154

[22] Cooper, E. L., *et al*., Radioactivity in Food and Total Diet Samples Collected in Selected Settlements in the USSR, *J. Environ. Radioact., 17* (1992), 2+3, pp. 147-158

[23] Kostiainen, E., 1^{37}Cs in Finnish Wild Berries, Mushrooms and Game Meat in 2000-2005, *Boreal. Environ, Res., 12* (2007), 1, pp. 23-28

[24] Stanković, S., *et al*., Comparative Testing of Levels of Radiocaesium Contamination-137 and 134 of some bioindicators (in Serbian), *Proceedings*, XVII Yugoslav Symposium on Radiation Protection, Belgrade, Vinča Institute, 1993, pp. 65-68

[25] Stanković, S., Stanković, A., Radiocesium-137 in Cultivated and Forest Mushrooms (in Serbian), *Proceedings*, XIX Yugoslav Symposium on Radiation Protection, Golubac, 1997, pp. 175-178

[26] Duff, M. C., Ramsey, M. L., Accumulation of Radiocesium by Mushrooms in the Environment: a Literature Review, *J. Environ. Radioact., 99* (2008), 6, pp. 912-932

[27] Mietelski, J. W., *et al*., Radioactive Contamination of Polish Mushrooms, *Sci. Tot. Environ., 157* (1994), Dec., pp. 217-226

[28] Chiaravalle, A. E., *et al*., A Ten-Year Survey of Radiocontamination of Edible Balkan Mushrooms: Cs-137 Activity Levels and Assessed Dose to the Population, *Food Control, 94* (2018), Dec., pp. 263-267

[29] ***, Law on Nature Protection of the Republic of Serbia (in Serbian), Official Gazette of RS No.: 36/09 and 88/2010

[30] ***, The Cartagena Protocol on Biosafety to the Convention on Biological Diversity, ISBN: 92-807-1924 -6, Montreal, 2000

[31] ***, Decree on Control of the Use and Trade of Wild Flora and Fauna (in Serbian), Official Gazette of RS no.: 31/2005, 45/2005, 22/2007, 38/2008 and 09/2010
[32] ***, European Commission (EC), Council Regulation No 733/2008 of 15 July 2008 on the Conditions Governing Imports of Agricultural Products Originating in Third Countries Following the Accident at the Chernobyl Nuclear Power Station (Codified Version), 2008
[33] Hobbs, C., *Medicinal Mushrooms*, Botanica Press, Empire Grade, Santa Cruz, CA, 1995
[34] Wasser, S. P., Medicinal Mushrooms as a Source of Antitumor and Immunomodulating Polysaccharides, *Appl. Microbiol. Biotechnol., 60* (2002), 3, pp. 258-274
[35] Wasser, S. P., Weis A. L., Therapeutic Effects of Substances Occurring in Higher Basidiomycetes Mushrooms: a Modern Perspective, *Crit. Rev. Immunol., 19* (1999), 1, pp. 65-96
[36] Guillen, F. J., Baeza, A., Radioactivity in Mushrooms: A Health Hazard?, *Food Chem., 154* (2014), 1, pp. 14-25
[37] Baeza, A., *et al*., Radiocesium and Radiostrontium Uptake by Fruit Bodies of Pleurotus Eryngii Via Mycelium, Soil and Aerial Absorption, *App. Radiat. Isot., 53* (2000), 3, pp. 455-462
[38] Clarke, M. J., Smith, F. B., Wet and Dry Deposition of Chernobyl Releases, *Nature, 332* (1988), 6161, pp. 245-249
[39] ***, IAEA (International Atomic Energy Agency), Measurement of Radionuclides in Food and the Environment, Vienna, Technical Report Series No. 295, 1989
[40] ***, Gamma Vision-32: Gamma - Ray Spectrum Analysis and MCA Emulation, Software User s Manuel, ORTEC, 2001
[41] ***, ICRP (International Commission on Radiation Protection), Compendium of Dose Coefficients Based on ICRP Publication 60, ICRP Publication 119 Annals of ICRP, Vienna, 41(Suppl.), 2012
[42] ***, IAEA (International Atomic Energy Agency), International Basic Safety Standards for Protection against Ionizing Radiation and for the Safety of Radiation Sources, Vienna, Safety Series, No. 115, 1996
[43] Cardi, F., Belmusto, G., Radioactivity in Wild-Growing Mushrooms of the Calabria Region, South of Italy, Cogent Physics 4: 1354957, 2017
[44] Varfolmeeva, K. V., Culinary Processing of Dried Mushrooms as an Effective Way to Reduce the Content of Cesium-137 in Them (in Russian), *Radiation Hygiene, 12* (2019), 4, pp. 82-88
[45] ***, UNSCEAR (United Nations Scientific Committee on the Effects of Atomic Radiation): Sources and Effects of Ionizing Radiation, Report of the General Assembly, with Scientific Annexes, Annex B, Exposures of the Public and Workers from Various Sources of Radiation, United Nations, New York, 2008, 2010
[46] Mitrović, B. M., *et al*., Radionuclides and Heavy Metals in Soil, Vegetables, and Medicinal Plants in Suburban Areas of the Cities of Belgrade and Pančevo, Serbia, *Nucl Tecnol Radiat, 34* (2019), 3, pp. 278-384

Received on June 15, 2020
Accepted on August 24, 2020

Ана А. ЧУЧУЛОВИЋ, Јелена Н. СТАНОЈКОВИЋ, Драган С. ВЕСЕЛИНОВИЋ

ДВАДЕСЕТОГОДИШЊЕ ИСТРАЖИВАЊЕ РАДИОКОНТАМИНАЦИЈЕ ЈЕСТИВИХ ГЉИВА СРБИЈЕ
Нивои активности ^{137}Cs и процењена доза за популацију

Вргањ (*Boletus edulis*) и лисичарка (*Cantharellus cibarius*) се користи за људску исхрану у више облика: свеже, термички обрађене или конзервиране (усољене, мариниране, пастеризоване, сушене) и као такве се извозе из Србије. У сваком појединачном узорку утврђено је присуство ^{137}Cs, који је чернобиљског порекла и због тога су вргањ и лисичарка незаобилазни чиниоци радиоеколошких испитивања екосистема. Мониторинг животне средине коришћењем гљива је важан за процену радијационог оптерећења становништва одређеног простора. Високе вредности активности ^{137}Cs измерене у јестивим гљивама, указују на потенцијалну директну опасност по људе који их користе. Резултати упућују да се временом у испитиваним гљивама смањује ниво активности ^{137}Cs. Вредности годишњих ефективних доза зрачења су много мање од вредности препоручених за становништво (1 mSv)

Кључне речи: Boletus edulis, Cantharellus cibarius, ^{137}Cs, годишња ефективна доза