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RADIOACTIVITY IN THE ENVIRONMENT AND FOOD CHAIN AT MT. MALJEN, SERBIA

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Abstract. This paper provides activity concentrations of ^{40}K, ^{238}U, ^{226}Ra, ^{232}Th, and ^{137}Cs in samples of cultivated soil, hay, cow milk, cheese, mushrooms, and mosses collected at Mt. Maljen, Serbia, during 2018 and 2019. The average contents of ^{40}K (435 ± 33 Bq/kg), ^{238}U (42 ± 5 Bq/kg), ^{226}Ra (42 ± 4 Bq/kg) and ^{232}Th (47 ± 4 Bq/kg) in the soil are slightly higher than the global means. Radiation hazard index is less than one. ^{137}Cs is detected in soil (3.1 – 111 Bq/kg), hay (4.6 – 9.4 Bq/kg), cow milk (2.0 ± 0.2 Bq/kg), cheese (1.7 ± 0.2 Bq/kg), mushrooms (26 ± 2 Bq/kg), and mosses (21 ± 2 Bq/kg). Investigated dairy products are safe for consumption.

Key words: natural radionuclides, ^{137}Cs, radiation hazard parameters.

1. INTRODUCTION

Exposure of living organisms to ionizing radiation stems from both natural and artificial sources [1]. Natural radionuclides in the uranium and thorium series and their decay products, as well as ^{40}K, are the major contributors to irradiation due to their gamma-ray emission. The worldwide exposure resulting from the natural radiation sources varies geographically, with the mean value of 2.4 mSv/y (1–10 mSv/y). On average, the population receives 0.48 mSv/y from external terrestrial radiation, 0.39 mSv/y from cosmic radiation and cosmogenic radionuclides, 0.29 mSv/y from ingestion of ^{40}K and radionuclides in the uranium and thorium series, and 1.15 mSv/y from inhalation exposure to ^{222}Rn [1]. The content of naturally occurring radionuclides in the soil is related to the type of parent rock and soil genesis [2]. The presence of the natural and artificial radionuclides in the soil leads to their transfer to plants and onward through the food chain [3–6].

Artificial radionuclides, *e.g.* ^{131}I, ^{134}Cs, ^{137}Cs, and ^{90}Sr, that entered the environment as a result of nuclear tests and accidents, contribute to an additional irradiation of humans and non-human biota. After the Chernobyl accident in 1986, about 2.4% of the total radioactivity was deposited at the territory of the former Republic of Yugoslavia [7]. On the other hand, the contamination from the 2011

nuclear accident in Fukushima did not reach the Republic of Serbia due to its great distance from the accident site. Some of the fission products present in the soil are of particular radiological significance because of their chemical similarity to essential elements (^{134,137}Cs–K; ^{89,90}Sr–Ca) which renders them easily incorporated into the food chain. Thus, the long-lived ^{137}Cs (half-live about 30 years) deposited in Serbia after the Chernobyl accident, is still present in the environment and transferred amongst different environmental compartments. The ^{137}Cs transport through terrestrial food chain depends on many factors: ecosystem type, soil type, moisture and clay content, rainfall, vegetation, activities of both animals and humans [8–9]. Consequently, ^{137}Cs is primarily present in the soil and some bioindicators such as mosses, mushrooms, berries and game meat [5, 10–13].

Radionuclides present in the soil and food can lead to human exposure by external and internal irradiation. Therefore, monitoring gamma-ray emitters is a very useful method to: 1) estimate and assess the terrestrial radiation dose to the human population; 2) recognize regions of elevated natural radiation hazard [14], and 3) determine the content of artificial radionuclides in the environment and food chain. Our study addresses these three goals – we collected different environmental samples at Mt. Maljen, in the Republic of Serbia, determined activity concentration of the natural (^{40}K, ^{238}U, ^{226}Ra, and ^{232}Th) and artificial (^{137}Cs) radionuclides, and calculated the radiation hazard parameters to evaluate the state of the environment in this region of Serbia.

2. MATERIALS AND METHODS

2.1. SAMPLING AND MEASUREMENT

During the summers of 2018 and 2019, we collected samples at Mt. Maljen (Fig. 1), located in the west of Serbia, around 20 km south of the Valjevo town (population 60,000). The mountain spreads in the east-west direction for approximately 25 km. The highest peak is Kraljev sto (elevation 1104 m a.s.l.). The mountain is a popular holiday and recreation destination throughout the year, but the land is mostly used for animal husbandry within individual holdings.

At each of the five locations: Mionica (elevation 200 m a.s.l), Brežđe (elevation 500 m a.s.l), Tometino polje (elevation 700 m a.s.l), Divčibare (elevation 980 m a.s.l), and Kraljev sto (elevation 1100 m a.s.l), we took samples of the cultivated soil (3 samples per location), hay (3 samples per location), cow milk and cheese (3 samples per location, excluding Divčibare) from individual holdings. In addition, we collected samples of mushrooms and mosses in the vicinity of these holdings and prepared composite samples: 2 samples of mushrooms and 2 samples of mosses. In total, 58 samples were collected and transported to an accredited Laboratory for Radiation Hygiene of the Faculty of Veterinary Medicine, University of Belgrade, Serbia, where they were analyzed.

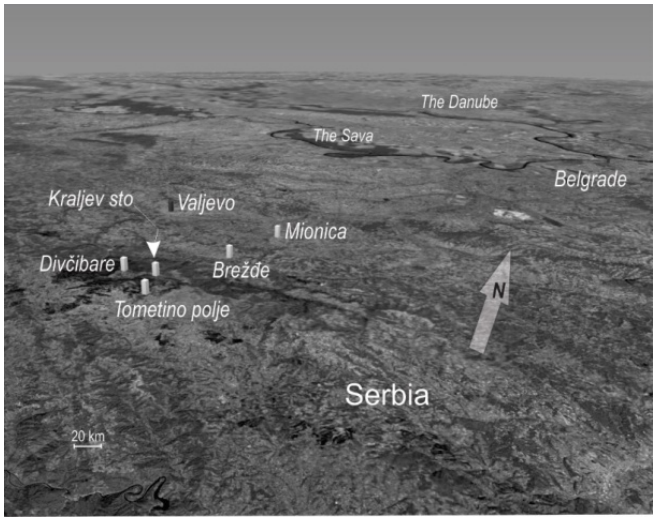

Fig. 1 – View of Serbia and sampling sites at Mt. Maljen. Locations of the nearby town Valjevo and capital of Serbia, Belgrade, are given for reference.

The cultivated soil samples were collected up to 15 cm soil depth. Before the analysis, the samples were crushed, dried at 105°C, sieved, homogenized, and stored in Marinelli beakers (1 l). The samples of hay and mosses were dried in the oven at a temperature of 105°C until constant weight, and then homogenized and stored in Marinelli beakers (0.5 l). Cow milk, cheese, and mushrooms were measured in Marinelli beakers (1 l).

Gamma-ray spectrometric determination of radionuclides content and concentrations was performed on high-purity germanium detectors (ORTEC) with a relative efficiency of 30% and 33%, and energy resolution 1.85 keV (1332.5 keV of ^{60}Co) and ORTEC low-background germanium radiation detector shields.

Software program GAMMA VISION-32 was used for the analysis of each obtained gamma-ray spectrum. All obtained results are expressed as (mean ± standard deviation). The counting time for the samples, as well as for background, was 60,000 s. Commercially available standards with mixed radionuclides were used for efficiency calibration of the detectors:

- for soil samples, we used ^{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, volume 1 l;
- for cow milk, cheese and mushrooms we used ^{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 baker, density (0.98±0.01) g/cm, volume 1 l; and
- for hay and moss samples, we used certified reference radionuclides ^{241}Am, ^{109}Cd, ^{139}Ce, ^{57}Co, ^{60}Co, ^{137}Cs, ^{85}Sr, ^{88}Y, ^{51}Cr, dispersed in milled grass (Marinelli beaker 0.5 l).

The minimum detectable activity (MDA) for each radionuclide was determined using the following equation (GammaVision®-32, 2006):

$$\text{MDA} = 2 \times (1 + \text{sqrt}\,(1 + 2B_1)) / \varepsilon \times I \times T \times M, \quad (1)$$

where B_1 is background reported in the identified peak summary (counts), ε is absolute efficiency of detector, I is the gamma emission probability, T is counting time, and M is the mass of the sample (in kg). The minimum detectable activity of ^{137}Cs was 0.3 Bq/kg for hay and 0.1 Bq/kg for milk and cheese.

2.2. CALCULATION OF RADIATION HAZARD PARAMETERS

Radium equivalent activity

We used radium equivalent activity (Ra_{eq}) to compare the activity concentrations of materials with different contents of ^{226}Ra, ^{232}Th, and ^{40}K. We calculated Ra_{eq} using [1]:

$$Ra_{eq}\ [\text{Bq/kg}] = C_{Ra} + 1.43C_{Th} + 0.077\ C_K, \quad (2)$$

where C_{Ra}, C_{Th}, and C_K are the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K (in Bq/kg), respectively.

Absorbed dose rate

We calculated absorbed dose rate ($\dot{D}$) using conversion factors under an assumption that all decay products of ^{226}Ra and ^{232}Th are in equilibrium with their precursors [1]:

$$\dot{D}\ [\text{nGy/h}] = 0.462\ C_{Ra} + 0.604\ C_{Th} + 0.042\ C_K. \quad (3)$$

Annual effective dose equivalent

We used the absorbed dose rate data (Eq. 3), adopted the conversion factor of 0.7 Sv/Gy [1] and assumed that on average, people in Serbia spend 20% of their time outdoors, and thus obtained annual effective dose equivalent (AEDE):

$$\text{AEDE}\ [\mu\text{Sv/y}] = \dot{D}\ [\text{nGy/h}] \times 24\ [\text{h}] \times 365\ [\text{days}] \times 0.7\ [\text{Sv/Gy}] \times 0.2. \quad (4)$$

External hazard index

Beretka and Mathew [15] defined the external hazard index (H_{ex}):

$$H_{ex} = C_{Ra}/370 + C_{Th}/259 + C_K/4810. \quad (5)$$

The radiation hazard is insignificant when H_{ex} value is less than one. The external hazard index equal to one corresponds to the upper limit of radium equivalent activity (370 Bq/kg).

Annual gonadal dose equivalent

To calculate annual gonadal dose equivalent (AGDE) to the population arising from the presence of naturally occurring radionuclides ^{226}Ra, ^{232}Th, and ^{40}K in the soil, we used [16]:

$$\text{AGDE}\ (\mu\text{Sv/y}) = 3.09\ C_{\text{Ra}} + 4.18\ C_{\text{Th}} + 0.314\ C_{\text{K}}. \quad (6)$$

Excess lifetime cancer risk outdoors

We calculated excess lifetime cancer risk outdoors ($\text{ELCR}_{\text{outdoor}}$) using [17–18]:

$$\text{ELCR}_{\text{outdoor}} = \text{AEDE} \times \text{DL} \times R_f, \quad (7)$$

where DL is the duration of life (70 years average) and R_f is the risk factor, *i.e.* fatal cancer risk per sievert. For stochastic effects, we used $R_f = 0.05\ \text{Sv}^{-1}$ for the whole population [19].

3. RESULTS AND DISCUSSION

Table 1 gives the results of gamma-ray spectrometry analysis of ^{40}K, ^{238}U, ^{226}Ra, ^{232}Th and ^{137}Cs in the cultivated soil collected at different altitudes at Mt. Maljen. In the soil, the activity concentration of ^{40}K, ^{238}U, ^{226}Ra, and ^{232}Th were in the range (173–641) Bq/kg, (25–50) Bq/kg, (26–51) Bq/kg, and (22–73) Bq/kg, respectively. The maximum content of the natural radionuclides was detected in Brežđe (500 m), while the minimum content in the highest location Kraljev sto (1100 m). According to UNSCEAR [18] the world average activity concentrations of ^{40}K, ^{238}U, ^{226}Ra, and ^{232}Th are 420 Bq/kg, 33 Bq/kg, 32 Bq/kg, and 45 Bq/kg, respectively. Our results show that the activity concentrations of the natural radionuclides are slightly higher than the global averages, except in Kraljev sto (1100 m).

Table 1

Activity concentration of ^{40}K, ^{238}U, ^{226}Ra, ^{232}Th and ^{137}Cs (in Bq/kg) in the cultivated soil

Sampling site	Altitude (m)	^{40}K	^{238}U	^{226}Ra	^{232}Th	^{137}Cs
Mionica	200	487±35	48±5	50±5	59±5	11±1
Brežđe	500	641±50	50±5	51±4	73±6	21±2
Tometino polje	700	396±30	42±4	39±4	38±4	111±5
Divčibare	900	478±35	44±5	44±5	44±4	3.1±0.3
Kraljev sto	1100	173±15	25±3	26±3	22±3	23±2
Average	–	435±33	42±5	42±4	47±4	34±2

An analysis of uncultivated soil at Mt. Maljen [10] showed that at lower altitudes (200–650 m), the content of the natural radionuclides was above the

global means, but as altitude increased, their content decreased. The mean values of the ^{40}K, ^{238}U, ^{226}Ra and ^{232}Th in the uncultivated soil were 254 Bq/kg, 33 Bq/kg, 33 Bq/kg and 29 Bq/kg, respectively, *i.e.* about two-fold lower than the results obtained in the present study. Activity concentrations of U, Th and K depend on parent rock, soil type and texture [20], but the differences we see between the cultivated and uncultivated soil most likely arise from the use of mineral fertilizers which can increase a content of these radionuclides in the soil [21–22].

Table 2 lists the radiation hazard parameters for the investigated locations. The values of Ra_{eq} ranged from 70 Bq/kg (Kraljev sto) to 200 Bq/kg (Brežđe) and were below the limit of 370 Bq/kg for Ra_{eq} in building materials [15]. The value of 370 Bq/kg corresponds to an annual effective dose equivalent of 1 mSv, and presents the dose limit for the general population. The mean values of absorbed dose rate (66 nGy/h) and AEDE (81 μS/y) were slightly higher than the world average of 59 nGy/h [1] and 70 μS/y [18], respectively. However, with the mean external hazard index of 0.39, the radiation hazard is insignificant in this region.

Table 2

Radiation hazard parameters for the cultivated soil samples

Sampling site	Altitude (m)	Ra_{eq} (Bq/kg)	$\dot{D}$ (nGy/h)	AEDE (μS/y)	H_{ex}	AGDE (μSv/y)	$ELCR_{outdoor} \times 10^{-3}$
Mionica	200	168	79	97	0.46	554	0.34
Brežđe	500	200	95	116	0.55	664	0.41
Tometino polje	700	121	58	71	0.33	404	0.25
Divčibare	900	140	67	82	0.39	470	0.29
Kraljev sto	1100	70	33	40	0.19	227	0.14
Average	–	140	66	81	0.39	464	0.28
Average world values		370[15]	59[1]	70[18]	≤ 1[23]	1000[19]	0.29 [18]

The annual gonadal dose equivalent ranged from 227 μSv/y to 664 μSv/y, with a mean of 464 μSv/y (Table 2). These values were lower than 1 mSv/y which is the recommended limit by the International Commission on Radiological Protection [19] for the general public. The excess lifetime cancer risk outdoors ($ELCR_{outdoor}$) gives the probability of cancer incidence in a population during the lifetime due to exposure to natural radionuclides in the soil. Our results show that $ELCR_{outdoor}$ values varied between 0.14 and 0.41, with the mean value (0.28×10^{-3}) slightly lower than the world average of 0.29×10^{-3} [18].

Anthropogenic radionuclide ^{137}Cs was also detected in the cultivated soils from Mt. Maljen (Table 1). Before the Chernobyl accident, the activity concentration of ^{137}Cs in the soil in Serbia was less than 5 Bq/kg [7] and the deposition of ^{137}Cs in the soil was unequal and non-homogenous. Mitrović *et al.* [10] reported that the ^{137}Cs content in the uncultivated soil at Mt. Maljen significantly increased with altitude – from 19 Bq/kg at 200 m a.s.l., to 259 Bq/kg at 1100 m a.s.l. These findings are in contrast with the present study where we observe that the ^{137}Cs

activity concentration in the cultivated soil increases as elevation increases from 200 m to 700 m (from 11 Bq/kg to 111 Bq/kg), but at the highest altitudes of 900 m and 1100 m, the ^{137}Cs content decrease several-fold (to 3.1 Bq/kg and 23 Bq/kg, respectively). This low activity concentration of ^{137}Cs at the highest altitudes could be explained by the differences in soil type (cultivated and uncultivated) on one hand, but also by the fact that the sampling in these two studies was not performed at identical locations. Further, the application of agro-technical measures, such as ploughing and use the fertilizers, can significantly reduce the content of radiocaesium in the cultivated soil [24]. In contrast, the higher ^{137}Cs activity concentration (111 Bq/kg) detected at Tometino polje (700 m), is probably a result of the non-uniform contamination of the environment after the Chernobyl accident, as well as the radionuclide retention by the soil structure at this location. An investigation conducted at the Mountain of Tara, Serbia, five years after the accident, showed that the ^{137}Cs content in the soil varied from 88 Bq/kg to 462 Bq/kg, depending on the soil type [25]. The major factors impacting radionuclide retention and mobility in the soil are: pH value, content of inorganic ions and organic substances, characteristic and content of clay minerals, microorganism, fungi, etc. [8–9]. Hence, to determine the exact reasons for the ^{137}Cs increase at this particular site at Mt. Maljen, a further analysis, encompassing physico-chemical properties of the soil, is needed.

The presence of radionuclides in the soil is a starting point for their transfer to plants, animals, and humans. Anthropogenic radionuclide ^{137}Cs, which is chemically similar to potassium, gets closely involved in nutrient cycles and may be present in the food chain for many decades after deposition [12, 26–27]. Our study confirms this finding for the hay, milk and cheese samples (Tables 3 and 4).

Table 3

Activity concentration of ^{40}K and ^{137}Cs in the hay (dry weight, in Bq/kg)

Sampling site	Altitude (m)	^{40}K	^{137}Cs
Mionica	200	663 ± 40	<MDA
Brežđe	500	704 ± 40	<MDA
Tometino polje	700	236 ± 17	4.6 ± 0.4
Divčibare	900	460 ± 35	6.0 ± 0.6
Kraljev sto	1100	455 ± 33	9.4 ± 1.5

In the hay, ^{137}Cs was detected in the samples collected from higher altitudes (700–1000 m), while at the lower altitudes, its activity concentration was below the maximum detectable activity (Table 3). Similarly, ^{137}Cs was detected in the cow milk (2.0 Bq/kg) and cheese (1.7 Bq/kg) only at the highest altitude, location Kraljev sto (Table 4). According to the Serbian Rulebook [28], the maximal permitted concentration of ^{137}Cs in foodstuff is 15 Bq/kg, so the investigated milk and cheese samples were safe for human consumption.

Table 4

Activity concentration of ^{40}K and ^{137}Cs in the cow milk and cheese (in Bq/kg)

Sampling site	Altitude (m)	^{40}K	^{137}Cs
cow milk			
Mionica	200	48 ± 4	<MDA
Brežđe	500	59 ± 4	<MDA
Tometin polje	700	49 ± 4	<MDA
Kraljev sto	1100	47 ± 3	2.0 ± 0.2
cow cheese			
Mionica	200	46 ± 4	<MDA
Brežđe	500	42 ± 3	<MDA
Tometino polje	700	39 ± 3	<MDA
Kraljev sto	1100	36 ± 3	1.7 ± 0.2

In a previous study conducted at Mt. Maljen [10], ^{137}Cs was detected in the hay at all of the investigated altitudes (200–1100 m a.s.l.), and its concentration varied between 1.1 Bq/kg and 38.6 Bq/kg. On the other hand, ^{137}Cs in the cow milk (6.9 Bq/kg) and cheese (3.6 Bq/kg) was detected only at the higher altitude of 1100 m a.s.l., the same as in the present study. It is worth noting that overall, the ^{137}Cs concentrations in the environmental samples decreased by a factor 2–3 compared to 2002–2007 [10]. This finding is in agreement with other studies conducted in Europe. For example, a long-term investigation performed in France [27] showed that the effective half-lives of ^{137}Cs in the pasture ecosystem are 11 and 9 years for the grass and milk, respectively.

Transfer of ^{137}Cs through different compartments of the food chain is another question often addressed in radioecological investigations. Although our study did not investigate this particular issue, it is interesting to note that the ^{137}Cs transfer might be influenced by the content of potassium. On one hand, Corcho-Alvarado *et al.* [29] showed that the transfer of ^{137}Cs to grass was not affected by the variations of the K content in the soil, while Karunakara *et al.* [30] reported that the grass to milk ^{137}Cs transfer was influenced by the ^{40}K activity concentration in the grass, and that the higher transfer occurs when the ^{40}K content is lower.

The natural ^{40}K was detected in the milk and cheese, with the average activity concentration of 51 Bq/kg and 41 Bq/kg, respectively (Table 4), which is in agreement with the results of Alharshan *et al.* [31] who reported that content of ^{40}K in the milk was in the range 40–98 Bq/kg.

Due to their physiological characteristics, mushrooms and mosses are good bioindicators of radioactive pollution of the environment (Table 5), even decades after the Chernobyl accident [32]. Mushrooms are consumed by humans for their nutritional and medicinal benefits, but they can be a significant source of internal contamination with radiocaesium [33]. After the Chernobyl accident in several European countries, the activity concentration in mushrooms was higher than 100,000 Bq/kg dry weight [33], but today, these concentrations are much lower

[10, 34–35]. An investigation performed in the Calabria region, Southern Italy, showed activity concentration of ^{137}Cs in wild-growing mushrooms between 0.3 Bq/kg and 73.1 Bq/kg, but the effective dose due to its ingestion (0.25 μS/y to 1.35 μS/y) was lower than the maximum recommended level of 1 mSv/y for the public [13].

Table 5

Activity concentration of ^{40}K and ^{137}Cs in mushrooms and mosses (in Bq/kg)

Sample	^{40}K	^{137}Cs
Mushrooms (f.w.)	91 ± 8	26 ± 2
Mosses (d.w.)	119 ± 10	21 ± 2

f.w. – fresh weight; d.w. – dry weight

Results published by Mitrović *et al.* [10] showed that activity concentration of ^{137}Cs in mushrooms collected at Mt. Maljen depended on the mushroom species and varied between 93 Bq/kg and 385 Bq/kg d.w. (average 258 Bq/kg), while in mosses, the ^{137}Cs activity concentration was 1200 Bq/kg d.w., which is several-fold higher than our results. Gamma-ray spectrometric analyses of mosses collected in 92 sites in Bosnia and Herzegovina showed that ^{137}Cs was present in all tested samples (4 Bq/kg to 1612 Bq/kg d.w.) and that radiocaesium accumulation depends on the moss species, location, substrate, and altitude [36]. The above findings indicate that mushrooms and mosses are good bioindicators of radiocaesium in the environment over a prolonged period of time.

4. CONCLUSION

The results of our study confirm the importance of radiation monitoring of the environment with the aim to protect people and non-human biota from external and internal irradiation. Our radioecological investigation at Mt. Maljen showed that the average values of activity concentrations of ^{238}U, ^{226}Ra and ^{232}Th in the cultivated soil were slightly higher than the global averages, except at the highest location where the content of the natural radionuclides was lower.

The mean values of absorbed dose rate and annual effective dose equivalent were also slightly higher than the world averages, but the radiation hazard index was less than one, indicating an insignificant radiation hazard in this region.

The artificial ^{137}Cs was present in the cultivated soil, hay, mushrooms, and mosses. In mushrooms and mosses, the activity concentration of the radionuclide decreased several-fold compared to the previous investigation performed at Mt. Maljen. This radionuclide was also detected in the cow milk and cheese samples, however, only at the highest investigated location, and in concentrations lower than the maximal permitted concentration, making them safe for human consumption.

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