Natural and anthropogenic radioactivity in the environment of mountain region of Serbia

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Natural and anthropogenic radioactivity in the environment of mountain region of Serbia

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The activity concentrations of ^{40}K, ^{238}U, ^{232}Th and ^{137}Cs have been measured using a gamma spectrometric method in different samples from the environment of two mountains in Serbia (altitude 1000–1100 m), during the period 2002–2007. The mountains Maljen and Tara (popular tourist destinations) are near Belgrade. On mountain Maljen, samples were taken at 4 different altitudes (200 m, 650 m, 1000 m and 1100 m), and on mountain Tara at altitudes of 1000 m and 1100 m. On mountain Maljen it was found that the level of ^{137}Cs activity increased with altitude in samples of soil, grass, hay and cow, sheep and goat milk. On the contrary, ^{40}K activity decreased with altitude in samples of soil, grass and hay. The highest activity concentrations of ^{137}Cs were found in bioindicators: sheep meat, venison, wild boar meat, moss and mushrooms. These results indicate that ^{137}Cs is present in mountain region of Serbia even 20 years after the nuclear accident in Chernobyl. Deposition of ^{137}Cs was almost two times higher on the Maljen mountain compared to Tara mountain. An average annual dose arising from ^{137}Cs was 7.4 μSv due to ingestion of cow milk and 6.3 μSv due to ingestion of mushrooms at the Maljen mountain.

1. Introduction

Radioactive contamination of environment with anthropogenic radionuclides in Serbia appeared after the nuclear accident in Chernobyl (1986). Even 20 years after the accident, the presence of ^{137}Cs may be detected in the environment, especially in bio-indicator organisms, because of its long half-life (30 years). Hecht[11] concluded, after the Chernobyl accident, that mosses, lichens and mushrooms are typical representatives of bio-indicator plants for radiocesium. High activity ^{137}Cs in mushrooms has been found by other scientists.[3,7,13,24] Wild animals are good indicators of radioactive contamination of the environment too. Because of their nutritional behavior, they accumulate higher levels of ^{137}Cs in their tissues. Hecht[10] investigated the activity level of ^{137}Cs in game meat before and after the Chernobyl accident. He found a significant increase of radiocesium activity.

Worldwide use of nuclear energy, application and nuclear weapon probes, coal combustion, production and application of phosphorus fertilizers, the mining industry and formation of radioactive waste dumps contribute to distribution of natural radioactivity. The quantity of radionuclides from these cited sources impact the environment significantly even now.[2,5,18,24] One of the most important anthropogenic sources of environment pollution with primordial radionuclides ^{226}Ra, ^{232}Th and ^{40}K is production and application of phosphorus fertilizers.[1] Uranium of anthropogenic origin found in soil is most often in a form which is easily available for plants, and therefore the possibility for its food chain incorporation is higher.[6]

A radiological investigation of a certain area assumes collection of samples from the environment which properly represents the potential danger to population health. For that purpose, soil, grass, domestic animal products samples,[4,26] meat of wild animals and mushrooms[12,16,17] and mosses[8,20,25] were investigated.

The goal of this paper was to determine the level of activity and presence of radionuclides ^{40}K, ^{238}U, ^{232}Th and ^{137}Cs in the environment and food chain: soil–plants–animals–humans in the territory of two Serbian mountains, which are popular tourist destinations located near Belgrade.

2. Materials and methods

The samples from the environment were collected in the period 2002–2007 in the area of Maljen and Tara mountains (Fig. 1).

Fig. 1 Geographic map of Serbia – location of Tara and Maljen mountains.

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The samples were collected at different altitudes on the Maljen mountain (200 m, 650 m, 1000 m, and 1100 m) and at an altitude of 1000 m and 1100 m on Tara mountain.

Soil samples were collected at a depth of 10–20 cm, homogenized, then dried at a temperature of 105 °C and put in 1 l Marineli beakers. Grass and hay samples were collected in amounts of 2–5 kg, ground and burned to mineral ash. Plastic vessels of volume 200 ml were used for measurements.

Milk samples were collected in amounts of 5 l, evaporated to 1 l and then placed in Marineli beakers. Fresh meat and cheese samples were homogenized and measured into 1 l Marineli beakers.

Mushrooms samples were first cleaned of soil and other impurities and dried at 80 °C until constant mass and measured into 200 ml vessels.

All samples were kept at least 30 days before measurement.

The activity of samples was determined by gamma spectrometric measurement on an HPGe detector (Ortec), with relative efficiency 30% and energy resolution 1.85 keV (1332.5 ^{60}Co). The analysis of each measured γ-ray spectrum has been carried out by a software program GAMMA VISION®-32.

The total uncertainty value (σ_t) is composed of the random and systematic errors:

$$\sigma_t = \sqrt{\sum \sigma_{ri}^2 + \frac{1}{3}\sum \sigma_{si}^2}$$

where: σ_{ri} = the individual random error (counting, random summing), σ_{si} = the individual systematic error (nuclide uncertainty from library, efficiency fitting uncertainty from calibration, calibration source uncertainty).

The analysis of ^{238}U by gamma spectrometry relies on the hypothesis of equilibrium conditions between parent nuclide ^{238}U and daughters. ^{226}Ra activity was determined from the ^{226}Ra daughters in equilibrium. We have used the weighted mean of the activities of three photopeaks of ^{214}Bi (609.3, 1120.0, and 1764.5 keV). In the case of ^{232}Th, three photopeaks of ^{228}Ac (338, 911.2 and 969 keV) were used in the same way. The activity of ^{40}K was derived from the 1460.8 keV gamma line of this isotope.

The detector efficiency calibration was performed for different geometries and different matrices, according to the sample type we measured. 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 Marineli beaker, density (0.98 ± 0.01) g/cm^3, volume 1 l;

Table 1 ^{40}K, ^{238}U, ^{232}Th, ^{137}Cs activity concentration in different samples at different altitudes on Maljen mountain (Bq kg^{-1})[a]

Samples/altitude	No. of samples	^{40}K	^{238}U	^{232}Th	^{137}Cs
Soil (f.w.)[b]					
200 m	10	449.9 ± 5.1	60.9 ± 5.6	56.7 ± 1.7	18.8 ± 0.6
650 m	10	394.1 ± 4.9	45.5 ± 3.3	41.5 ± 1.7	46.9 ± 0.7
1000 m	10	69.6 ± 2.3	10.9 ± 1.1	7.5 ± 0.3	63.9 ± 1.9
1100 m	10	102.3 ± 1.0	13.5 ± 1.0	11.3 ± 0.5	259.2 ± 0.9
Grass (f.w.)					
200 m	10	722 ± 21	4.0 ± 0.9	2.4 ± 0.5	1.8 ± 0.1
650 m	10	223.5 ± 6.7	<0.5	<0.5	<0.3
1000 m	10	386 ± 17	<1.5	<0.3	8.9 ± 0.1
1100 m	10	196.6 ± 5.9	<0.9	<0.2	9.3 ± 0.2
Hay (f.w.)					
200 m	10	946 ± 28	<9	<2	1.1 ± 0.3
650 m	10	1000 ± 35	<8.5	<2.6	1.5 ± 0.3
1000 m	10	956 ± 29	<12	<1.5	11.3 ± 1.3
1100 m	10	360 ± 10	<0.7	<0.2	38.6 ± 1.5
Cow's milk (f.w.)					
200 m	10	62.9 ± 2.1	<0.1	<0.1	<0.3
650 m	10	66.4 ± 1.9	<0.3	<0.1	<0.1
1000 m	10	64.2 ± 2.1	<0.4	<0.1	<0.1
1100 m	10	88.0 ± 2.7	<0.6	<0.1	6.9 ± 0.2
Sheep's milk (f.w.)					
650 m	10	87.1 ± 3.1	<1.5	<0.1	0.9 ± 0.1
1000 m	10	53.4 ± 1.7	<0.9	<0.1	12.3 ± 1.3
1100 m	10	80.8 ± 2.5	<0.8	<0.1	21.7 ± 1.5
Goat's milk (f.w.)					
200 m	10	98.2 ± 2.9	<0.5	<0.1	<0.1
1000 m	10	119.6 ± 4.1	<1.4	<0.2	24.1 ± 1.1
Cow's cheese (f.w.)					
650 m	10	66.2 ± 2.4	<1.3	<0.2	<0.2
1000 m	10	43.1 ± 2.9	<0.9	<0.1	3.6 ± 0.1
Sheep meat (f.w.)					
200 m	10	129.9 ± 4.1	<0.6	<0.1	<0.2
650 m	10	160.2 ± 5.1	<0.7	<0.1	4.5 ± 0.1
1000 m	10	129.4 ± 4.5	<0.6	<0.1	45.8 ± 1.9

[a] Mean ± standard deviation. [b] Fresh weight.

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

The first calibration standard was used for fresh sample measurements and the second was used for the soil samples measurement.

The ash samples were measured in a cylindrical geometry. Calibration for cylindrical geometry was performed using natural radioactive elements, such as potassium (^{40}K) and lanthanum (^{138}La).[21] We used ash as a matrix and mixed it with KCl and La_2O_3.[19,22] The efficiency curve was applied for the energy region 600 keV–1800 keV (eff = exp(3.972 − 1.163 × ln E).

Our laboratory participated in IAEA AQCS Programme Intercomparation Exercise, IAEA-385.[23]

According to the IAEA[15] and Vilic,[27] the average annual effective dose that an individual receives due to the radiocesium ingestion by consumption of contaminated meat, milk and mushrooms was calculated using the following formula:

$$D = D_{\mathrm{f}} \times C_{\mathrm{sample}} \times M_{\mathrm{ingested}}$$

where: D is annual individual effective dose (Sv); D_{f} is effective dose per unit intake of ^{137}Cs *via* ingestion (1.3 × 10^{-8} Sv/Bq), C_{sample} is activity concentration of ^{137}Cs in ingested food (Bq kg^{-1}) and M_{ingested} is mass of food ingested during one year (kg).

3. Results and discussion

The average ^{40}K, ^{238}U, ^{232}Th and ^{137}Cs activity concentration in soil, fodder and animal product samples, collected in the area of the Maljen mountain, are presented in Table 1.

The results show that ^{137}Cs is incorporated in the food chain soil–plant–animals on Maljen mountain. After the Chernobyl nuclear accident, about 2.4% of total polluted radionuclides (without inert gasses), or 5% ^{131}I and 10% ^{137}Cs, were deposited on the territory of the former Republic of Yugoslavia. Rainfall activity in the Yugoslavia territory was 880 Bq m^{-2} in the zone of minimal concentration, and 102 000 Bq m^{-2} in the zone of maximal concentration.[9] Radiocesium in the Yugoslavian environment, as a consequence of radioactive rainfall, was incorporated into the food chain: soil–plants–animals–human.

The ^{137}Cs activity concentration increased with altitude in all samples. The ^{137}Cs activity concentration in the soil increased several times from an altitude of 200 m (23.5 Bq kg^{-1}) to 1100 m (259.2 Bq kg^{-1}).

Similarly, in samples of hay, which is a basic feedstuff for ruminants in winter time, a ^{137}Cs activity concentration increase from 1.1 to 38.6 Bq kg^{-1} with increasing altitude was found.

The ^{137}Cs activity concentration in cow's milk was in the range from 0.3 to 6.9 Bq kg^{-1}. The highest activity in milk was found at the altitude 1000–1100 m, which can be explained as a result of the increased activity in soil, grass and hay at these altitudes. The ^{137}Cs activity in samples of cow's cheese was 3.6 Bq kg^{-1}.

During the milk processing into products with high milkfat, such as these types of cheeses, the decrease in ^{137}Cs activity occurs.[14] Fr (retention factor) for ^{137}Cs is 0.07, and Pe (processing efficiency) is 0.12. The ratio Fr/Pe = 0.07/0.12 = 0.58, means that in cow cheese the ^{137}Cs activity was 0.58 times lower than in milk. ^{137}Cs activity in cow cheese was: 6.9 Bq kg^{-1} × 0.58 = 4.0 Bq kg^{-1}, which corresponds to the results obtained in this investigation (3.6 Bq kg^{-1}).

^{137}Cs activity concentrations in cow's, sheep's and goat's milk sampled at 650 m altitude were below the detection limit. On the contrary, at 1000–1100 m altitude a significant increase of ^{137}Cs activity in sheep's (21.7 Bq kg^{-1}) and goat's milk (24.1 Bq kg^{-1}) was detected. At the same altitude ^{137}Cs activity in cow milk is equal to the activity at 650 m. It can be explained by a specific way of grazing regarding the fact that they consume soil together with grass. Ingestion of soil adhered to vegetation has been identified as a potentially important source of radionuclide to grazing animals.[4] These animal species are good indicators of radionuclide pollution of environment.

The same effect can be observed in sheep's meat. At an altitude of 200 m to 650 m, a low level of ^{137}Cs was detected. The ^{137}Cs activity concentration increased 10 times (4.5–45.8 Bq kg^{-1}) when the altitude increased to 1000–1100 m. This is the direct consequence of feeding.

Beside anthropogenic, primordial radioactivity was investigated in the food chain (Table 1.). Relatively high activities of ^{40}K was detected in soil, grass and hay. It was 6–10 times higher than in animal products milk, cheese and meat. These results are in correlation with results from Poland.[23] With increasing altitude, the activity of ^{40}K tended to decrease in soil samples. The activity concentration of ^{238}U and ^{232}Th decreased by 4 to 6 times in soil samples with altitude increase. This can be explained by the fact that soils in lower areas are submitted to more agrotechnical measures and use of fertilizers of a phosphate and

Table 2 ^{40}K, ^{238}U, ^{232}Th, ^{137}Cs activity concentration in bioindcators organisms at the Maljen Mountain (1000 m altitude) (Bq kg^{-1})^a

Samples /altitude	No. of samples	^{40}K	^{238}U	^{232}Th	^{137}Cs
Mushrooms (d.w.)^b					
Leccinium spp.	7	97.2 ± 3.3	<2.1	<0.3	93.3 ± 3.0
Lactraius Trivialis	5	118.4 ± 0.8	<3.0	<0.7	294.0 ± 8.5
Lactraius Deliciosus	6	105.5 ± 3.5	<3.2	<0.3	385 ± 11
Russula spp.	6	142.3 ± 5.8	<2.6	<1.5	258.1 ± 8.8
Game animals (f.w.)^c					
Venison	5	87.8 ± 3.1	12.2	<1.5	22.1 ± 1.5
Wild boars meat	5	48.2 ± 2.1	<6.5	<5	30.7 ± 1.8
Moss (d.w.)^b	5	191.2 ± 8.1	29.8 ± 3.2	6.4 ± 2.9	1200 ± 36

^{a} Mean ± standard deviation. ^{b} Dry weight. ^{c} Fresh weight.

potassium basis, compared to soils at higher mountains areas. The long-continued application of phosphate and potassium fertilizers can elevate ^{238}U and ^{40}K concentration in soil.[6]

The activity concentrations of the natural series of ^{232}Th in soil samples were at the normal environmental levels. The concentration of uranium and thorium in the Earth's crust is in the range of 1.1–10 ppm (13.5–123 Bq kg^{-1})[2] for ^{238}U, and 10 ppm (39.4 Bq kg^{-1})[16] for ^{232}Th. The measured activity of natural radionuclides ^{238}U and ^{232}Th in grass and hay was low. In the animal products the activity was at the detection limit.

The results obtained by gamma spectometric analysis of mushrooms, mosses and game samples collected on Maljen mountain are presented in Table 2.

The investigated samples represent environment pollution bioindicators, and increased activity of ^{137}Cs was expected. The highest activity of ^{137}Cs was found in species *Lactarius deliciosus* (385.3 Bq kg^{-1}), and it was confirmed by Spanish scientists.[3] The measured activity of ^{137}Cs at this location was lower compared to ^{137}Cs activity in mushrooms in Romania,[7] which was 20–2800 Bq kg^{-1} dry matter. Hove[13] assumed that mushrooms are the main source of radioactive contamination of ruminants in Norway. Karlen[17] indicated that the increased contamination of does which was found in August and September in 1988 was the result of mushroom consumption, which can contribute up to 20% of the rumen content. This can explain the higher level of ^{137}Cs activity concentration in doe's and wild boar's meat found on Maljen mountain.

The activity concentration of ^{137}Cs in moss samples was significantly higher (1200 Bq kg^{-1}) compared to all investigated samples in the food chain on Maljen mountain. High accumulation of ^{137}Cs in moss was reported by other scientists too.[20] This confirms that the most of the ^{137}Cs radioactive pollution of the environment remains in moss, even 20 years after the accident.

The activity level of natural radionuclides was very low, both in mushrooms and game meat, while in moss it was in range of 6.1 to 29.8 Bq kg^{-1} f.w.

The activity of natural and produced radionuclides in the environment samples from Tara mountain is shown in Table 3.

The ^{137}Cs activity concentration measured in soil samples at Tara mountain was about 50% lower compared to the activity measured at Maljen mountain. A similar conclusion can be derived for samples of fresh grass collected on Tara mountain, from 6 locations at altitude 1000–1100 m.

The level of natural radioactivity in non cultivated soil of the mountain region of Maljen and Tara, compared with values obtained from lowland region of Vojvodina is presented in Table 4. The results show that the level of natural radionuclides activity in non cultivated soil of mountains Maljen and Tara is similar, and it is two times lower compared to non cultivated soil in region of Vojvodina.

Comparing ^{137}Cs activity concentration in cow's milk from mountains Tara and Maljen, it can be observed that on Tara mountain it is on the detection limit, and on Maljen mountain it is increased 4–10 times.

The ^{137}Cs activity in sheep's milk collected on the Tara mountain was 7 times lower compared to the samples collected on Maljen mountain. The same trend repeats in sheep's milk at an altitude of 1000 m. Observed differences in ^{137}Cs activity concentration can be explained as a result of different amounts of rainfall at the time of the Chernobyl accident.

The activity levels of natural radionuclides ^{40}K, ^{238}U, ^{232}Th in the soil samples and grass at Tara mountain are at the same level as those collected at Maljen mountain.

The activity of natural and produced radionuclides in samples of moss and game meat collected on Tara mountain are shown in Table 5.

Table 4 Comparasion of the natural radioactivity of soil samples from different regions in Serbia (Bq/kg)

Radionuclide	Maljen[a]	Tara[a]	Vojvodina[5]
^{232}Th	34 ± 1	29 ± 2	53 ± 8
^{238}U	36 ± 3	30 ± 3	51 ± 9
^{40}K	297 ± 3	233 ± 6	554 ± 92

[a] Mean ± standard deviation.

Table 3 ^{40}K, ^{238}U, ^{232}Th, and ^{137}Cs activity concentration in samples of Tara Mountain (Bq kg^{-1})[a]

Samples/altitude	No. samples	^{40}K	^{238}U	^{232}Th	^{137}Cs
Soil (f.w.)[b]					
1000 m	10	71.5 ± 2.9	11.4 ± 1.3	8.3 ± 0.4	91.2 ± 3.5
1100 m	10	395 ± 11	50.1 ± 5.1	48.9 ± 2.8	104.1 ± 8.2
Grass (f.w.)[b]					
1000 m	10	257.2 ± 8.5	<0.8	1.2 ± 0.2	4.7 ± 0.8
1100 m	10	375 ± 11	<1	1.5 ± 0.2	5.4 ± 0.2
Cow milk (f.w.)[b]					
1000 m	10	125.4 ± 7.3	0.8 ± 0.2	<0.4	<0.6
1100 m	10	108.8 ± 4.2	0.6 ± 0.1	<0.2	<0.3
Sheep milk (f.w.)[b]					
1000 m	10	38.2 ± 0.9	<0.7	<0.6	2.5 ± 0.4
1100 m	10	45.8 ± 1.5	<0.5	<0.4	3.2 ± 0.5
Sheep meat (f.w.)[b]					
1000 m	10	89.3 ± 3.3	<1.3	<0.8	5.2 ± 1.1
1100 m	10	129.4 ± 4.5	<0.6	<0.1	7.8 ± 1.9

[a] Mean ± standard deviation. [b] Fresh weight.

Table 5 ^{40}K, ^{238}U, ^{232}Th, ^{226}Ra, ^{137}Cs activity concentration in bioindicators of Tara Mountain (Bq/kg)[a]

Sample	Altitude (m)	No. of samples	^{40}K	^{238}U	^{232}Th	^{137}Cs
Mosses (d.w.)[b]	1000	5	21.0 ± 1.0	4.2 ± 1.3	<0.8	574 ± 17
	1100	5	197.7 ± 5.9	22.1 ± 3.5	7.2 ± 2.3	836 ± 24
Wild boars meat (f.w.)[c]	1000	5	157.0 ± 5.1	<3.2	<0.3	54.1 ± 5.6
Venison (f.w.)[c]	1000	5	125.0 ± 4.3	<4.3	<2.5	24.0 ± 7.2

[a] Mean ± standard deviation. [b] Dry weight. [c] Fresh weight.

Table 6 Comparison of average ^{137}Cs activity in environmental samples from Maljen and Tara mountains at altitude of 1000–1100 m (Bq/kg)

Samples	Maljen	Tara
Soil (f.w.)[a]	161.5	97.6
Grass (f.w.)[a]	9.1	5.2
Cow milk (f.w.)[a]	5.7	0.4
Sheep milk (f.w.)[a]	17.0	2.8
Sheep meat (f.w.)[a]	45.8	6.5
Moss (d.w.)[b]	1200	705
Wild boars meat (f.w.)[a]	30.7	54.1
Venison (f.w.)[a]	22.1	24.0

[a] Fresh weight. [b] Dry weight.

In Table 6 are presented average ^{137}Cs activities in different environmental samples from Maljen and Tara mountains at altitude of 1000–1100 m (Bq/kg).

In moss samples collected at two different altitudes, three samples at each location, ^{137}Cs activity was from 573.7 to 836.0 Bq kg^{-1} d.m. The same results were also found in ref. 8. Comparing ^{137}Cs activity measured in the moss on two mountains in Serbia it was found that on Tara mountain activity was 1.2–4 times lower. This can be explained by different amounts of rainfall at the time of the Chernobyl accident at these locations.

The activity of natural radionuclides ^{40}K, ^{238}U, ^{232}Th in moss and game samples at both locations was in the same range.

The annual effective dose for the population in the region of the Maljen and Tara mountains (Table 7) using above presented ^{137}Cs activity in milk and milk based products, meat of domestic and game animals, and in eatable mushrooms was estimated.

Consumption of mushrooms is not a common habit in the nutrition of human population in Serbia. Therefore, there are insufficient data about the amount of human consumption of mushrooms in Serbia. The average value for forest mushroom consumption in central European countries is about 1.3 kg f.w. per person annually[24] and we used this data for calculation.

Table 7 Estimated effective dose from ^{137}Cs from consumed food (μSv)

Samples	Average annual consuming (kg)	Effective equivalent dose (μSv)	
		Maljen	Tara
Cow milk	100.0	7.4	0.5
Sheep milk	0.5	0.1	0.02
Sheep meat	3.0	1.8	0.25
Wild boars meat	1.0	0.4	0.7
Venison	1.0	0.3	0.3
Mushrooms	1.3	6.3	—

Official statistical data in Serbia about consumption of other foodstuffs per person were used in this paper.

The effective dose due to ^{137}Cs activity in the animal products is below radiological health concern, except in the cases of cow's milk (7.4 μSv) and mushrooms (6.3 μSv) at Maljen mountain.

4. Conclusion

Natural and produced radionuclide content (^{40}K, ^{238}U, ^{232}Th, ^{137}Cs) in different samples from the environment of the Maljen and Tara mountains was measured. On the basis of the obtained results it can be observed that ^{137}Cs activity increases with altitude at the Maljen mountain location. It is incorporated in food chain soil–plants–animals–humans. Increases of ^{137}Cs activity concentration in animal products is a direct result of its presence in the soil as a consequence of the Chernobyl accident. Mushrooms, mosses and game meat confirmed that they are the best bioindicators of radioactive pollution, many years after the accident. At the Maljen location it was found that increases of ^{137}Cs activity concentration in the food chain at higher altitudes is a result of increased amounts of rainfall after the Chernobyl accident. The lower level of ^{137}Cs was measured in the food chain at the Tara mountain location, which can be explained by lower amounts of rainfall after the accident. The activity level of natural radionuclides ^{40}K, ^{238}U, ^{232}Th in investigated samples was low and at the same level of activity for natural radionuclides in other countries.

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