

A REVIEW: NATURAL AND ARTIFICIAL RADIONUCLIDES AND
RADIATION HAZARD PARAMETERS IN THE SOIL OF
MOUNTAIN REGIONS IN SERBIA

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Abstract: This review paper discusses the content of natural (⁴⁰K, ²³⁸U, ²²⁶Ra, and ²³²Th) and artificial (¹³⁷Cs) radionuclides in the soil of the mountains of Maljen, Tara and Kopaonik in the Republic of Serbia over 2002–2015. In addition, the paper gives radiation hazard parameters, i.e., radium equivalent activity, absorbed dose rate, annual effective dose equivalent, external hazard index, annual gonadal dose equivalent, and excess lifetime cancer risk outdoors that we calculated from the obtained content of the natural radionuclides in the soil samples. We compared the parameters to previously published results for different parts of the country and looked into the radioecological status of the investigated areas.

Key words: soil, radionuclides, gamma spectrometry, radiation hazard parameters, mountain region.

Introduction

Ionizing radiation and radioactivity are an integral part of the environment. Natural radionuclides, characterized by a long half-life, are present in rocks, minerals and soil. These primordial terrestrial radionuclides are divided into two groups.

The first group includes three natural decay series, headed by ²³⁸U, ²³²Th and ²³⁵U, and their decay products (Atwood, 2010), which are alpha, beta and gamma emitters. In nature, uranium is present as a mixture of ²³⁸U (99%, half-life 4.5×10^9 years), ²³⁵U (0.71%, half-life 7.034×10^8 years), and ²³⁴U (0.006%, half-life 2.46×10^5 years). Thorium-232 (half-life 1.41×10^{10} years) is less radioactive than uranium but

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more abundant in nature. The most important radioactive decay isotopes are: ^{226}Ra (half-life 1602 years) that belongs to the ^{238}U series, and ^{228}Ra (half-life 5.7 years) from the ^{232}Th series. A daughter nuclide of ^{228}Ra is a radioactive gas radon, which contributes the largest dose to the radiation from natural radioactive sources.

Radionuclides in the second group are ^{40}K and ^{87}Rb . Potassium-40 (half-life 1.25×10^9 years) is a radioactive isotope (contributing 0.012%) of potassium that is abundant in nature. Rubidium-87 (half-life 4.88×10^{10} years) is more abundant in nature (27.83%) than ^{40}K . These two radionuclides present in the soil significantly contribute to the total dose of irradiation to humans and biota (Fisenne, 1993).

An important source of human exposure to radiation is the decay of naturally occurring radionuclides in the soil. Radionuclides are not uniformly distributed; their concentration depends on geographic location, the geological origin of the soil, but also anthropogenic activities. All of these factors contribute to the variation of radiation exposure. According to the UNSCEAR report (2008), the average value of worldwide exposure due to the natural radiation sources is 2.4 mSv/y, with a range of 1–10 mS/y. The population receives the highest radiation doses due to the inhalation exposure to ^{222}Rn (1.15 mSv/y), from external terrestrial radiation (0.48 mSv/y), cosmic radiation and cosmogenic radionuclides (0.39 mSv/y), and ingestion of ^{40}K and radionuclides from the uranium and thorium series (0.29 mSv/y).

Materials containing naturally occurring radionuclides and their radioactive decay products that are not disturbed by human activities are called Naturally Occurring Radioactive Material (NORM) (IAEA, 2003). Some activities, such as mining, oil and gas production, coal combustion, use of fertilizers and release of fertilizer production wastes may lead to concentrated NORM materials.

The environmental contamination of Serbia with a fission radionuclide ^{137}Cs (half-life 30 years) is a consequence of the nuclear accident in Chernobyl (in 1986) and nuclear weapons testing in the atmosphere. The ^{137}Cs activity concentration in different regions of the former Republic of Yugoslavia ranged between 1 kBq/m² and 10 kBq/m², depending on the prevailing meteorological conditions during the release (EU Commission, 1991). Due to its long half-life, ^{137}Cs can still be found in the environment, especially in the soil and bioindicator organisms, such as mosses, fungi, blueberries and wild animals (Steinnes and Njåstad, 1993; Delfanti et al., 1999; Čučulović et al., 2012; Mitrović et al., 2013b, 2014a, 2016c; Todorović et al., 2013). The retention of ^{137}Cs in the soil depends on the soil physico-chemical characteristics and its clay content, but also on the climate, surrounding vegetation, etc. (Cremers et al., 1988; Rosén et al., 1999; Fujii et al., 2014).

Our paper gives a review of the activity concentration of natural (^{40}K , ^{238}U , ^{226}Ra , and ^{232}Th) and artificial (^{137}Cs) radionuclides in the soil from the mountains of Tara, Maljen and Kopaonik, with an emphasis given to estimating the radiation hazard parameters.

Materials and Methods

The soil samples were collected over 2002–2015 in the areas of the mountains of Maljen, Tara, and Kopaonik (Figure 1). The mountains of Tara and Kopaonik are nature parks and thus all their reserves are under the state protection. The studied areas present some of favorite tourist destinations for sports and recreation.



Figure 1. A view of Serbia with the location of the mountains of Maljen, Tara and Kopaonik.

The soil samples were collected from the surface down to a depth of 20 cm. In the laboratory, the samples were crushed, dried at 105 °C, sieved, homogenized and put into Marinelli beakers (0.5 ml and 1 l). All samples were hermetically sealed and stored for 21 days to ensure equilibrium between ^{226}Ra and its daughters.

We used gamma-ray spectrometric measurements on High Purity Germanium detectors (ORTEC and Canberra), with a relative efficiency of 18%, 20% and 30%, and an energy resolution of 1.85 keV (1332.5 ^{60}Co), to determine the activity concentrations of the investigated radionuclides.

The analysis of each measured gamma-ray spectrum was performed by a software program GAMMA VISION-32. All obtained results are expressed as (mean \pm standard deviation).

The activity concentrations of ^{40}K and ^{137}Cs were derived directly from their 1460.8 keV and 661.66 keV gamma energies, respectively. For other radionuclides, we included the following steps in the analysis:

- We assumed equilibrium conditions between the parent nuclide ^{238}U and its daughters, ^{234}Th (63.2 keV) and $^{234\text{m}}\text{Pa}$ (1001 keV).
- We obtained the ^{226}Ra activity concentration from the gamma energy of 186.1 keV corrected for ^{235}U , and ^{226}Ra daughters in equilibrium, ^{214}Bi (609.3 keV, 1120.2 keV and 1764.5 keV) and ^{214}Pb (351.9 keV).
- We used three photo peaks of ^{228}Ac (338 keV, 911.2 keV, and 969 keV) for the activity concentration of ^{232}Th .

Calculation of radiation hazard parameters

Radium equivalent activity

Since the distribution of ^{226}Ra , ^{232}Th , and ^{40}K in the soil is not uniform, we used radium equivalent activity (Ra_{eq}) to compare the specific activity of materials with different amounts of ^{226}Ra , ^{232}Th , and ^{40}K . The definition of Ra_{eq} is based on an assumption that 370 Bq/kg of ^{226}Ra , 259 Bq/kg of ^{232}Th , and 4810 Bq/kg of ^{40}K produce the same gamma dose rate. We calculated Ra_{eq} using (Beretka and Mathew, 1985):

$$Ra_{eq} \text{ (Bq/kg)} = C_{\text{Ra}} + 1.43C_{\text{Th}} + 0.077C_{\text{K}} \quad (1)$$

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 converted the mean activity concentrations of ^{226}Ra , ^{232}Th , and ^{40}K into 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 (UNSCEAR, 2008):

$$\dot{D} \text{ (nGy/h)} = 0.462C_{\text{Ra}} + 0.604C_{\text{Th}} + 0.042C_{\text{K}} \quad (2)$$

Annual effective dose equivalent

To estimate the annual effective dose equivalent ($AEDE$), the conversion coefficient from the absorbed dose in the air to the effective dose must be considered. We used the absorbed dose rate data (Eq. 2), adopted the conversion factor of 0.7 Sv/Gy (UNSCEAR, 2008), and assumed that on average, people in Serbia spend 20% of their time outdoors, and thus obtained:

$$AEDE \text{ (}\mu\text{Sv/y)} = \dot{D} \text{ (nGy/h)} \times 24 \text{ (h)} \times 365 \text{ (days)} \times 0.7 \text{ (Sv/Gy)} \times 0.2 \quad (3)$$

External hazard index

Beretka and Mathew (1985) defined the external hazard index (H_{ex}) as:

$$H_{ex} = C_{\text{Ra}}/370 + C_{\text{Th}}/259 + C_{\text{K}}/4810 \quad (4)$$

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

Annual gonadal dose equivalent

We used the following equation (Arafa, 2004) 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:

$$AGDE (\mu\text{Sv/y}) = 3.09C_{\text{Ra}} + 4.18C_{\text{Th}} + 0.314C_{\text{K}} \quad (5)$$

Excess lifetime cancer risk outdoors exposure

Excess lifetime cancer risk outdoors exposure ($ELCR_{outdoor}$) gives the probability of developing cancer over a lifetime at a given exposure level. We calculated $ELCR_{outdoor}$ using (UNSCEAR, 2000; ICRP, 1990):

$$ELCR_{outdoor} = AEDE \times DL \times R_f \quad (6)$$

where DL is the duration of life (70 years average) and R_f is the risk factor (in Sv), i.e. fatal cancer risk per sievert. For stochastic effects, we used $R_f = 0.05$ mSv for the whole population (ICRP, 2007).

Results and Discussion

Mountains are complex ecological and very sensitive ecosystems. The characteristics of mountain ecosystems vary depending on altitude, climate conditions, geological characteristics, biomes, water flows, etc. Any changes in biotic or abiotic elements can cause an interruption in the ecosystem.

Specific activities of natural radionuclides in the soil depend on geological and geographical characteristics of a given region. In the mountain regions of Maljen, Tara and Kopaonik, serpentine soils are present. Characteristics of this soil type are extremely stressful for plant growth (Kostić et al., 1998; Vicić et al., 2014) – since they are susceptible to erosion, and often shallow and rocky, vegetation is usually sparse and poor (Tatić and Veljović, 1991; Jakovljević et al., 2001; Stevanović et al., 2003). The geological background of Mt. Kopaonik is represented by granites, serpentinites, slates, marbles, andesites, and limestones. The differences in geological characteristics between the investigated mountains also affect the content of radionuclides in the soil, which will be summarized below.

The presence of anthropogenic radionuclides, such as ^{137}Cs , is mainly a consequence of the nuclear accident in Chernobyl in 1986. The ensuing soil contamination was a result of a number of factors, such as a distance from the accident location, rainfall index, and soil characteristics (UNSCEAR, 2000).

The mountain of Maljen

Maljen is a mountain located in the west of Serbia. It spreads in the east-west direction for approximately 25 km. The highest peak is Kraljev sto (elevation – 1100 m a.s.l.). The geologic structure of Mt. Maljen consists of ultrabasic rocks (peridotites and serpentized peridotites). Magnesium oxide present in the soil makes this area unfavorable for plant survival (Tatić and Veljović, 1991).

The soil samples from Mt. Maljen were collected at four altitudes over 2002–2007 (Mitrović et al., 2009). Table 1 gives the gammaspectrometric results.

Table 1. ^{40}K , ^{238}U , ^{226}Ra , ^{232}Th , and ^{137}Cs activity concentrations in the soil collected at Mt. Maljen (Bq/kg)^a.

Altitude (m)	^{40}K	^{238}U	^{226}Ra	^{232}Th	^{137}Cs
200	450 ± 5	61 ± 6	54 ± 5	57 ± 2	19 ± 1
650	394 ± 5	46 ± 3	51 ± 5	42 ± 2	47 ± 1
1000	70 ± 2	11 ± 1	11 ± 1	8 ± 1	64 ± 2
1100	102 ± 1	14 ± 1	13 ± 1	11 ± 1	259 ± 1
Average	254	33	33	29	97

^aMitrović et al. (2009).

The mean activity concentrations of the radionuclides on Mt. Maljen were 254 Bq/kg for ^{40}K , 33 Bq/kg for ^{238}U , 33 Bq/kg for ^{226}Ra , 29 Bq/kg for ^{232}Th , and 97 Bq/kg for ^{137}Cs (Table 1). According to the UNSCEAR report (2000), the world average activity concentrations in the soil are 420 Bq/kg, 33 Bq/kg, 32 Bq/kg, and 45 Bq/kg for ^{40}K , ^{238}U , ^{226}Ra , and ^{232}Th , respectively. Our results showed that the content of natural radionuclides at lower altitudes was above the world average values. However, as altitude increased, the activity of natural radionuclides in the soil decreased by a factor of 4–6. This finding can be explained by different geochemical characteristics of the soil at different altitudes, and by the fact that soils in lower areas are generally subject to agro-technical measures and use of fertilizers based on phosphate and potassium (Bolca et al., 2007).

In contrast to this finding, there was a several-fold increase in the ^{137}Cs activity concentration from an altitude of 200 m (19 Bq/kg) to 1100 m (259 Bq/kg), which could be explained by an increased amount of rainfall at higher altitudes after the Chernobyl accident. Contamination of the soil with ^{137}Cs is a starting point for transfer of this radionuclide to plants and onwards through the food chain. Its presence was detected in mosses, mushrooms, game meat and milk of animals from the test sites (Mitrović et al., 2009). The highest content of ^{137}Cs was found in sheep meat (45.8 Bq/kg) and goat milk (24.1 Bq/kg) sampled from animals grazing at 1000 m a.s.l.

Based on the measured activity concentrations of natural radionuclides in the soil (Table 1), we calculated radiation hazard parameters for Mt. Maljen (Table 2). The radium equivalent activity was below 170 Bq/kg (Table 2). For comparison, a value of 370 Bq/kg is the criterion limit for Ra_{eq} in building materials (Beretka and Mathew, 1985), and its corresponding annual effective dose equivalent is 1 mSv, which, in turn, represents the dose limit for the general population. Further, the locations at lower altitudes showed an absorbed dose rate higher than the world average of 59 nGy/h (UNSCEAR, 2008). The calculated annual effective dose equivalents at lower altitudes were also higher than the world mean of 70 μ Sv/y (UNSCEAR, 2000), while the annual gonadal dose equivalent values were lower than the level of 1 mSv/y, which is a recommended limit by the International Commission on Radiological Protection (2007) for the general public. According to UNSCEAR (2000), the gonads, bone marrow, and bone surface are considered as the organs of importance since they are active. The probability of cancer incidence in a population during the lifetime due to an exposure to natural radionuclides ($ELCR_{outdoor}$) in the soil at lower altitudes was slightly higher than the world average value of 0.29×10^{-3} (UNSCEAR, 2000), but the mean $ELCR_{outdoor}$ value was lower (0.20×10^{-3}).

Table 2. Radiation hazard parameters for the soil collected at Mt. Maljen.

Altitude (m)	Ra_{eq} (Bq/kg)	\dot{D} (nGy/h)	AEDE (μ S/y)	H_{ex}	AGDE (μ Sv/y)	$ELCR_{outdoor} \times 10^{-3}$
200	169	78	95	0.46	544	0.36
650	141	65	80	0.38	455	0.30
1000	27	13	16	0.07	88	0.06
1100	37	17	21	0.10	120	0.08
Average	94	43	53	0.25	302	0.20
Average world values	370 ^a	59 ^b	70 ^c	≤ 1 ^d	1000 ^e	0.29×10^{-3} ^f

^aBeretka and Mathew (1985); ^bUNSCEAR (2008); ^cUNSCEAR (2000); ^dICRP (2000); ^eICRP (2007); ^fUNSCEAR (2000).

As a summary of the above results, the obtained external hazard index for altitudes above 200 m was less than one (Table 2), i.e., less than the limit recommended by ICRP (2000), implying that the investigated area is radioecologically safe to use.

The mountain of Tara

Farther west and south from Maljen is the mountain of Tara. It covers an area of 250 km² and geologically belongs to the Dinarides. The highest peak is Kozji rid (1591 m a.s.l.). The climate is distinctly mountainous, and the winter period is cold

with plenty of snow. The soil is mainly uncultivated, dominated by the forest ecosystem (80%). The soil is composed of various limestones (MgCO_3 , CaCO_3) and shale (Si) rocks. Forests mainly consist of beech, spruce and fir.

Table 3 gives the activity concentrations of radionuclides in the soil of Mt. Tara, while Table 4 shows the calculated radiation hazard parameters. In all soil samples, the activity concentrations of natural radionuclides were below the world average (Table 3), except the altitude of 1100 m where the content of ^{238}U , ^{226}Ra , and ^{232}Th was higher (Djurić et al., 1996; Mitrović et al., 2009; Rakić et al., 2014). The obtained results showed that the level of natural radionuclides' activity in non-cultivated soil of the mountains of Maljen and Tara was similar (Tables 1 and 3). Popović et al. (2009) reported that the mean activities of ^{226}Ra , ^{232}Th , and ^{40}K in the soil from Mt. Tara collected over a period of 1983–1993 were 42 Bq/kg (22–92 Bq/kg), 46 Bq/kg (24–69 Bq/kg) and 613 Bq/kg (356–1126 Bq/kg), respectively. Differences in radionuclides' content in the soil are a consequence of soil type. For example, higher activity concentrations of ^{40}K , ^{238}U , and ^{232}Th can be found in shale soils than in the soil composed mainly of limestone rocks (Djurić et al., 1996).

Table 3. ^{40}K , ^{238}U , ^{226}Ra , ^{232}Th and ^{137}Cs activity concentrations in the soil collected at Mt. Tara (Bq/kg).

Altitude (m)	^{40}K	^{238}U	^{226}Ra	^{232}Th	^{137}Cs	Reference
300	420 ± 25	34 ± 3	/	43 ± 3	/	Djurić et al. (1996)
1000	72 ± 3	11 ± 1	11 ± 1	8.3 ± 0.4	91 ± 4	Mitrović et al. (2009)
1000	368 ± 37	30 ± 2	/	31 ± 3	/	Djurić et al. (1996)
1082 (Site 1)	143 ± 10	/	23 ± 3	/	221 ± 6	Rakić et al. (2014)
1082 (Site 2)	237 ± 10	/	14 ± 1	/	189 ± 6	Rakić et al. (2014)
1100	395 ± 11	50 ± 5	50 ± 5	49 ± 2	104 ± 8	Mitrović et al. (2009)
Average	300	31	31	30	98	

The values of radiation hazard parameters on all sample locations were below the recommended maximum values (Tables 2 and 4). The exception again was the soil collected at 1100 m a.s.l. where absorbed dose rate (\dot{D}), annual effective dose equivalent ($AEDE$) and excess lifetime cancer risk ($ELCR_{outdoor}$) were about 1.2-fold higher than the recommended levels.

Table 4. Radiation hazard parameters for the soil collected at Mt. Tara.

Altitude (m)	Ra_{eq} (Bq/kg)	\dot{D} (nGy/h)	$AEDE$ ($\mu\text{Sv/y}$)	Hex	$AGDE$ ($\mu\text{Sv/y}^1$)	$ELCR_{outdoor} \times 10^{-3}$
1000	28	13	16	0.08	91	0.06
1100	150	69	85	0.41	483	0.32
Average	89	41	51	0.24	287	0.19

Anthropogenic ^{137}Cs was also detected in the soil (Table 3). In 1991, five years after the Chernobyl accident, the activity concentration of this radionuclide in the surface layers was significantly lower in limestone soils (88 Bq/kg) than in shale soils (462 Bq/kg) (Djurić et al., 1996; Popović et al., 1996; Popović and Spasić, 2006; Popović et al., 2009), and it is still present in the soil (Rakić et al., 2014).

Transfer of this radioisotope from the soil to plants strongly depends on the soil type and characteristics of the plant species. The study by Mitrović et al. (2009) showed that radiocesium was present in the food chain: soil-plants-animals. Between 2002 and 2007, the medium activity concentration of ^{137}Cs was 5.1 Bq/kg in the grass, 2.9 Bq/kg in sheep milk, and 6.5 Bq/kg in sheep meat. Small ruminants, such as sheep and goats, are good bioindicators for radiocesium contamination of the environment, because the ingestion of soil can be a major source of radiocontamination for free-grazing ruminants (Assimakopoulos et al., 1993).

After the Chernobyl accident, the activity concentrations of ^{134}Cs and ^{137}Cs in herbal teas from Serbia reached 5000 Bq/kg (Petrović, 2010), but results obtained 28 years later showed a significant reduction in the ^{137}Cs concentration in teas from mountain regions (Mitrović et al., 2014). In herbal teas collected from Mts Maljen and Tara, high activity concentrations of ^{137}Cs (77 Bq/kg and 73 Bq/kg, respectively) were observed in plant species *V. myrtillus* (bilberry). Some organisms, e.g. bilberry but also mosses, mushrooms and game, are good bioindicators of environmental pollution with radiocesium (Grdović et al., 2010; Mitrović et al., 2013, 2016a,b; Rakić et al., 2014).

The mountain of Kopaonik

The south-western part of Serbia is dominated by Kopaonik, which is one of the highest mountain ranges in the country. It extends in the northwest-southeast direction for 80 km, and has a width of around 40 km in the middle. The highest peak is Pančičev vrh (elevation – 2017 m a.s.l.). The Kopaonik National Park covers an area of about 12000 ha, and it is a very popular tourist destination. The climate is subalpine with the annual average precipitation rate up to 1000 mm.

The results of gamma spectrometric determination of radionuclides in the soil collected on nine locations at Mt. Kopaonik during 2013–2016 (Mitrović et al., 2014, 2016; Džoljić et al., 2017) and radiation hazard parameters are presented in Tables 5 and 6, respectively.

The mean activity concentrations of radionuclides at Mt. Kopaonik were 723 Bq/kg for ^{40}K , 78 Bq/kg for ^{238}U , 75 Bq/kg for ^{226}Ra , and 76 Bq/kg for ^{232}Th (Table 5), which is two-fold higher than the world average activity concentrations of these radionuclides in the soil (UNSCEAR, 2000). The obtained differences

between the results can be attributed to the geological structure at the sampling locations. For example, at Lisina, a sampling site with an elevation of 1300 m, the geological period belongs to the Palaeozoic, and rocks are comprised of granite, granodiorite and quartz, known for its high content of radionuclides. On the other hand, higher up at Gobelja, metamorphic rocks (crystalline shales) are found. Furthermore, the results showed an increase of natural radionuclides ^{238}U , ^{226}Ra , and ^{232}Th with altitude. An exception was the Đerekare location where somewhat higher radionuclide content was detected.

Table 5. ^{40}K , ^{238}U , ^{226}Ra , ^{232}Th and ^{137}Cs activity concentrations in the soil collected at Mt. Kopaonik (Bq/kg).

Sampling site	Altitude (m)	^{40}K	^{238}U	^{226}Ra	^{232}Th	^{137}Cs
Brus ^a	429	526 ± 16	27 ± 4	29 ± 2	32 ± 2	31 ± 1
Jošanička banja ^a	557	269 ± 8	33 ± 4	29 ± 3	32 ± 6	142 ± 4
Đerekare ^a	950	920 ± 27	70 ± 6	78 ± 6	89 ± 7	6.0 ± 0.2
Brzeće ^a	980	620 ± 19	33 ± 5	34 ± 4	36 ± 3	32 ± 1
Brezeće ^b	1100	650 ± 40	37 ± 5	30 ± 3	41 ± 3	89 ± 5
Lisina ^a	1300	730 ± 21	184 ± 10	174 ± 10	133 ± 5	79 ± 2
Suvo Rudište ^b	1700	780 ± 50	93 ± 8	88 ± 6	111 ± 7	9.8 ± 0.8
Gobelja ^a	1934	722 ± 21	62 ± 5	65 ± 4	63 ± 2	30 ± 1
Pančićev vrh ^a	2017	1291 ± 39	160 ± 10	152 ± 11	151 ± 9	4.2 ± 0.2
Average		723	78	75	76	47

^aMitrović et al. (2016); ^bDžoljić et al. (2017).

Mt. Kopaonik was under a prolonged attack during the NATO action against the Federal Republic of Yugoslavia in 1999. The bombing led to the destruction of the terrain and disruption of the forest ecosystem (UNEP/UNCHS BTF, 1999). The gamma-spectrometric analysis of the soil taken from the Gobelja location where severe bombing activity took place, showed that activity concentrations of ^{238}U and ^{226}Ra were in equilibrium, implying a natural origin of the radionuclides (Mitrović et al., 2016). This study, however, did not investigate soil samples from the bombing craters. The average activity concentration of ^{137}Cs (47 Bq/kg) on Mt. Kopaonik was two-fold lower than on Mt. Maljen (97 Bq/kg) and Mt. Tara (98 Bq/kg). These variations can be explained by a different deposition of Chernobyl radiocesium, differences in forest type (Koarashi et al., 2016), physical and chemical properties of soils (Koarashi et al., 2012), as well as the sedimentary structure of this location (Mitrović et al., 2016).

The radium equivalent activity in the soil was mostly below 370 Bq/kg (Table 6). The exceptions were the locations of Lisina and Pančićev vrh where Ra_{eq} exceeded this limit (415 Bq/kg and 458 Bq/kg, respectively). As a consequence of the high activity concentration of natural radionuclides in the soil, the values of

absorbed dose rate (\dot{D}) and annual effective dose equivalent ($AEDE$) were higher than the global average of 58 nGy/h (UNSCEAR, 2008) and 70 μ Sv/y (UNSCEAR, 2000), respectively. However, at lower altitudes, in Brus and Jošanička banja, these two parameters were below the global means (Table 6).

Table 6. Radiation hazard parameters for the soil collected in Mt. Kopaonik.

Sampling site	Altitude (m)	Ra_{eq} (Bq/kg)	\dot{D} (nGy/h)	$AEDE$ (μ Sv/y)	H_{ex}	$AGDE$ (μ Sv/y)	$ELCR_{outdoor} \times 10^{-3}$
Brus ^a	429	115	55	67	0.31	389	0.25
Jošanička banja ^a	557	95	44	54	0.26	308	0.20
Đerekare ^a	950	276	128	158	0.75	902	0.59
Brzeće ^a	980	133	63	78	0.36	450	0.29
Brezeće ^b	1100	139	66	81	0.37	468	0.31
Lisina ^a	1300	420	191	235	1.14	1323	0.89
Suvo Rudište ^b	1700	307	140	172	0.83	981	0.65
Gobelja ^a	1934	211	98	121	0.57	691	0.46
Pančičev vrh ^a	2017	467	216	264	1.26	1506	1.00
Average		240	111	137	0.65	780	0.52

^aMitrović et al. (2016); ^bDžoljić et al. (2017).

In all locations except Lisina and Pančičev vrh, the annual gonadal dose equivalent ($AGDE$) values were lower than 1 mSv/y. Similarly, the average value for the external hazard index (H_{ex}) was less than one (Table 6). Still, values higher than 1 were recorded in Lisina and Pančičev vrh. The probability of cancer incidence in a population during the lifetime due to an exposure to natural radionuclides in the soil ($ELCR_{outdoor}$) at higher altitudes was higher than the world average value of 0.29×10^{-3} (UNSCEAR, 2000).

In addition to the natural radionuclides, the artificial ^{137}Cs was detected in the soil, grass, spruce, cow milk and moss samples from Mt. Kopaonik (Mitrović et al., 2016; Džoljić et al., 2017). In the soil, the lowest activity concentration of ^{137}Cs was recorded in the area of Pančičev vrh, and the maximum in Jošanička banja (Table 5). A decrease in the ^{137}Cs content in the soil with an increasing altitude is probably due to stronger soil erosion at higher altitudes (Nešić et al., 2009).

Radionuclides in the soil of Serbia

Determination of radionuclides' content in the soil has been a subject of many studies with the purpose of risk assessment and protection of the humans, non-human biota and the environment from radioactive pollution. Studies performed by Momčilović et al. (2010) and Tanič et al. (2014) in the area of an abandoned uranium mine (Grabovnica mine) at Mt. Stara Planina showed that the terrestrial gamma dose rate due to ^{238}U (31–237 Bq/kg), ^{232}Th (6–109 Bq/kg), and ^{40}K (64–

977 Bq/kg) in the soil was two-fold higher than that of the surrounding area. In the vicinity of the mine compounds, the radionuclide content was within the usual range for this mountain or slightly higher. In a part of Mt. Stara Planina farther south, at sampling points of Smilovci, Smilovsko jezero, Kamenica, and Gornji Krivodol, the activity concentrations of natural radionuclides in the soil were lower and ranged between 393 Bq/kg and 543 Bq/kg, 29 Bq/kg and 57 Bq/kg, 25 Bq/kg and 51 Bq/kg, and 28 Bq/kg and 69 Bq/kg for ^{40}K , ^{238}U , ^{226}Ra , and ^{232}Th , respectively (Vranješ et al., 2016). Apart from the natural radionuclides, ^{137}Cs was also detected in the environment of Mt. Stara Planina, i.e. in the soil, mosses and medicinal plants (Vranješ et al., 2016).

Gamma-spectrometric determination of radionuclides present in the soil from Mt. Zlatibor (Dragović et al., 2010a) showed that the content of ^{40}K ranged between 212 Bq/kg and 740 Bq/kg, ^{238}U between 10.6 Bq/kg and 69.0 Bq/kg, ^{226}Ra between 8.3 Bq/kg and 87.5 Bq/kg, ^{232}Th between 10.4 Bq/kg and 41.4 Bq/kg, and ^{137}Cs between 73 Bq/kg and 155 Bq/kg. This study showed enrichment of ^{226}Ra with respect to ^{238}U in some soil samples, probably due to greater mobility of uranium, which, in turn, led to a depleted content of this element relative to radium. In Mt. Zlatibor, anthropogenic ^{137}Cs was detected in bioindicators, as well as ants, mosses, lichens (Dragović et al., 2010a,b) and medicinal plants (Mitrović et al., 2013, 2014, 2016).

Investigation of radionuclide content in the soil has also been conducted in urban areas (Grdović et al., 2010; Janković Mandić and Dragović, 2010; Dragović et al., 2012; Milenković et al., 2015; Mitrović et al., 2016b; Vukašinović et al., 2018), agriculture soil (Vukašinović et al., 2009, 2010; Bikit et al., 2011; Mitrović et al., 2013b; Forkapić et al., 2017) and in the vicinity of industrial plants (Vitorović et al., 2012; Vukašinović et al., 2014). Gamma-spectrometric determination of ^{238}U in the soil sampled from 21 different locations in Serbia and Montenegro showed that uranium content ranged from 1.28 ppm on Slatina (approximately 15.7 Bq/kg) to 4.80 ppm in Vranje (approximately 58.8 Bq/kg), with the mean value of 2.76 ppm (approximately 33.8 Bq/kg) (Dragović et al., 2006b). The absorbed gamma dose rates (\dot{D}) ranged between 24.5 nGy/h and 97.6 nGy/h, with the mean of 66.8 nGy/h, annual effective dose equivalent (AEDE) varied from 42.3 $\mu\text{Sv/y}$ to 119 $\mu\text{Sv/y}$, with the mean of 81.9 $\mu\text{Sv/y}$ (Dragović et al., 2006a); and the radiation hazard was insignificant for population living in the investigated areas.

It is also worth noting that many studies have been conducted to determine the uranium content in various environmental samples from locations where depleted uranium ammunition was used during the NATO bombing in 1999 (Miljević et al., 2001; Esposito et al., 2002; Popović et al., 2008, 2010; Sarap et al., 2014). These locations are now included as an integral part of the monitoring program at the territory of the Republic of Serbia (Official Gazette RS, 2010).

Conclusion

Determining natural radioactivity in the soil is a necessary step in the evaluation of the radiation hazard parameters. The average values of activity concentrations of ^{238}U , ^{226}Ra and ^{232}Th at Mts Maljen and Tara were within the range of values obtained for other parts of Serbia and worldwide (UNSCEAR, 2008). However, the measured concentrations were approximately two-fold higher at Mt. Kopaonik.

The radiation hazard parameters obtained for Mts Maljen and Tara were similar to values reported for other parts of Serbia implying that the investigated areas were radioecologically safe for humans.

Mt. Kopaonik showed the average values of absorbed dose rate, annual effective dose equivalent and external hazard index two-fold higher than at Mts Maljen and Tara. At two particular locations, Lisina and Pančičev vrh, the values of absorbed dose rate and annual effective dose equivalent were about four-fold higher than those on Mts Maljen and Tara. External hazard index was also higher than one, indicating an elevated radiation hazard.

The values of annual gonadal dose equivalent, as a measure of the genetic significance of the annual dose equivalent received by the population, were less than 1 mSv in the investigated areas, except on Mt. Kopaonik, locations Lisina and Pančičev vrh, where this parameter was higher than 1 mSv.

The calculated excess lifetime cancer risk implied that the chance of contracting cancer for residents at the study area of Mts Maljen and Tara was below the global average of 0.29×10^{-3} (UNSCEAR, 2000). However, the probability of cancer incidence on Mt. Kopaonik was higher than the world average.

Anthropogenic ^{137}Cs was detected in all investigated mountain regions of Serbia.

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PRIRODNI I PROIZVEDENI RADIONUKLIDI U ZEMLJIŠTU
PLANINSKIH REGIONA REPUBLIKE SRBIJE I
PARAMETRI RADIJACIONOG RIZIKA

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R e z i m e

U radu su prikazani rezultati gamaspektrometrijskog određivanja sadržaja prirodnih (^{40}K , ^{238}U , ^{226}Ra i ^{232}Th) i proizvedenog (^{137}Cs) radionuklida u zemljištu sa područja planina Maljen, Tara i Kopaonik. Uzorci zemljišta prikupljeni su u periodu od 2002. do 2015. godine na različitim nadmorskim visinama. Na osnovu sadržaja radionuklida u zemljištu odredili smo parametre radijacionog rizika: radijum ekvivalentnu aktivnost, jačinu doze, godišnju efektivnu dozu spoljašnjeg zračenja, indeks spoljašnjeg hazarda, godišnju gonadnu dozu i faktor rizika pojave kancera na području planinskih regiona Republike Srbije, i potom ih uporedili sa ranije objavljenim rezultatima za pojedine regione Republike Srbije.

Ključne reči: zemljište, radionuklidi, gama spektrometrija, parametri radijacionog rizika, planinski region.

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