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NATURAL RADIONUCLIDES AND ^{137}Cs IN MOSS AND LICHEN IN EASTERN SERBIA

by

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The paper presents the results of radionuclides determination in moss (*Homolothecium sp., Hypnum Cupressiforme sp.,* and *Brachythecium sp.*) and lichen (*Cladonia sp.*) sampled in the region of Eastern Serbia during 1996-2010. The activities in moss are in the range of 100-500 Bq/kg d. w. for ^{40}K, and 5-50 Bq/kg d. w. for ^{226}Ra and ^{232}Th, while the "soil-to-moss" transfer factors are 0.45 for ^{40}K, 3 for ^{226}Ra, and 0.3 for ^{232}Th. The spatial distribution of the ^{137}Cs activities is highly non-uniform; some values reach 500 Bq/kg d. w., with less than 10% of the samples, mainly the ones taken prior to 2000, with the activity above 1000 Bq/kg d. w. The variations in the content of natural radionuclides among the moss species are not significant. The frequency pattern of the activities of natural radionuclides in lichen is similar to the one in moss, but the activities in lichen are to some extent lower. The mean activity of ^{137}Cs in lichen is below 400 Bq/kg d. w. The mean activities of ^{7}Be in moss and lichen sampled in 2006 and 2008 are in the range of 41-122 Bq/kg d. w., with pronounced variations between the sampling sites.

Key words: moss, lichen, radionuclides, Eastern Serbia

INTRODUCTION

In the last several decades, moss and lichen have been established as reliable bioindicators of radioactivity in the environment, since they have no rooting system and the content of elements in them is generally due to precipitation and dry deposition. Natural radionuclides in both species are mostly accumulated by resuspension of soil and dust from soil and vegetation cover. Moss and lichen have been often used in different studies on global deposition of radionuclides from nuclear atmospheric tests, monitoring of radiocontamination following nuclear plant accidents, and radioactivity and pollution monitoring in the vicinity of uranium plants [1-7]. Further, moss and lichen have been used in studies on contamination by depleted uranium ammunition in Kosovo and Metohia and Southern Serbia [8], as well as in Bosnia and Herzegovina [9].

The moss method was first used in the Scandinavian countries as a complementary method to classic instrumentation pollution monitoring [10-12]. Today, moss biomonitoring is a part of pollution monitoring programmes in most of the European countries as it gives evidence of anthropogenic impact in urban areas due to vehicular traffic and fossil fuel combustion. The method is also used to identify sources of heavy metals pollution, such as ore exploitation, agricultural activities, *etc.* [13-15].

The lichen indicator method is suitable for detection of long-term atmospheric contamination by uranium and other heavy metals, as lichen accumulates uranium and other elements and retains them for several years after the source of pollution has been exhausted [16-20]. Concentrations of radionuclides and heavy metals are significantly higher in lichen thallus than in various organs of higher plants [21]. Moss and lichen have significantly different soil-to-plant transfer factors than higher plants: 2.3 and 43.8 for ^{226}Ra and ^{137}Cs, respectively, but only 0.5 for ^{40}K, which is less than for higher plants [22].

Potassium-40 is a primordial natural radionuclide with a long half-life of 1.25 10^9 years, and a biological half-life of 30 days. It comprises around 0.0119% of the total potassium, which is found in large amounts in soils, plants, animals and humans. It is most abundant among natural radionuclides, and it is a chemical analogue to caesium.

Natural thorium-232 is also a long lived radionuclide (half-life 1.6 10^6 years).

The half-life of radium-226, a daughter of ^{238}U, is 1600 years. Radium-226 is found in different quantities in soils and rocks containing natural uranium. Its biological half-life is up to 45 years [23].

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Beryllium-7 (half-life 53.28 days) is produced by cosmic rays in spallation processes in the upper troposphere and lower stratosphere. Its variations in mean annual concentrations in air reflect changes in its atmospheric production rate, and its seasonal patterns are correlated to the stratosphere-troposphere exchange, vertical mixing within the troposphere and precipitation. ^{7}Be is widely used as an indicator of atmospheric transport processes. In mid-latitudes, the ^{7}Be seasonal variations show a maximum in summer and a minimum in winter [24-29].

Due to its long half-life (30.2 years) and the abundance in fission processes, caesium-137 has been the most significant fission product and indicator of anthropogenic pollution in the environment. In the 1990's, concentrations of ^{137}Cs in ground level air, mainly originating from the Chernobyl nuclear accident in 1986, were the order of μBq/m^3, with maxima in summer and winter. The winter maxima were attributed to resuspension of the Chernobyl fallout, with the stratospheric contribution less significant [27, 30-32].

Prior to the nuclear plant accident in Chernobyl, the ^{137}Cs concentrations in mosses were in a wide range of values, somewhere as high as 20 kBq/kg d. w., with variations up to 80% that were more due to habitat than to differences in species [33]. In Finland, the ^{137}Cs concentrations in lichen were 2400 Bq/kg in the 1960s, around 200 Bq/kg in 1985, and reaching 25 kBq/kg in 1986/1987 [34]. In a long-term study (1987-1993), the ^{137}Cs ecological half-life of 58.6 months and 10.9 months was found for moss and lichen, respectively [35]. After 2000, the distribution of ^{137}Cs in mosses and lichens over Europe was uneven, from 10 Bq/kg d. w. to more than 1 kBq/kg d. w. [36-37].

A number of recent studies have analysed radionuclides in moss and lichen in Serbia. The ^{137}Cs activities in soils and lichen in the mountains of Serbia and Montenegro increase with altitude (from 1.2 kBq/kg d. w. to 18.6 kBq/kg) [38-40]. The activities of ^{137}Cs in moss in the Belgrade city area (mountain Avala) are significantly lower – up to 221 Bq/kg [41], while in Southern Serbia, the activities reach 578 Bq/kg [42]. Moss has also been used to estimate deposition of ^{7}Be in Serbia [43-44].

In this paper, the results of long term determination (1996-2010) of radionuclides in moss and lichen in the region of Eastern Serbia are presented. The large number of data over a long time period presented in the paper could be a significant statistics base for modelling the distribution of radionuclides in the region.

MATERIALS AND METHODS

The site

The study was performed in the region of Eastern Serbia (fig. 1). The climate in the area is continental, with cold winters and hot summers.

Figure 1. Map of the Republic of Serbia (with marked sampling sites)

The sampling area included the National Park Djerdap, the towns of Kladovo, Negotin, and Zaječar, the spas of Gamzigrad, Sokobanja, and Jošanica, and the mountains Ozren and Stara Planina (fig. 1). The Djerdap National Park is located in the north-east of Serbia, on the border with Romania. It lies along the right bank of the river Danube and includes a narrow wooded mountain region, with an altitude of 50-800 meters a. s. l. Spa Sokobanja lies between the Carpathian and Balkans mountains (Rtanj and Ozren). Ozren is a well-known health resort, rich in forests and vegetation. Spa Jošanica lies in the foot of the Bukovik Mountain, at the altitude of 200-500 meters a. s. l. Geothermal waters of Sokobanja and Jošanica spas are considered among highly radioactive waters in Serbia. Gamzigrad spa is in the vicinity of the Zaječar town (11 km), in the valley of the Black Timok river, at an altitude of 160-180 m a. s. l.

Experimental

Samples of moss and lichen were randomly collected in the region of Eastern Serbia from 1996-1998 and 2000-2010. Up to 200 samples of mosses and 30 samples of lichen (*Cladonia sp.*) were collected over the period. *Homolothecium sp., Hypnum Cupressiforme sp.,* and *Brachythecium sp.* were most frequently sampled moss species (approximately 30 samples per each species).

The samples were cleaned, dried at room temperature and homogenised, then soaked in paraffin in Marinelli vessels (1 L), and left for 30 days to reach the radioactive equilibrium. Activities of the radionuclides were determined on an HPGe/ORTEC/Ametek detector (relative efficiency 34%, resolution 1.65 keV at 1.33 MeV), and an HPGe detector (Canberra, relative efficiency 25 %, resolution of 1.95 keV at 1.33 MeV).

The total standard error of the method (including relative error in geometric efficiency estimation, photo peak counts estimation, sample volume determination, *etc.*) was estimated to about 20%. Spectral analysis was performed with the Gamma Vision 32 software package. The activities of ^{226}Ra and ^{232}Th were determined by their decay products ^{214}Bi (609.3 keV; 1120.3 keV, and 1764.5 keV), ^{214}Pb (352 keV), and ^{228}Ac (338.4 keV; 911 keV, and 968.9 keV), respectively. The activity of ^{137}Cs was determined from its 661.6 keV line. The activities of ^{40}K were determined from its 1460 keV γ-line. The activities of ^{7}Ba were determined from its 477 keV γ-line. The average counting time interval was 60.000 s. Geometric calibration was performed with different radioactive reference materials, in the sampling geometry (Marinelli 1 L): (1) Silicone Resin (Czech Metrological Inst. CMI, Cert. No. 931-OL-191-01 Type MBSS 2 (^{241}Am, ^{133}Ba, ^{109}Cd, ^{139}Ce, ^{57}Co, ^{60}Co, ^{137}Cs, ^{54}Mn, ^{113}Sn, ^{85}Sr, ^{88}Y, 980.0 g, 0.98   0,01 g/cm^3, 1000   cm^3, ref. date 1. 7. 2001); (2) Soil standard (Inst. Radiological Protection, Belgrade: QAP 9803, 95.1 g, 29. 11. 2002); (3) Vegetation (Inst. Radiological Protection, Belgrade: QAP 9709, 23.12. 2002); (4) Silicone raisin (CMI, Cert. No. 9031-OL-032/05 Type MBSS 2 (^{241}Am, ^{109}Cd, ^{139}Ce, ^{57}Co, ^{60}Co, ^{137}Cs, ^{113}Sn, ^{85}Sr, ^{88}Y, 441.0 g, 0.98   0,01 g/cm^3, 45.0   4.5 cm^3, ref. date 15. 2. 2005); (5) Silicone raisin (CMI, Cert. No. 9031-OL-159/08 Type MBSS 2 (^{241}Am, ^{133}Ba, ^{109}Cd, ^{139}Ce, ^{57}Co, ^{60}Co, ^{137}Cs, ^{54}Mn, ^{113}Sn, ^{85}Sr, ^{88}Y, 980.0 g, 0.98   0,01 g/cm^3, 1000   10 cm^3, ref. date 1. 4. 2008).

RESULTS AND DISCUSSION

Natural radionuclides in moss and lichen in Eastern Serbia, 1996-2010

The activities of natural radionuclides ^{40}K, ^{226}Ra, and ^{232}Th in moss and lichen sampled in Eastern Serbia, from 1996-1998 and 2000-2010 are presented in tab.1. Since no significant variations between the sites were found, the results are given as "mean   standard deviation" (Bq/kg dry weight), with coefficients of variation (%) in brackets.

The activities of natural radionuclides in moss in Eastern Serbia measured in 1996-1998 and in 2000-2010 were within the range of values reported for the region [39, 42, 44], but somewhat lower than the activities reported for moss sampled in urban areas [41]. This could be explained by the fact that in our study the sampling sites were not treated with fertilizers, and, therefore, the resuspended concentrations of natural radionuclides from soils, mainly ^{40}K, were lower. Higher variations in the content of natural radionuclides in moss in 1996-1998 were due to larger sampling area (mountain Stara Planina included), than later (2000-2010), when the sampling sites were more closely grouped (Gamzigrad, Sokobanja, Djerdap, Jošanica) (fig. 1). The content of natural radionuclides was to some extent lower in lichen than in moss (tab. 1), but not significantly. It should be however noted that lichen was collected from higher points (trees) and the effect of resuspension from soil was less pronounced.

Table 1. Activities of natural radiounuclides in moss and lichen, Eastern Serbia

Period		^{40}K [Bq/kg d. w.]		^{226}Ra [Bq/kg d. w.]		^{232}Th [Bq/kg d. w.]	
1996-1998	Moss	281	193 (69)	54	29 (54)	19	11 (58)
	Lichen	209	46 (22)	13	4 (31)	11	3 (27)
2000-2010	Moss	223	84 (38)	13	6 (46)	12	6 (50)
2006-2010	Lichen	207	51 (25)	12	5 (42)	9	2 (22)

The frequencies of the ^{40}K, ^{226}Ra, and ^{232}Th activities in moss in Eastern Serbia from 1996-2010 are given in figs. 2 and 3. The activities of ^{40}K in moss were mainly within the range of 100-500 Bq/kg d. w., while the majority of the ^{226}Ra and ^{232}Th activities were spread within the range of 5-50 Bq/kg d. w. (figs. 2 and 3). Less than 5% of ^{232}Th activities was in the range of 50-100 Bq/kg d. w., while ^{226}Ra activities higher than 100 Bq/kg d. w. were measured in less than 10% of the samples.

Based on our data of the mean activities of natural radionuclides in soils sampled in the immediate vicinities of the sampling sites for moss and lichen (621 Bq/kg for ^{40}K, 18 Bq/kg for ^{226}Ra and 32 Bq/kg for ^{232}Th), the soil-to-moss transfer factors were calculated: 0.45 for ^{40}K, 3 for ^{226}Ra and 0.3 for ^{232}Th. The results correspond with the data reported by other authors [22, 42].

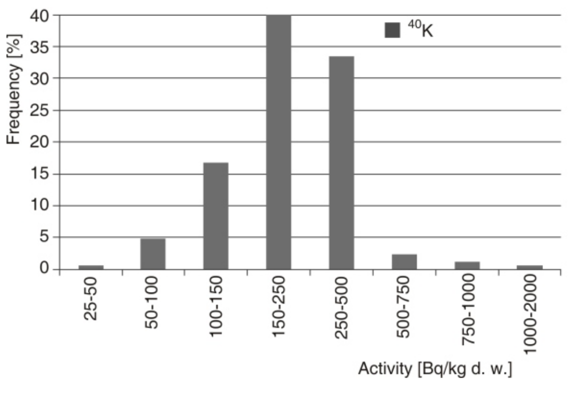

Figure 2. Frequency of ^{40}K activities [%] in moss in Eastern Serbia, 1996-2010

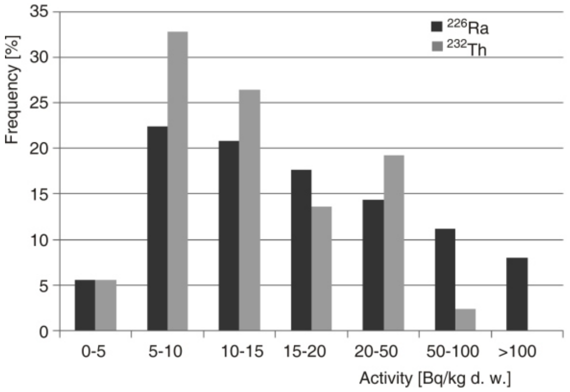

Figure 3. Frequency of ^{226}Ra and ^{232}Th activities [%] in moss in Eastern Serbia, 1996-2010

The frequencies of the activities of natural radionuclides in lichen in 1996-2010 are given in figs. 4 and 5. The frequency patterns resemble the patterns seen in moss (figs. 2 and 3), with some differences concerning ^{40}K due to several very high activities of this radionuclide (above 500 Bq/kg d. w.) measured in moss.

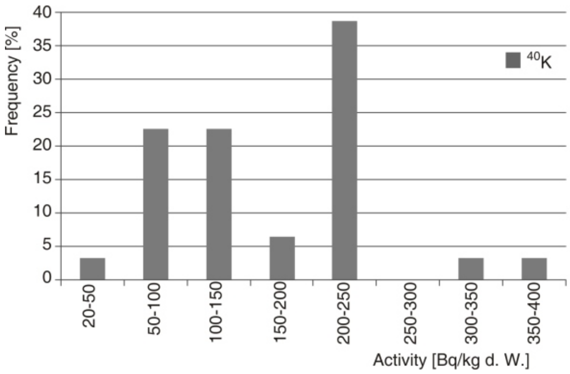

Figure 4. Frequency of ^{40}K activities [%] in lichen in Eastern Serbia, 1996-2010

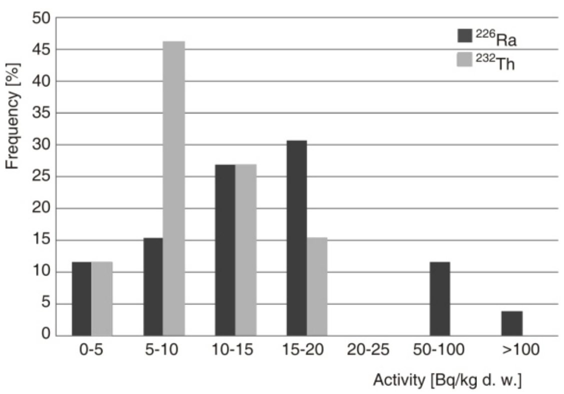

Figure 5. Frequency of ^{226}Ra and ^{232}Th activities [%] in lichen in Eastern Serbia, 1996-2010

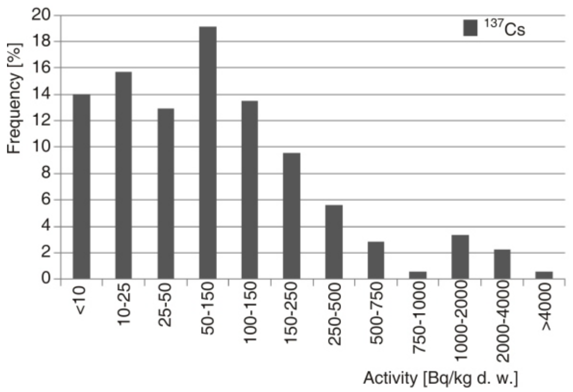

Figure 6. Frequency of ^{137}Cs activities [%] in moss in Eastern Serbia, 1996-2010

^{137}Cs in moss and lichen in Eastern Serbia, 1996-2010

The activities of ^{137}Cs measured in moss in Eastern Serbia, from 1996-2010 are presented in tab. 2. The results are given as a range of values if the activities differ more than one-fold (*i. e*., variations higher than 100%), and as "mean   standard deviation" otherwise. The coefficients of variations are given in brackets. Individual high values (max) that were not included in the calculations are also given in brackets. LLD stands for lower limit of detection.

The distribution of ^{137}Cs activities in moss confirms the non-uniform spatial distribution of this radionuclide in the region. The wide range of values and highly pronounced local variations in our study compared to some other authors over the same period [42-44], could be attributed to the fact that the sampling sites were not fertilized and subjected to extensive anthropogenic activities. The temporal decrease in the ^{137}Cs activities in moss is well pronounced, with some local exceptions.

The frequencies of the ^{137}Cs activities in moss in Eastern Serbia, from 1996-2010, are presented in fig. 6. Over the study period (15 years), the activities of ^{137}Cs in moss were measured in the wide range of LLD-500 Bq/kg d. w., with less than 10% of samples with the activity higher than 1000 Bq/kg d. w.

Table 2. Activities of ^{137}Cs in moss in Eastern Serbia, 1996-2010

Period	Site	^{137}Cs [Bq/kg d. w.]
1996-1998	Stara Planina	16-384
	Zaječar	33-748
	Kladovo/Djerdap	166-9900
2000-2005	Zaječar/Gamzigrad/ Sokobanja	6-279 (max 2365)
	Kladovo/Djerdap	LLD-745
2006-2010	Gamzigrad/Sokobanja/ Jošanica	72 45 (62.5%) (max 1239)
	Kladovo/Djerdap	LLD-223 (max 1131)

The activities of anthropogenic ^{137}Cs in lichen sampled in Eastern Serbia in 1996-1998 and 2006-2010 are presented in tab. 3. The results are given as "mean ± standard deviation", with the coefficients of variation in brackets. Higher content of caesium in lichen was measured on sites with higher content of this radionuclide in moss, but the variations of the ^{137}Cs concentrations in lichen were generally lower.

Table 3. Activities of ^{137}Cs in lichen in Eastern Serbia, 1996-2010

Period	Site	^{137}Cs [Bq/kg d. w.]
1996-1998	Eastern Serbia	233 56 (24)
2006	Sokobanja	14 9 (64)
2006-2008	Gamzigrad	164 53 (32)
2008-2010	Djerdap	283 86 (30)

The frequencies of the ^{137}Cs activity in lichen are given in fig. 7. This frequency pattern shows just few extremes (above 400 Bq/kg d. w.).

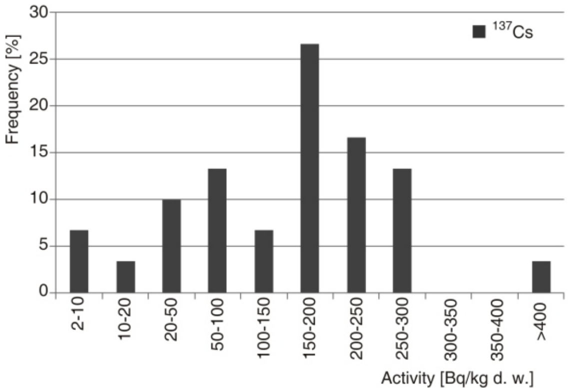

Figure 7. Frequency of ^{137}Cs activities [%] in lichen in Eastern Serbia, 2996-2010

Natural radionuclides and ^{137}Cs in moss species: *Homolothecium sp.*, *Hypnum Cupressiforme sp.*, and *Brachythecium sp.*

Concentrations of natural radionuclides and anthropogenic ^{137}Cs in most frequently sampled moss species: *Homolothecium sp.* (H), *Hypnum Cupressiforme sp.* (HC) and *Brachythecium sp.* (B) are presented in tab. 4. The results are given as a range of values if the activities differ more than one-fold (*i. e.*, variations higher than 100%), and as "mean standard deviation" otherwise. The coefficients of variations are given in brackets. LLD stands for lower limit of detection.

The variations in the content of natural radionuclides in the three moss species were in the range 48-96% (tab. 4), but the differences among the species were not significant. The concentrations of ^{137}Cs in the three moss species showed high values prior to 2000, followed by a steep decrease afterwards.

To estimate the correlation between the moss species for different radionuclides, the linear Pearson correlation coefficients for (^{226}Ra, ^{232}Th) and (^{137}Cs, ^{40}K) pairs were calculated. High correlation (0.68) was obtained between ^{226}Ra and ^{232}Th, as expected, and low (0.24) between ^{137}Cs and ^{40}K.

^{7}Be in moss and lichen in Eastern Serbia, 2006-2008

The activities of ^{7}Be in moss and lichen sampled in Eastern Serbia in 2006 and 2008 are presented in tab. 5. The activity of ^{7}Be was calculated on the date of the sampling (in May 2006 and in June 2008). The activities are presented as "mean ± standard deviation" from multiple samples taken within each year (10-17 samples of moss per year and per site, and 6 samples of lichen per year and per site were analyzed), with coefficients of variation in brackets.

The ^{7}Be concentrations from two sampling sites, both in moss and in lichen, were significantly higher (20-60%) in 2008 than in 2006 (tab. 5). The variations between the sites and within a year were also high (28-71%), mainly due to short half-life of ^{7}Be and the differences in microclimate and topology of the sites. Since the ^{7}Be activities in air were not measured simultaneously, the activities in moss and lichen could not be compared with the activities in air. Still, the ^{7}Be concentrations in our study are comparable with its activities in moss and lichens in Southern Serbia [42]. Higher values (195-560 Bq/kg d. w.), with a strongly pronounced non-uniformity in spatial distribution, of the ^{7}Be concentrations in moss (*Hypnum Cupressiforme*) in Northern and Central

Table 4. Activities of natural radionuclides and ^{137}Cs in *Homolothecium sp.* (H), *Hypnum Cupressiforme sp.* (HC), and *Brachythecium sp.* (B), in Eastern Serbia, 1996-2010

Species	Period	^{40}K [Bq/kg d. w.]	^{226}Ra [Bq/kg d. w.]	^{232}Th [Bq/kg d. w.]	Period	^{137}Cs [Bq/kg d. w.]
H	1996-2010	246 134 (55)	19 18 (95)	15 11 (73)	1997-1999	123-9990
					2000-2010	1.0-435
HC		348 190 (55)	25 24 (96)	18 11 (61)	1997-1999	271-3100
					2000-2010	15-438
B		204 97 (48)	15 9 (60)	12 7 (58)	1997-1999	LLD-1417
					2000-2010	LLD-146

Table 5. ^{7}Be in moss and lichen in Eastern Serbia, in 2006 and 2008

Sample type	Site	^{7}Be [Bq/kg d. w.]	
		2006	2008
Moss	Sokobanja	61 29 (48)	122 45 (37)
Lichen	Sokobanja	–	–
Moss	Gamzigrad	66 33 (50)	82 23 (28)
Lichen	Gamzigrad	41 29 (71)	103 43 (42)

Serbia were reported by [43]. Even higher activities of ^{7}Be in moss, up to 920 Bq/kg, were shown by [44]. Assuming that moss contains about one year of accumulated ^{7}Be from air [44], the ^{7}Be concentration in moss could provide a reliable insight of the ^{7}Be concentrations in the ground level air of the area.

CONCLUSIONS

The activities of natural radionuclides in moss in Eastern Serbia measured in 1996-2010 were within the range reported for the region, but somewhat lower than the activities reported for moss sampled in urban areas. Since the sampling site in our study was not treated with fertilizers, the resuspended concentrations of natural radionuclides from soils, mainly ^{40}K, were lower. Higher variations in the content of natural radionuclides in moss in 1996-1998 were due to the fact that the sampling area was larger, and included a mountain region, than later (2000-2010), when the sampling sites were more closely grouped. The content of natural radionuclides was to some extent less in lichen than in moss, but not significantly. It was probably due to the fact that lichen was collected from higher points (trees) and the effect of resuspension from soil was less pronounced. The frequency patterns of the natural radionuclides activities in lichen resembled the patterns seen in moss.

The distribution of the ^{137}Cs activities in moss confirmed the non-uniform spatial distribution of this radionuclide in the region. The wide range of values and highly pronounced local variations could be attributed to the fact that the sampling sites were not subjected to extensive anthropogenic activities. The temporal decrease in the ^{137}Cs activities in moss was well pronounced, with some local exceptions. Higher content of caesium in lichen was measured on sites with higher content of this radionuclide in moss.

The differences among the different moss species were not significant. However, to support this conclusion, a further analysis is needed.

The variations in the ^{7}Be concentrations, both for moss and lichen, between the sites and within each year, were high, mainly due to its short half-life and the differences in microclimate and topology of the sites.

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ПРИРОДНИ РАДИОНУКЛИДИ И ^{137}Cs У МАХОВИНАМА И ЛИШАЈЕВИМА ИСТОЧНЕ СРБИЈЕ

У раду су представљени резултати одређивања радионуклида у маховинама (*Homolothecium sp., Hypnum Cupressiforme sp*. и *Brachythecium sp.*) и лишају (*Cladonia sp*.) који су узорковани у региону Источне Србије током 1996-2010 године. Активности у маховинама износе 100-500 Bq/kg суве материје за ^{40}K, и 5-50 Bq/kg суве материје за ^{226}Ra и ^{232}Th. Добијени су трансфер фактори "земљиште-маховина": 0.45 за ^{40}K, 3 за ^{226}Ra и 0.3 и ^{232}Th. Просторна дистрибуција активности ^{137}Cs је изразито неуниформна; мање од 10% узорака, углавном оних пре 2000. године, има активност већу од 1000 Bq/kg суве материје. Варијације садржаја природних радионуклида у маховинама нису значајне. Фреквенционе криве активности природних радионуклида у лишају сличне су онима у маховинама, док су активности измерене у лишају нешто ниже. Средња активност ^{137}Cs у лишају мања је од 400 Bq/kg суве материје. Средње активности ^{7}Be у маховинама и лишају, који су узорковани током 2006. и 2008. године, износе 41-122 Bq/kg суве материје, са израженим варијацијама између локација.

Кључне речи: маховина, лишај, радионуклиди, Источна Србија