Arch. Biol. Sci., Belgrade, 66 (2), 691-700, 2014 DOI:10.2298/ABS1402691Č

THE ACTIVITY CONCENTRATIONS OF ^{40}K, ^{226}RA, ^{232}TH, ^{238}U AND ^{7}BE IN MOSS FROM SPAS IN EASTERN SERBIA IN THE PERIOD 2000-2012

ANA ČUČULOVIĆ[1], M. SABOVLJEVIĆ[2] and D. VESELINOVIĆ[3]

[1] *INEP- Institute for the Application of Nuclear Energy, University of Belgrade,* 11080 Zemun, Serbia
[2] *Institute of Botany and Garden, University of Belgrade,* 11000 Belgrade, Serbia
[3] *Faculty of Physical Chemistry, University of Belgrade,* 11001 Belgrade, Serbia

Abstract- In this work we present the activity concentrations of natural radionuclides ^{40}K, ^{226}Ra, ^{232}Th, ^{238}U and ^{7}Be in mosses. One hundred and sixty-seven moss samples were collected between 2001 and 2012 from the territory of the Sokobanja, Banja Jošanica and Gamzigradska Banja spas. They were classified into 23 species. The activity concentrations (Bq/kg) in moss from Sokobanja spa were: ^{40}K 25-427; ^{226}Ra 0.3-36; ^{232}Th 1.0-37; ^{238}U 0.4-28 and ^{7}Be 29-210; from Banja Jošanica spa they were: ^{40}K 90-242; ^{226}Ra 2.4-11.7; ^{232}Th 2.0-12.7; ^{238}U 1.6-11.3 and ^{7}Be 142-212; Gamzigradska Banja spa: ^{40}K 95-351; ^{226}Ra 8.0-21; ^{232}Th 5.1-19; ^{238}U 6.7-18 and ^{7}Be 20-144. The activity concentrations of dominant natural radionuclides (potassium, radium, thorium and uranium) in the moss samples were within the usual ranges for the territory of Serbia.

Key words: Mosses, radionuclides, eastern Serbia, radioactivity

INTRODUCTION

Primordial radionuclides found on Earth have existed in their current form since before the Earth was formed and they are the most common and most significant source of ionizing radiation in the environment, both from the viewpoint of total irradiation of the population and local high radiation doses. They have a long half-life (10^5-10^{16} years) and differ significantly in physical and geochemical properties. These radionuclides are ^{235}U, ^{238}U, ^{232}Th, ^{226}Ra, ^{222}Rn and ^{40}K. The most abundant natural radionuclide in the lithosphere is ^{87}Rb, followed by ^{232}Th, ^{40}K and ^{238}U.

Uranium is naturally found as a mixture of three long-life isotopes: ^{238}U (99.28%), ^{235}U (0.71%) and ^{234}U (0.0006%). Uranium-238 serves as a raw material for obtaining ^{239}Pu, which is used as a fission material. The physical half-life of ^{238}U is $4.5x10^9$ years, and the biological half-life is from 1-500 days (depending on the mobility of uranium compounds) (Kljajić et al., 1995). In the organism, uranium, regardless of its chemical form and exposure time, behaves as a toxin as it acts as a source of ionizing radiation and as a chemical toxic element. Uranium-238 and its products ^{226}Ra and ^{222}Rn, whose half-lives are 1 600 years and 3.8 days respectively, present the greatest danger to human health. ^{226}Ra is also present in materials originating from coal and wood burning (forest fires). Radium-226 is found in different quantities in soils and rocks containing natural uranium. Its biological half-life is about 45 years (Eisenbud and Gesell, 1997). Due to its long physical half-life, increased chemical activity, low elimination from the organism, its amount in the organism increases. Thorium-232 is the most significant member of the thorium chain of radioactive decomposition. Natural thorium-232 is also a long-lived radionuclide (half-life $1.4x10^{10}$ years). Its concentration is very small in the biosphere, due to its small specific activity and insolubility. Potassium-40 is a primordial natural radionuclide with a long half-life of $1.25x10^9$ years, and a biological half-life of 58 days. Potassium is a monovalent lithophile element under natural conditions. In nature, ^{40}K is found in a mixture with stable potassium isotopes ^{39}K and ^{41}K (^{39}K-93.08%, ^{40}K-0.1119% and ^{41}K-6.9%). Among the naturally occurring primordial radionuclides, ^{40}K ($T_{1/2}$=$1.28x10^9$ y) is very abundant in soil, as the molar fraction of ^{40}K is 0.0117. In a living organism, potassium is evenly distributed. Potassium is a chemical analogue to cesium. Among natural radionuclides, only ^{40}K is considered essential as it is part of the human organism and is under homeostatic control. ^{238}U and its products ^{226}Ra and ^{222}Rn present the greatest risk for human health (Gržetić and Jelenković, 1995; Veselinović et al., 2004; Mason, 1996; United Nations Committee, 2000).

Ionizing radiation reaching the surface of the Earth from the depth of cosmic space and the sun, has a large energy (0.01-10 GeV). These rays act on the environment in two ways: directly and indirectly, causing secondary radiation and forming cosmogenic radionuclides. Cosmogenic nuclides (^3H, ^7Be, ^{10}Be, ^{14}C, ^{26}Al, ^{32}Si, ^{36}Cl) are produced by cosmic rays in the atmosphere and are used in many research applications, such as paleoclimatology, solar activity reconstructions, dating methods, etc. (Kovaltsov and Usoskin, 2009). Due to their low concentrations, relatively short half-life and low radiation intensity, they are not very significant for total population irradiation. Berilium-7 forms under the action of cosmic radiation on oxygen and nitrogen in the atmosphere. About 75% of ^7Be is produced in the stratosphere, and about 25% in the higher layers of the troposphere (Masarik and Beer, 1999; Papastefanou et al., 1999). ^7Be aerosols remain in the stratosphere for about a year, while the retention time in the troposphere is about 6 weeks. It reaches lower layers, surface waters and seawater by rainfall and is used as a tracer when investigating movement of air mass through the atmosphere. Changes of ^7Be concentrations in surface layers of air depend on several factors: change of air mass movement speed in the tropopause, variation of vertical mass movement in the troposphere, variation in horizontal transport of air mass from medium latitudes towards Polar Regions and on the fall amount. The ^7Be radioisotope has a half-life of 53.12 days and emits a photon energy of 477.6 keV, with a relative intensity of 10.56% (Masarik and Beer, 1999; Papastefanou et al., 1999).

Mosses are bioindicators of environment pollution for radionuclides and other pollutants. They are organisms with specific ecology and biological features as compared to vascular flora. Investigation of radionuclide activity levels in mosses provides a reliable insight into the contamination degree of ecological systems by radioactive isotopes (Chakrabortty and Paratkar, 2006; Basile et al., 2008; Čučulović and Veselinović, 2008; Čučulović and Veselinović, 2009; Čučulović et al., 2010; Čučulović et al., 2012; Čučulović et al., 2012; Čučulović et al., 2010; Čučulović et al., 2012).

Many factors influence the concentration of polluting substances in moss: microclimate, plant age and morphology, amount of emitted and settled polluting substances, physicochemical properties of polluting substances, regime and form of water supply, chemical composition and pH of the substrate on which they grow. Parameters which influence the level of activity of radionuclides in plant material are the amount of released and deposited radionuclides, physicochemical properties of the radionuclides, meteorological-climatic conditions, physicochemical properties of the soil where the plant grows, the species and physiological-morphological characteristics of the plant itself and the way it is grown. Various locations are contaminated differently with radionuclides due to varying biosphere contamination. The surface of the plant leaf is also very significant for radionuclide uptake: if it is bigger, more radionuclides are retained on it. The age of the plant is also important for radionuclide accumulation: younger plants absorb a 2-3 times greater amount of radionuclides than older ones.

MATERIALS AND METHODS

Samples of mosses were collected from 2000 to 2012 in the territory of Sokobanja, Banja Jošanica and Gamzigradska banja spas. Sokobanja is located in the central part of eastern Serbia and is part of the Zaječar region. Samples from the territory of the Sokobanja municipality were taken on the following locations: in the city of Sokobanja (hotel Sunce) (locality **SB1**), Lepterija (1.5 km east of the center of Sokobanja) (**SB2**), in Soko city (400 m from Lepterija) (**SB3**), Mt. Ozren (about 4.5 km from the center of Sokobanja) (**SB4**), the Ophthalmology Hospital (5 km from Sokobanja southeast, close to the Ozren Special Hospital for treating eye problems) (**SB5**). In the period 2000-2012, samples were taken as follows: locality **SB1** 6 samples, **SB2** 41, **SB3** 1, **SB4** 72 and **SB5** 1 sample, in total 121 samples. Banja Jošanica is located in the northwestern part of the Sokobanja valley, between the western part of Mt. Rtanj and the eastern slopes of Mt.Bukovik, close to Sokobanja. In the territory of Banja Jošanica, samples were taken in the following locations: in the park (**BJ1**), close to mineral water springs (**BJ2**) in the Jošanica river banks (**BJ3**). In the period 2009-2012, in location **BJ1** 5 samples were taken, in **BJ2** 3 samples and in **BJ3** 6 samples were taken, in total 14 samples. Gamzigradska Banja is located close to Zaječar. In the territory of Gamzigradska Banja samples were taken on the left (**GB1**) and right bank of the Crni Timok River (**GB2**), near the Gamzigrad Rehabilitation Center (**GB3**), near Hotel Kastrum (**GB4**), hotel annexes (**GB5**), hydroelectric power station (HE) Gamzigrad (**GB6**) and in the archeological locality of Felix Romuliana (**GB7**). In the period 2006-2012 in locality **GB1** 7 samples were taken, in **GB2** 1, in **GB3** 5, in **GB4** 3, in **GB5** 7, in **GB6** 4 and in locality **GB7** 2 samples were taken, in total 32 samples. Selections of localities were based on the presence of large enough amounts of mosses and the absence of immediate sources of pollution.

In this work 23 moss taxa were analyzed (167 samples), that grew in the sampling localities: (**1**) *Brachythecium midleanum* (Schimp.) Schimp. (2 samples), (**2**) *Dicranum scoparium* Hedw. (3), (**3**) *Bryum argenteum* Hedw. (2), (**4**) *Ctenidium molluscum* (Hedw.) Mitt. (1), (**5**) *Tortella tortuosa* (Hedw.) Limpr. (2), (**6**) *Homalothecium philippeanum* (Spruce) Schimp. (7), (**7**) *Brachythecium rivulare* Schimp. (5), (**8**) *Syntrichia ruralis* (Hedw.) F. Weber & D. Mohr. (2), (**9**) *Plagiomnium cuspidatum* (Hedw.) T.J. Kop. (11), (**10**) *Homalothecium sericeum* (Hedw.) Schimp. (48), (**11**) *Leucodon sciuroides* ((Hedw.) Schwaegr (13), (**12**) *Homalothecium lutescens* (Hedw.) H. Rob (1), (**13**) *Hylocomium splendens* (Hedw.) Schimp. (8), (**14**) *Hypnum cupressiforme* Hedw. (13), (**15**) *Syntrichia calcicola* J. J. Amann (1), (**16**) *Anomodon attenuatus* (Hedw.) Huebener. (3), (**17**) *Brachythecium rutabulum* (Hedw.) Schimp. (4), (**18**) *Homalothecium sp.* (14), (**19**) *Grimmia pulvinata* Hedw. Sm. (6), (**20**) *Pylaisia polyantha* (Hedw.) Schimp. (9), (**21**) *Anomodon viticulosus* (Hedw.) Hook & Tayl. (9), (**22**) *Isothecium myosuroides* Brid. (1) and (**23**) *Neckera crispa* Hedw. (2). The nomenclature follows Sabovljevic et al. (2008). The vouchers are deposited in the bryophyte collection of the BEOU.

The samples were cleaned, dried at room temperature and homogenized, then soaked in paraffin in Marinelli vessels (1L), and left for 30 days to reach the radioactive equilibrium. Activities of radionuclides were determined on an HPGe-ORTEC/Ametek detector (relative efficiency 34%, resolution 1.65 keV at 1.33 MeV). The sample weight was about 0.1 kg. 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 activities of ^{40}K were determined from its 1460 keV γ-line. The activities of ^{7}Be were determined from its 477 keV γ-line. The average counting time interval was 6 x 10^4 s. Geometric calibration was performed with different radioactive reference materials, in the sampling geometry (Marinelli 1L): (1) Silicone Resin (Czech Metrological Inst. CMI, Cert. No. 931-OL-191-01 Type MBSS

Table 1. Locality (L), sampling year (Y), moss species (S), sample number (N), and activity (A) (Bq/kg) (lowest-highest (A_1-A_2), individual (A) and average (Ā)) ^{40}K, ^{226}Ra, ^{232}Th, ^{238}U and ^{7}Be in spas in eastern Serbia

L	Y	S	N	^{40}K (Bq/kg)		^{226}Ra (Bq/kg)		^{232}Th (Bq/kg)		^{238}U (Bq/kg)		^{7}Be (Bq/kg)	
				A_1-A_2; *A*	Ā	A_1-A_2; *A*	Ā	A_1-A_2; *A*	Ā	A_1-A_2; *A*	Ā	A_1-A_2; *A*	Ā
SB1	2001	**14**	2	119-319	219	18	---	15	---	---	---	---	---
	2002	**14**	1	319	---	17	---	15	---	---	---	---	---
	2006	**5**	1	250	---	8.5	---	6.7	---	---	---	48	---
	2008	**9**	2	65-148	107	5.1-5.4	5.2	5.9-5.1	5.5	8.6-10.5	9.5	126-151	139
SB2	2000	**13**	1	173	---	17	---	19	---	---	---	---	---
	2001	**13**	2	173-229	201	16-17	16.5	15	15	---	---	---	---
	2002	**13**	3	171-229	201	16-18	17	15-19	17	---	---	---	---
	2006	**4**	1	325	---	12.3	---	9.3	---	---	---	89	---
		5	1	176	---	4.6	---	5.0	---	---	---	94	---
	2007	**13**	1	202	---	---	---	---	---	---	---	---	---
	2008	**9**	2	243-352	298	8.8-10.0	9.4	5.6-8.4	7.0	8.7-10.0	9.3	108-188	148
		10	1	169	---	4.1	---	2.7	---	4.1	---	196	---
		14	1	167	---	9.1	---	4.7	---	8.8	---	83	---
		16	1	178	---	4.0	---	3.1	---	7.9	---	139	---
		17	1	219	---	8.0	---	7.3	---	11.6	---	119	---
	2009	**9**	1	200	---	6.1	---	4.9	---	7.9	---	---	---
		10	2	210-236	223	20	20	19-28	24	18-19	19	---	---
		16	1	115	---	8.8	---	6.2	---	8.2	---	---	---
	2010	**9**	2	142-186	164	8.4-12.2	10.3	4.8-5.3	5.0	7.6-8.8	8.2	121-149	135
		16	1	187	---	9.2	---	8.0	---	10.6	---	119	---
		17	2	251-323	287	20-24	22	26-31	28.5	9.2-20	14.6	33-150	92
	2012	**10**	8	101-163	122	0.3-9.7	5.8	1.5-5.9	3.2	1.0-8.6	6.7	76-144	117
		17	1	267	---	34	---	37	---	28	---	104	---
		21	5	108-145	132	2.5-7.5	5.3	1.0-5.6	3.6	3.9-9.9	7.3	70-112	92
		22	1	102	---	6.5	---	2.3	---	3.6	---	92	---
		23	2	115-117	116	1.4-7.8	4.6	1.6-3.0	2.3	0.4-8.5	4.4	74-122	98
SB3	2008	**10**	1	148	---	5.4	---	5.9	---	8.6	---	126	---

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 1.7.2001); (2) Vegetation (Inst. Radiological Protection, Belgrade: QAP 9709, 23.12.2002); (3) 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). Nuclides were identified using a library-driven search routine and quantitative analyses were carried out using the appropriate detector calibration. Radionuclide results were reported in Bq/kg on a dry weight basis.

RESULTS

Activity values of ^{40}K, ^{226}Ra, ^{232}Th, ^{238}U and ^{7}Be (Bq/kg) in mosses collected on the territory of Sokobanja, Banja Jošanica and Gamzigradska Banja spas, in the period from 2000 to 2012, are given in Table 1.

Table 1. Continued

SB4	2001	**14**	1	277	---	27	---	22	---	---	---	---	---
		15	1	414	---	32	---	34	---	---	---	---	---
	2002	**14**	1	187	---	20	---	16	---	15	---	---	---
	2006	**1**	2	123-208	166	7.4-18	12.7	8.1-16	12.0	7.4-15	11.2	35-99	67
		2	2	83-143	113	8.3-9.4	8.9	2.1-5.8	4.0	---	---	37	---
		18	2	103-188	146	10.4-13.5	12.0	10.9-19	15	12.8-14.5	13.7	---	---
		12	1	427	---	36	---	34	---	---	---	60	---
		14	4	95-209	150	7.3-12.5	9.4	6.6-14.9	11.0	8.8-16	12.4	29	---
		10	3	215-387	310	13.8-32	21	10.2-37	18	16-21	19	67-112	90
	2007	**2**	1	81	---	25	---	29	---	12.8	---	---	---
		13	1	304	---	27	---	26	---	15	---	---	---
		14	2	265-286	276	22-24	23	20-24	22	11.8-19	15.4	---	---
	2008	**6**	2	159-203	181	7.8-18	12.9	7.1-10	8.5	4.8-7.5	6.2	54-69	62
		7	2	184-239	212	8.1-12.0	10.0	9.2-10.7	10.0	8.9-9.9	9.4	110-144	127
		8	1	267	---	12.7	---	10.2	---	10.7	---	109	---
		9	1	65	---	5.1	---	8.0	---	8.0	---	79	---
		10	4	164-326	244	3.7-20	10.0	6.2-22	12.1	2.4-16	8.6	90-133	113
	2009	**6**	1	166	---	13.0	---	12.6	---	11.0	---	---	---
		7	1	25	---	8.20	---	12.8	---	8.2	---	---	---
		9	1	156	---	8.00	---	5.0	---	5.1	---	---	---
		10	5	145-255	221	4.5-19	12.1	5.6-20	11.9	4.3-19	10.7	---	----
	2010	**6**	4	159-211	186	6.0-9.6	8.1	3.8-6.8	5.8	4.3-9.16	7.7	88-128	108
		7	2	101-150	126	6.9-11.1	9.0	10.3-12.7	11.5	3.9-11.2	15.1	60	---
		9	2	141-178	160	3.0-4.3	3.6	1.8-6.4	4.1	7.9-9.0	8.5	155-190	173
		10	11	179-415	250	3.8-28	12.7	4.6-27	12.5	3.8-18	10.8	60-210	119
	2012	**8**	1	247	---	15.5	---	14.8	---	10.0	---	104	---
		10	8	56-275	172	2.3-10.4	6.6	3.6-14.2	6.5	1.7-16.7	6.9	78-116	92
		14	1	118	---	4.6	---	4.2	---	2.5	---	80	---
		21	4	132-402	219	4.5-12.7	8.7	4.0-9.1	6.7	1.6-11.8	6.1	116-132	123
SB5	2006	**18**	1	388	---	26	---	36	---	---	---	56	---
BJ1	2009	**18**	1	190	---	6.6	---	7.7	---	6.2	---	---	---
	2010	**18**	2	141-201	171	6.1-11.7	8.9	6.0-9.6	7.8	2.1-5.7	3.9	151-208	180
	2012	**14**	2	112-132	122	2.8-9.1	5.9	3.4-4.9	4.1	5.8-7.4	6.6	190-198	194
BJ2	2009	**18**	1	251	---	12.0	---	15.0	---	12.9	---	---	---
	2010	**18**	2	137-172	155	3.8-9.4	6.6	7.3-12.1	9.7	9.9-11.3	10.6	178-212	195
BJ3	2009	**18**	1	169	---	10.0	---	12.0	---	11.1	---	---	---
	2010	**18**	2	208-222	215	2.4-6.7	4.5	6.4	6.4	8.3-11.3	9.8	174-200	187
	2012	**10**	3	90-242	152	3.2-11.2	7.1	3.3-12.7	6.7	1.6-9.8	7.0	142-152	147
GB1	2006	**19**	2	134-243	189	12.1-14.7	13.4	5.1-7.1	6.4	12.9-14.9	13.9	20	---
		20	1	130	---	18	---	10.2	---	13.7	---	---	---
	2008	**19**	3	226-361	273	17-21	18	12.4-19	15.6	8.9-12.3	11.1	144	---
		20	1	123	---	16.8	---	11.7	---	9.5	---	---	---

Table 1. Continued

GB2	2006	**19**	1	95	---	8.0	---	10.0	---	7.2	---	20	---
GB3	2006	**20**	2	116-128	122	15.6-20	17.8	8.8-12.3	10.5	11.8-14.3	13.1	---	---
	2008	**20**	3	144-276	230	10.2-14.5	12.2	6.7-11.3	8.7	8.9-11.6	10.2	---	---
GB4	2006	**11**	1	224	---	15.4	---	10.0	---	7.5	---	44	---
		20	1	306	---	12.0	---	13.1	---	14.5	---	18	---
	2008	**20**	1	280	---	12.4	---	15.4	---	13.1	---	72	---
GB5	2006	**11**	2	178-226	202	18-20	19	14.6-15	14.8	16-18	17	---	---
	2008	**11**	5	196-351	249	10.1-18	14.6	6.7-17	10.4	8.2-17	11.8	60-136	110
GB6	2006	**3**	1	112	---	14.3	---	7.8	---	8.1	---	---	---
		11	2	226-239	233	15.6-17	16	6.6-12.3	9.5	7.8-14.3	11.0	81	---
	2008	**3**	1	101	---	18	---	11.5	---	9.1	---	---	---
	2009	**11**	3	209-228	216	10.7-17	15.3	7.0-16	14.3	7.1-13.9	12.8	---	---
GB7	2006	**10**	2	138-158	148	11.9-16	13.7	6.9-17	13.3	6.7-14.7	13.8	---	---

The activity concentrations of ^{40}K (Bq/kg) in mosses were: Sokobanja from 25 (**S**B4, 2009, sample 7) to 427 (**SB4**, 2006, sample 12); Banja Jošanica from 90 (**BJ3**, 2012, sample 10) to 242 (**BJ3**, 2012, sample 10) and Gamzigradska Banja from 95 (**GB2**, 2006, sample 19) to 351 (**GB5**, 2008, sample 11). (Table 1)

The activity concentrations of ^{226}Ra (Bq/kg) in mosses were: Sokobanja from 0.3 (**SB2**, 2012, sample 10) to 36 (**SB4**, 2006, sample 12); Banja Jošanica from 2.4 (**BJ3**, 2010, sample 18) to 11.7 (**BJ1**, 2010, sample 18) and Gamzigradska Banja from 8.0 (**GB2**, 2006, sample 19) to 21 (**GB1**, 2008, sample 19).

The activity concentrations of ^{232}Th (Bq/kg) in mosses were: Sokobanja from 1.0 (**SB2**, 2012, sample 21) to 37 (**SB2**, 2012, sample 17), Banja Jošanica from 2.00 (**BJ3**, 2009, sample 18) to 12.7 (**BJ3**, 2012, sample 10) and Gamzigradska Banja from 5.07 (**GB1**, 2006, sample 19) to 19 (**GB1**, 2008, sample 19).

The activity concentrations of ^{238}U (Bq/kg) in mosses from Sokobanja were from 0.4 (**SB2**, 2012, sample 23) to 28 (**SB2**, 2012, sample 17); Banja Jošanica from 1.6 (**BJ3**, 2012, sample 10) to 12.9 (**BJ3**, 2009, sample 18) and Gamzigradska Banja from 6.7 (**GB7**, 2006, sample 10) to 18 (**GB5**, 2006, sample 11), while activity concentrations of ^{7}Be (Bq/kg) in mosses from Sokobanja were from 29 (**SB4**, 2006, sample 14) to 210 (**SB4**, 2010, sample 10); Banja Jošanica from 142 (**BJ3**, 2012, sample 10) to 212 (**BJ2**, 2010, sample 18) and Gamzigradska Banja from 20 (**GB2**, 2006, sample 20 and **GB2**, 2006, 20) to 144 (**GB1**, 2008, sample 20). From Table 1 it follows that the lowest activity concentration of ^{238}U was measured in the moss sample of *Neckera crispa* (0.4 Bq/kg), while the highest activity concentration was measured in the moss sample of *Brachythecium rutabulum* (Hedw.) Schimp. (28 Bq/kg). Both species grow on the Sokobanja territory. The lowest activity concentration of ^{7}Be was measured in the moss sample of *Pylaisia polyantha* (Hedw.) Schimp. (20 Bq/kg), while the highest activity concentration was measured in the moss sample of *Homalothecium sp. (14)* (212 Bq/kg).

In the period 2000-2012 the average activity concentration of ^{40}K (Bq/kg) in mosses from Sokobanja was 226 ± 73, from Banja Jošanica 178 ± 40 and from Gamzigradska Banja 194 ± 63 (Fig. 1, Tables 1, 2 and 3). In the same period the average activity concentration of ^{226}Ra and ^{232}Th (Bq/kg) in mosses from Sokobanja was 13.4 ± 6.8, i.e. 13.0 ± 8.1, from Banja Jošanica 7.7 ± 2.4, i.e. 7.4 ± 3.9 and from Gamzigradska Banja 14.9 ± 3.0, i.e. 11.5 ± 2.6. The average activity concentration of ^{238}U (Bq/kg) in mosses from Sokobanja was 10.1 ± 2.8, from Banja Jošanica 8.5 ± 3.0 and from Gamzigradska Banja 11.6 ± 2.4, while ^{7}Be (Bq/kg) in mosses from Sokobanja was 102±27, Banja Jošanica 181±19.7 and Gamzigradska Banja

Table 2. Average activities of ^{40}K, ^{226}Ra, ^{232}Th, ^{238}U and ^{7}Be (Bq/kg), standard deviation in moss samples collected on localities **SB1**, **SB2**, **SB3**, **SB4** and **SB5** in the period 2000-2012

YEAR	Radionuclides				
	^{40}K	^{226}Ra	^{232}Th	^{238}U	^{7}Be
	(Bq/kg)				
	SB1				
2001	319	18	15	---	---
2002	320	17	15	---	---
2006	250	8.5	6.6	---	48
2008	107 ± 59	5.2 ± 0.2	5.5 ± 0.6	9.5 ± 1.4	---
	SB2				
2000	173	17	19	---	---
2001	201 ± 29	16.5 ± 0.7	15	---	---
2006	250 ± 105	8.4 ± 5.5	7.2 ± 3.0	---	92 ± 3
2008	221 ± 71	7.3 ± 2.6	5.3 ± 2.3	8.5 ± 2.5	139 ± 39
2009	190 ± 52	13.8 ± 7.3	14.5 ± 11.0	13.3 ± 6.05	---
2010	218 ± 71	20 ± 15	16.0 ± 15.4	15.2 ± 13.9	114 ± 48
2012	131 ± 39	7.2 ± 7.4	5.2 ± 8.3	7.7 ± 6.0	105 ± 23
	SB3				
2008	148	5.4	5.9	8.6	126
	SB4				
2000	---	---	---	---	---
2001	346 ± 97	29.5 ± 3.5	28 ± 8.5	---	---
2006	208 ± 110	15.6 ± 9.7	14.8 ± 10.4	13.9 ± 4.3	67 ± 31
2008	229 ± 52	10.4 ± 5.3	10.4 ± 4.7	8.4± 3.6	102 ± 28
2009	194 ± 52	11.2 ± 4.4	11.2 ± 4.7	9.7 ± 3.7	---
2010	216 ± 69	10.5 ± 6.6	10.3 ± 6.7	9.7 ± 4.1	120 ± 43
2012	188 ± 79	7.9 ± 3.4	7.1 ± 3.5	6.6 ± 4.2	102 ± 21
	SB5				
2008	388	26	36	---	56

Table 3. Average activities of ^{40}K, ^{226}Ra, ^{232}Th, ^{238}U and ^{7}Be (Bq/kg), standard deviation in moss samples collected on localities **BJ1**, **BJ2** and **BJ3** in the period 2009-2012

YEAR	Radionuclides				
	^{40}K	^{226}Ra	^{232}Th	^{238}U	^{7}Be
	(Bq/kg)				
	BJ1				
2009	190	6.6	7.7	6.2	---
2010	171 ± 42	8.9 ± 4.0	7.8 ± 2.6	3.9 ± 2.5	180
2012	122 ± 14	5.9 ± 4.5	4.1 ± 1.0	6.6 ± 1.1	194 ± 6
	BJ2				
2009	251	12.0	15.0	12.9	---
2010	155 ± 25	6.6 ± 3.9	9.7 ± 3.4	10.6 ± 1.0	195
	BJ3				
2009	169	10.0	12.00	11.1	---
2010	215 ± 10	4.5 ± 3.0	6.4	9.8 ± 2.1	187
2012	152 ± 80	7.1 ± 4.0	6.7 ± 5.2	7.0 ± 4.7	147 ± 5

Table 4. Average activities of ^{40}K, ^{226}Ra, ^{232}Th, ^{238}U and ^{7}Be (Bq/kg), standard deviation in moss samples collected on localities **GB1**, **GB2**, **GB3**, **GB4**, **GB5**, **GB6** and **GB7** in the period 2006-2009

YEAR	Radionuclides				
	^{40}K	^{226}Ra	^{232}Th	^{238}U	^{7}Be
	(Bq/kg)				
	GB1				
2006	169 ± 64	14.9 ± 2.96	7.5 ± 2.6	13.8 ± 1.0	20
2008	253 ± 102	18.0 ± 2.0	14.7 ± 3.3	10.7 ± 1.7	144
	GB2				
2006	95	8.0	10.0	7.2	20
	GB3				
2006	122 ± 8	17.8 ± 3.1	10.5 ± 2.5	13.1 ± 1.8	---
2008	230 ± 75	12.2 ± 2.2	8.7 ± 2.4	10.2 ± 1.4	---
	GB4				
2006	265 ± 58	13.7 ± 2.4	11.5 ± 2.2	11.0 ± 4.9	44
2008	280	12.4	15.4	13.1	72
	GB5				
2006	202 ± 34	19.0 ± 1.0	14.8 ± 0.28	17.0 ± 1.0	81
2008	249 ± 59	14.7 ± 3.1	10.4 ± 4.1	11.8 ± 3.8	110 ± 35
	GB6				
2006	192 ± 70	15.6 ± 1.4	8.9 ± 3.0	10.1 ± 3.7	216 ± 24
2008	101	18.0	11.5	9.1	---
2009	216 ± 10	15.3 ± 3.9	14.3 ± 2.2	12.8 ± 1.4	---
	GB7				
2006	148 ± 14	14.0 ± 2.9	11.9 ± 7.2	10.7 ± 5.7	---

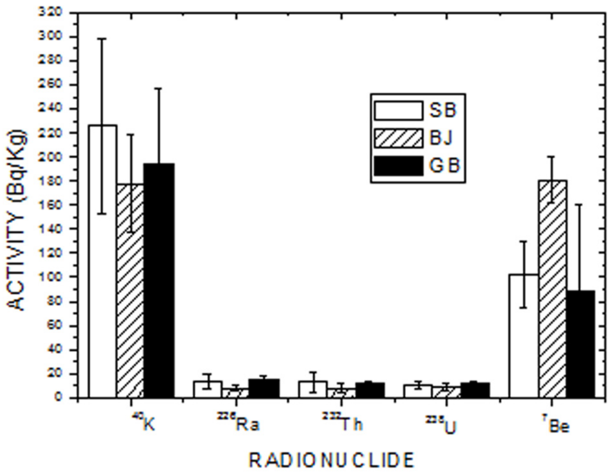

Fig. 1. Average activity concentrations of ^{40}K, ^{226}Ra, ^{232}Th, ^{238}U and ^{7}Be (Bq/kg) their standard deviations in moss samples collected in the period 2000-2012 in Sokobanja, Banja Jošanica and Gamzigradska banja

89±72. Activity concentrations of natural radionuclides ^{40}K, ^{226}Ra, ^{232}Th and ^{238}U were higher in mosses from Sokobanja and Gamzigradska Banja than in mosses from Banja Jošanica.

The activities of natural radionuclides were equilibrated, while the activity concentrations of all of the investigated radionuclides were in the range of already reported measurements in Serbia (Popović et al., 2009; Grdović et al., 2010; Dragović et al., 2010; Krmar et al. 2013).

DISCUSSION

Natural radionuclides ^{40}K, ^{226}Ra, ^{232}Th, ^{238}U and ^{7}Be were detected in all investigated samples. The results of these investigations confirmed that mosses are good indicators of environmental pollution with radionuclides.

Previous investigations have shown that average activity concentrations of ^{40}K in moss samples in Serbia were: Belgrade 245 Bq/kg (Popović et al., 2009);

Belgrade surrounding area: 294 Bq/kg (110-490 Bq/kg) (Grdović et al., 2010); Borovac (southern Serbia): 178-298 Bq/kg (Popović, et al. 2009); Zlatibor: 44.5-692 Bq/kg (Dragović et al., 2010). In our work, the lowest activity concentration of ^{40}K was measured in *Brachythecium rivulare* moss (25 Bq/kg), while the highest was measured in *Homalothecium lutescens* moss (427 Bq/kg), which grow on the territory of Sokobanja, more precisely Mt. Ozren.

Other investigations have shown that the average activity concentrations of ^{226}Ra in mosses in Serbia were as follows: in the Belgrade surrounding area: 6.1-189 Bq/kg (Grdović et al., 2010); Zlatibor: 0.9-25.8 Bq/kg (Dragović et al., 2010); Vojvodina: 2.2-36 Bq/kg (Krmar et al., 2013). The lowest activity concentration of ^{226}Ra was measured in *Homalothecium sericeum* moss (0.3 Bq/kg), while the highest was measured in *Homalothecium lutescens* moss (36 Bq/kg), which grow on the territory of Sokobanja.

Other investigations have shown that the average activity concentrations of ^{232}Th in moss from: the Belgrade surrounding area were up to 45 Bq/kg (Grdović et al., 2010), Zlatibor 0.8-13.7 Bq/kg (Dragović et al., 2010)) and Vojvodina up to 20 Bq/kg (Krmar et al. 2013), while the average activity concentrations of ^{238}U in moss from the Belgrade surrounding area were up to 113 Bq/kg (Grdović et al., 2010), Zlatibor 1.7-25.1 Bq/kg (Dragović et al., 2010). The average activity concentrations of ^{7}Be in *Hypnum cupressiforme* moss collected on the territory of Vojvodina were from 201 to 920 Bq/kg. These depend on the season when the moss was sampled and are higher in the summer than the winter (Krmar et al. 2009; Krmar et al., 2013).

From this work, it follows that the lowest activity concentration of ^{232}Th was measured in *Anomodon viticulosus* moss (1.0 Bq/kg), while the highest value was measured in *Brachythecium rutabulum* (37 Bq/kg), growing on the territory of Sokobanja. It also follows that the lowest activity concentration of ^{238}U was measured in *Neckera crispa* moss (0.4 Bq/kg), while the highest value was measured in *Brachythecium rutabulum* (28 Bq/kg), also from the territory of Sokobanja. The lowest activity concentration of ^{7}Be was measured in *Pylaisia polyantha* moss (20 Bq/kg), while the highest value was measured in *Homalothecium sp.* (14) moss (212 Bq/kg).

The results obtained in this work indicate that the measured activity concentrations of dominant radionuclides (potassium, radium, thorium and uranium) in moss samples from eastern Serbia are within the observed range for the territory of Serbia

Acknowledgments -This work was supported by the Ministry of Education, Science and Technological Development of the Republic of Serbia, project code III 43009

REFERENCES

Basile, A., Sorbo, S., Aprile, G., Conte, B. and *R. Castaldo Cobianchi* (2008). Comparison of the heavy metal bioaccumulation capacity of an epiphytic moss and epiphytic lichen. *Environ. Pollut.* **151**, 401-407.

Boileau, L.J.R., Beckett, P.J. and *D.H.S. Richardson* (1982). Lichens and mosses as monitors of industrial activity associated with uranium mining in northern Ontario, Canada-part-1: field procedures, chemical analysis and inter-species comparisons. *Environ. Pollut.* 4, 69-84.

Čučulović, A. and *D. Veselinović* (2008). Radioaktivnost nekih bioindikatora sakupljenih na teritoriji Sokobanje i njene okoline. *Ekologica*. **15**, 87-91.

Čučulović, A. and *D. Veselinović* (2009). Nivoi aktivnosti ^{137}Cs, ^{40}K i ^{7}Be u uzorcima mahovina sa teritorije Gamzigradske banje. *Ekologica*. **16,** 371-374.

Čučulović, A., Čučulović, R., Marković, D. and *D. Veselinović* (2010). Mahovine Sokobanje i Banje Jošanice – biomonitori zagađenja životne sredine radionuklidima. *Ekologica.* **17**, 135-139.

Čučulović, A., Popović, D., Čučulović, R. and J. Ajtić (2012). Natural radionuclides and ^{137}Cs in moss and lichen in Eastern Serbia. *Nucl Technol Radiat*, Belgrade, **27** (1), 44-51.

Čučulović, A., Čučulović, R., Sabovljević, M. and D. Veselinović (2012). Activity concentrations of ^{137}Cs and ^{40}K in mosses from spas in Eastern Serbia. *Arch. Biol. Sci.*, Belgrade, **64**(3), 917-925.

Čučulović, A., Čučulović, R. and D. Veselinović (2012). Activity levels of ^{137}Cs, ^{40}K, ^{226}Ra and ^{232}Th in mosses from the territory Sokobanja and Banja Jošanica, XX International Scientific and Professional Meeting Ecological Truth, Eco-Ist 12, Proceedings, Zaječar, 135-140.

Dragović, S., Mihailović, N. and B. Gajić (2010). Quantification of transfer of ^{238}U, ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs in mosses of a semi-natural ecosystem. *J. Environ. Radioact*. **101,** 159-164.

Eisenbud, M. and T. Gesell (1997). Environmental Radioactivity from Natural., Industrial and Military Sources, Academic Press, Oxford, UK.

Grdović, S., Vitorović, G., Mitrović, B. Andrić, V., Petrujkić, B. and *M. Obradović* (2010). Natural and anthropogenic radioactivity of feedstuffs, mosses and soil in the Belgrade environment, Serbia. *Arch. Biol. Sci*., Belgrade, **62** (2) 301-307.

Gržetić, I. and *R.J. Jelenković* (1995). Prirodni radioaktivni elementi, geološko poreklo i oblici pojavljivanja i migracije, monografija: Jonizujuća zračenja iz prirode, Jugoslovensko društvo za zaštitu od zračenja, INN Vinča, Beograd, 3-39.

Kljajić, R., Šipka, V., Radenković, M. and R.Mitrović (1995). Ugljevi I mineralna đubriva kao izvor tehnološkog povećanja prirodne radioaktivnosti, monografija: Jonizujuća zračenja iz prirode, Jugoslovensko društvo za zaštitu od zračenja, INN Vinča, Beograd, 95-111.

Kovaltsov, G.A. and I.G. Usoskin (2009). Test of production models for Beryllium cosmogenic radionuclides in the Earths atmosphere. Proceedings of the 31st ICRC, ŁÓDŹ, 1-4.

Krmar, M., Radanović, D., Mihailović, D., Lalić, B., Slivka, J. and I. Bikit (2009). Temporal variations of ^{7}Be, ^{210}Pb and ^{137}Cs in moss samples over 14 month period. *Appl. Radiation. Isotop*.**69**, 1139-1147.

Krmar, M., Wattanavatee, K., Radinović, D., Slivka, J., Bhongsuwan, T., Frontasyeva, M.V. and S.S. Pavlov (2013). Airborne radionuclides in mosses collected at different latitudes. *J. Environ. Radioact.* **117**, 45-48.

Veselinović, D., Pavlović, B., Pešić, D. and B. Radak (2004). Veličine, jedinice i oznake međunarodnog sistema SI u fizičkoj hemiji i srodnim oblastima, Fakultet za fizičku hemiju, Velarta, Beograd.

Masarik, J. and J. Beer, (1999). Simulation of particle fluxes and cosmogenic nuclide production in the Earth's atmosphere. *J.Geophys. Res*., **104**, 12099-12111.

Mason, B. (1996). Principles of Geochemistry. New York, J. Wiley & Sons, Inc.

Papastefanou, C., Manolopoulu, M., Stoulos, S., Ioannidou, A. and E. Gerasopoulos (1999). Soil-to-plant transfer of ^{137}Cs, ^{40}K and ^{7}Be. *J. Environ. Radioact*., **45**, 59-65.

Popović, D., Todorović, D., Ajtić, J. and Nikolić J. (2009), Active biomonitoring of air radioactivity in urban areas. *Nucl Technol Radiat*, **2**, 100-102.

United Nations Committee (2000). Effects of Atomic Radiation, Sources and effects of ionizing radiation. United Nations Scientific Committee on the Effects of Atomic Radiation. UNSCEAR 2000 report to the general assembly, with scientific annexes, United Nations, New York.

Veselinović, D. and A. Čučulović (2009). Nivoi aktivnosti ^{137}Cs, ^{7}Be i ^{40}K u uzorcima mahovina sa teritorije Sokobanje, Naučno stručni skup sa međunarodnim učešćem: Zaštita i zdravlje na radu i zaštita životne sredine, Zbornik radova, Banja Luka, 627-633.