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# Natural radionuclide uptake by mosses in eastern Serbia in 2008-2013

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The results of the study on natural radionuclide content in 102 samples of the moss species randomly collected in 2008-2013 at 30 locations of eastern Serbia are presented in the paper. The activity concentration values of $^{238}$U, $^{226}$Ra, $^{232}$Th, $^{40}$K, and $^{7}$Be determined by gamma spectrometry were within the intervals: $^{238}$U (1.1–50) Bq kg$^{-1}$, $^{226}$Ra (1.1–41) Bq kg$^{-1}$, $^{232}$Th (1.4–28) Bq kg$^{-1}$, $^{40}$K (64–484) Bq kg$^{-1}$ and $^{7}$Be (88–227) Bq kg$^{-1}$, not standing out of the average data reported for this region. The distribution of the obtained data for $^{226}$Ra, $^{232}$Th, and $^{238}$U activity concentration in the analysed mosses has shown values up to 10 Bq kg$^{-1}$ with frequencies 47.1 %, 54.9 % and 48.0 %, respectively. The obtained activity concentration values of primordial $^{40}$K and cosmogenic radionuclide $^{7}$Be were up to 500 Bq kg$^{-1}$ and about 90 % of all the results for $^{7}$Be uptake by mosses were in the 200-250 Bq kg$^{-1}$ concentration range.

KEY WORDS: *$^{238}$U; $^{226}$Ra; $^{232}$Th; $^{40}$K; $^{7}$Be; activity concentrations; bioindicators; gamma-spectrometry; mosses*

The feature of living organisms to collect quantitative information on the physico-chemical characteristics of the biosphere makes them a good indicator of changes in the ecological status of certain site or area. Basic characteristics of such organisms, bioindicators, are the ability to accumulate several or selected elements with low sensitivity, wide distribution and presence in large amounts in various environments, no seasonal differences in availability and applicability, and the existence of correlation between accumulation and input to the ecosystem (1-3). The most common representatives of bioindicators are mosses. Mosses adopt nutrients from the atmosphere by dry and moist desorption through weakly or no developed cuticles, which makes them good bioindicators of environmental pollution with heavy metals, pesticides, and radionuclides (4-7). Having no real roots, leaves, and stem with large surface area in relation to their weight, mosses grow slowly with minimal morphological changes during the lifespan. They are widespread, long-lived, and easy accessible, which makes them suitable for applications as pollution indicators (8-12).

From the Earth's genesis, natural radioactivity has been omnipresent in the environment in different amounts and it is a source of continuous exposure to living organisms. Primordial natural radionuclides such as $^{238}$U, $^{226}$Ra, $^{232}$Th, and $^{40}$K have a long physical half-life and significantly differ in physical and geochemical properties from other radionuclides. Uranium is a mixture of three long-lived isotopes: $^{238}$U, $^{235}$U, and $^{234}$U, the most abandoned $^{238}$U having a half-life time 4.5x10$^{9}$ years. Unlike the partially soluble $^{238}$U, its progeny $^{226}$Ra with half-life time 1.6x10$^{3}$ years easily enters plants from soil and transfers to animals and humans. In an organism it behaves similarly to calcium and has high chemical activity. Natural $^{232}$Th is a long-lived radionuclide with half-life 1.6x10$^{6}$ years, present in the biosphere in low content due to its insolubility. Radionuclide $^{40}$K with a half-life of 1.25x10$^{9}$ years is the most abundant among natural radionuclides with caesium as the chemical analogue (13).

Terrestrial radionuclides $^{238}$U, $^{232}$Th, and $^{40}$K are present more homogeneously in the environment than manmade ones with worldwide average activity concentrations in soil 400 Bq kg$^{-1}$, 30 Bq kg$^{-1}$, and 35 Bq kg$^{-1}$, respectively (13, 14). Their uptake by plants depends upon different factors such as soil type, pH, texture, conductivity, carbonate and sulphite contents, etc. Uptake by mosses occurs mainly through the absorption of soil particles suspended in air and transmitted by wind, raindrops containing soil particles or atmospheric deposition of these radionuclides released by fossil fuel combustion in urban areas. For these reasons, the concentrations of natural radionuclides in mosses are only indirectly related to those in soil but are more related

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to air radionuclide content, which makes them good bioindicators of air pollution. In regard to moss metabolic processes, thorium and uranium are not relevant, but potassium is an essential component (8, 9).

Radionuclide $^{7}$Be is produced by cosmic rays in spallation processes in the upper troposphere and lower stratosphere. Its variations in annual mean concentrations in air reflect changes in the atmospheric production rate, and its seasonal patterns are correlated to the stratosphere–troposphere exchange, vertical mixing within the troposphere and precipitation. Thus, $^{7}$Be is widely used as an indicator of atmospheric transport processes. In mid-latitudes, the $^{7}$Be's seasonal variations show a maximum in summer and a minimum in winter and it is certainly one of the radionuclides regularly taken by mosses (15-18). Although extensive research on radionuclides in air and bioindicators due to the contamination from nuclear probes and reactor breakdowns is available, there are limited data on natural radionuclides content in air or mosses especially for rural areas (19-23). The purpose of this paper is to present activity concentrations of natural radionuclides in different moss species collected at the territory of eastern Serbia in the period 2008-2013.

## MATERIALS AND METHODS

Moss sampling was performed during 2008-2011 and 2013 when 102 samples of 30 various moss species were collected at 30 localities of eastern Serbia region. The localities were: Đurakovo (1), Golubac (2), Brnjica (3), Lepenski vir (4), Dobra (5), Donji Milanovac (6), Miroč (7), Radujevac (8), Prahovo (9), Kladovo (10), Karataš (11), Negotin (12), Bukovo (13), Badnjevo (14), Kanjon Vratne (15), Šarbanovac (16), Štubik (17), Manastir Gornjak-Krepoljin (18), Zlotska (Lazareva) pećina (19), Vrelo Mlave Žagubica (20), Tilva Njagra (21), Dubašnica (22), Porečke šume (23), Porečka bara (24), Zlatica (25), Bela reka (26), Kožica (27), Leva reka (28), Desna reka (29), and Čezava (30). The moss species sampled for analysis were: *Hypnum cupressiforme* Hedw. *(1), Homalothecium lutescens* (Hedw.) H. Rob. *(2), Brachythecium rutabulum* (Hedw.) Schimp. *(3), Homalothecium sericeum* (Hedw.) Schimp. *(4), Brachythecium salebrosum* (Hoffm. Ex. F. Weber&D. Mohr) Schimp. *(5), Isotheciym myosuroides* Brid. *(6), Leucodon sciuroides* (Hedw.) Schwaegr *(7), Bryoerythrophyllum recurvirostrum* (Hedw.) P.C. Chen *(8), Bryum funcki* Schwaegr. *(9), Homalothecium phillippeanum* (Spruce) Schimp *(10), Syntrichia ruralis* (Hedw.) F. Weber & D. Mohr *(11), Neckera complanata* (Hedw.) Huebener *(12), Dicranum scoparium* Hedw. *(13), Leucobryum glaucum* (Hedw.) Angst. *(14), Amblystegium serpens* (Hedw.) Schimp. *(15), Grimmia trichophylla* Grev. *(16), Oxyrrhynchium hians* (Hedw.) Loeske *(17), Brachytheciastrum velutinum* (Hedw.) Ignatov & Huttunen *(18), Orthorichum anomalum Hedw. (19), Ceratodon purpureus* (*Hedw.*) *Brid (20), Kindbergia praelonga* (Hedw.) Ochyra *(21), Plagiomnium undilatum* (Hedw.) T.J. Kop *(22), Anomodon viticulosus* (Hedw.) Hook & Tayl. *(23), Anomodon attenuatus* (Hedw.) Hueb. *(24), Grimmia pulvinata* (Hedw.) Sm. *(25), Bryum capillare* Hedw. *(26), Abietinella abietina* (Hedw.) Fleisch. *(27), Polytrichum juniperinum* Hedw. *(28), Pogonatum urnigerum* Hedw. *(29), Dicranella heteromalla* (Hedw.) Schimp. *(30*). Sampling was performed regularly in summer seasons (July and August) and selection was done based on the most abandoned species existing at certain locality.

Prior to gamma spectrometry analysis moss samples were cleaned from soil particles, dried at room temperature, and homogenised. Prepared in such way, each sampled moss species was put separately into a Marinelli beaker and sealed by paraffin for 40 days to reach radioactive equilibrium between radium and its progenies. The quantity of about 100.00 g of dried sample was enough to fill 1 L volume of the Marinelli beaker. Specific activities (Bq kg$^{-1}$) of radionuclides were determined using HPGe (ORTEC/Ametek) detector with relative efficiency of 34 % and 1.65 keV resolution at 1.33 MeV. Gamma-spectra analysis was performed with the Gamma Vision 32 software package. The activity concentrations of $^{226}$Ra were determined by its decay products $^{214}$Bi (609.3 keV; 1120.3 keV and 1764.5 keV), and $^{214}$Pb (295 keV; 352 keV). The activity concentration of $^{232}$Th was determined on the basis of 338.4 keV line; $^{228}$Ac on the basis of 911 keV and 968.9 keV lines; and $^{238}$U on the basis of 63.2 keV and 1001 keV lines. $^{40}$K was determined based on 1460 keV line. The activity concentration of $^{7}$Be was determined at 477 keV line with correction to the time elapsed between sampling and measurement. The average counting time interval was 6x10$^{4}$ s. Detector calibration was performed using three different radioactive reference materials in the Marinelli geometry: 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±10) cm$^{3}$, ref. date 1.7.2001]; 2) Vegetation (Inst. Radiological Protection, Belgrade: QAP 9709, 23.12.2002), and 3) Silicone resin [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; (0.98±0.01) g cm$^{-3}$; (1000±10) cm$^{3}$, ref. date 1.4.2008]. The total uncertainty budget included geometric efficiency estimation, photo-peak counts estimation, sample volume determination, counting time, etc. and was determined to be no more than 20 %. The results of gamma-spectrometry measurements were obtained as activity concentrations in Bq kg$^{-1}$ on a moss dry weight basis. Measurements were performed in a laboratory accredited according to ISO/IEC 17025, authorised for gamma-ray spectrometry analysis by a competent national authority and a member of the IAEA ALMERA network. Quality control and quality assurance procedures were regularly conducted including national and international (IAEA) inter-laboratory comparisons.

Based on the obtained data, descriptive statistical parameters were calculated for each year resulting with the annual average activity concentration values of natural radionuclides $^{238}U$, $^{226}Ra$, $^{232}Th$, $^{40}K$, and $^{7}Be$ in mosses with the corresponding standard deviation, minimum, maximum and median values.

## RESULTS AND DISCUSSION

Table 1 provides data on the number of locations, moss species, and samples taken for each sampling year, as well as the corresponding results on annual average specific activities (A, Bq kg$^{-1}$) of $^{40}K$, $^{226}Ra$, $^{232}Th$, $^{238}U$, and $^{7}Be$ in the mosses collected in the period 2008-2013 in eastern Serbia, with basic statistic parameters: maximum and minimum values, and standard deviation. The lowest and highest values (in Bq kg$^{-1}$) of natural radionuclide activity concentrations determined in this study were for $^{40}K$: 64-484 (Manastir Gornjak, *H. sericeum*, 2013; Lepenski vir, *I. myosuroides*, 2008), $^{226}Ra$: 1.1-41 (Čezava, *D. scoparium*, 2010; Karataš, *G. trichophylla*, 2011), $^{232}Th$: 1.4-28 (Vrelo Mlave Žagubica, *H. sericeum*, 2013; Lepenski vir, *I. myosuroides*, 2008), $^{238}U$: 1.1-50 (Čezava, *D. scoparium*, 2010; Karataš, *G. trichophylla*, 2011), and $^{7}Be$: 88-227 (Dubašnica, *P. juniperinum*, 2013; Kanjon Vratne, *A. viticulosus*, 2013).

The obtained minimum and maximum activity concentration values differ significantly and for radionuclides $^{238}U$, $^{226}Ra$, $^{232}Th$, $^{40}K$, and $^{7}Be$ their ratio was 45; 37; 20; 8.1, and 2.6, respectively. All of the selected species have shown the capacity to accumulate radionuclides since $^{226}Ra$, $^{232}Th$, $^{238}U$, $^{40}K$, and $^{7}Be$ activity concentrations exceeded limits of detection in all the analysed samples. Due to a performed random sampling, it was not possible to establish a reliable relationship between the determined activity levels and certain type of moss species. However, Figures 1 and 2 present fractions (in %) of all the analysed moss samples having radionuclide contents in certain activity concentration intervals (0-10 Bq kg$^{-1}$; 10-20 Bq kg$^{-1}$ etc. in Fig. 1 and 0-100 Bq kg$^{-1}$, 100-200 Bq kg$^{-1}$, etc. in Fig.2). Figure 1 shows fractions of all samples with $^{238}U$, $^{226}Ra$, and $^{232}Th$ content up to 50 Bq kg$^{-1}$. It is evident that the most frequent interval of activity concentrations is 0-10 Bq kg$^{-1}$. About 48.0 % of all the samples collected in 2008-2013 and analysed in this study have activity concentrations of $^{238}U$ in that range. The percentage of samples with $^{226}Ra$ and $^{232}Th$ contents of up to 10 Bq kg$^{-1}$ was 47.1 % and 54.9 %, respectively.

Figure 2 presents a percentage of the moss samples with $^{40}K$ and $^{7}Be$ content of up to 500 Bq kg$^{-1}$. The activity concentration values in the analysed mosses from eastern Serbia were up to 450 Bq kg$^{-1}$ for $^{40}K$ and up to 250 Bq kg$^{-1}$

**Table 1** *Number (N) of locations, moss species and samples collected each year and descriptive statistical parameters for $^{238}U$, $^{226}Ra$, $^{232}Th$, $^{40}K$ and $^{7}Be$ activity concentrations (Bq kg$^{-1}$) in the analysed moss samples*

| A (Bq kg$^{-1}$) | | | $^{238}U$ | $^{226}Ra$ | $^{232}Th$ | $^{40}K$ | $^{7}Be$ |
|---|---|---|---|---|---|---|---|
| **2008** | | | | | | | |
| **N** | | Minimum | 3.7 | 6.6 | 5.5 | 134 | 96 |
| Locations | 13 | Maximum | 29 | 30 | 28 | 484 | 201 |
| Species | 13 | Average | 13.7 | 14.5 | 12.4 | 275 | 141 |
| Samples | 20 | St. Deviation | 7.4 | 7.1 | 6.7 | 82 | 35 |
| **2009** | | | | | | | |
| **N** | | Minimum | 5.6 | 3.8 | 7.3 | 140 | - |
| Locations | 3 | Maximum | 18.3 | 21 | 20 | 346 | - |
| Species | 5 | Average | 11.5 | 10.5 | 10.5 | 200 | - |
| Samples | 8 | St. Deviation | 4.7 | 5.4 | 3.6 | 62 | - |
| **2010** | | | | | | | |
| **N** | | Minimum | 1.1 | 1.1 | 3.8 | 98 | 116 |
| Locations | 9 | Maximum | 21 | 19.0 | 17.3 | 249 | 188 |
| Species | 2 | Average | 10.1 | 8.8 | 9.7 | 184 | 147 |
| Samples | 18 | St. Deviation | 4.8 | 5.1 | 3.7 | 44 | 23 |
| **2011** | | | | | | | |
| **N** | | Minimum | 3.6 | 4.0 | 3.7 | 178 | 93 |
| Locations | 9 | Maximum | 50 | 41 | 23 | 446 | 220 |
| Species | 11 | Average | 14.1 | 13.2 | 12.2 | 271 | 140 |
| Samples | 21 | St. Deviation | 9.8 | 8.4 | 5.0 | 65 | 33 |
| **2013** | | | | | | | |
| **N** | | Minimum | 3.1 | 2.2 | 1.4 | 64 | 88 |
| Locations | 11 | Maximum | 22 | 26 | 27 | 443 | 227 |
| Species | 15 | Average | 10.5 | 10.9 | 9.6 | 207 | 152 |
| Samples | 35 | St. Deviation | 5.6 | 5.4 | 6.1 | 101 | 34 |

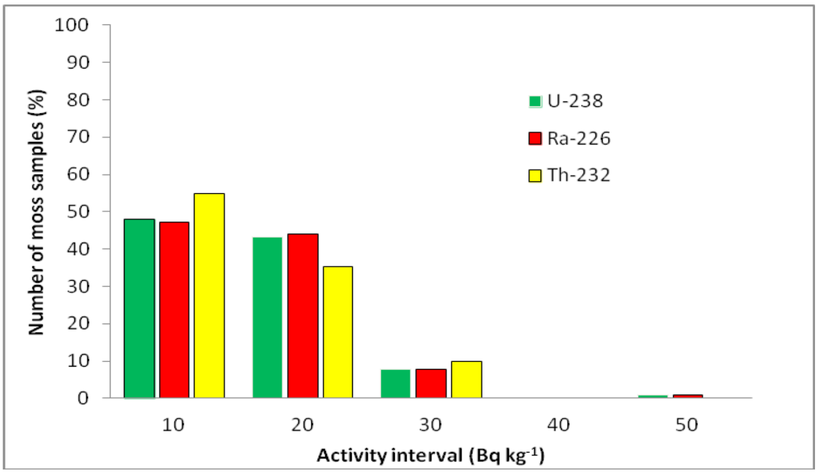


**Figure 1** *The fraction (in %) of analysed moss samples with $^{238}$U, $^{226}$Ra and $^{232}$Th activity concentrations up to 50 Bq kg$^{-1}$*

for $^{7}$Be, both with the maximum frequency of results around 200 Bq kg$^{-1}$.

The content of radionuclide $^{238}$U in mosses varied between 1.1 Bq kg$^{-1}$ and 50 Bq kg$^{-1}$ with yearly average activity concentrations 10.1±4.8 Bq kg$^{-1}$ to 14.1±9.8 Bq kg$^{-1}$ in 2008-2013, which is in accordance with our previous study on the $^{238}$U content in mosses from eastern Serbia spas: 0.4 Bq kg$^{-1}$ (*Necera crispa*) to 28 Bq kg$^{-1}$ (*Brachythecium rutabulum*) (24). These values are comparable with $^{238}$U average concentration 43 Bq kg$^{-1}$ obtained for mosses of Belgrade suburban area (26) and concentration range 1.7–25.1 Bq kg$^{-1}$ of $^{238}$U obtained for the mosses from central Serbia rural area (21).

The results of activity concentrations of $^{226}$Ra in the mosses from eastern Serbia varied within the interval 1.1-41 Bq kg$^{-1}$ with the yearly average activity concentration of 8.8 Bq kg$^{-1}$ up to 14.5 Bq kg$^{-1}$ in 2008-2013. Our previous research has shown $^{226}$Ra contents in the moss samples from spas in the range from 0.3 Bq kg$^{-1}$ (*H. sericeum*) up to 36 Bq kg$^{-1}$ (*H. lutescens*) (24). The obtained $^{226}$Ra concentrations are in good agreement with 11-75.0 Bq kg$^{-1}$ and 0.9–25.8 Bq kg$^{-1}$ reported by other authors for rural areas in Serbia (21, 26).

The average activity concentrations of $^{232}$Th in the mosses collected in the territory of Serbia were in the range from 8.4 to 41.4 Bq kg$^{-1}$ (21, 26) and the results of $^{232}$Th activity concentrations for the moss samples from eastern Serbia spas were from 1.0 Bq kg$^{-1}$ (*Anomodan viticulosus*) to 37 Bq kg$^{-1}$ (*Brachythecium rutabulum*) (24). In this study, the activity concentrations of $^{232}$Th were from 1.4 (*H. sericeum*, Vrelo Mlave) to 28 Bq kg$^{-1}$ (*I. myosuroides*, D. Milanovac) with the annual average activity concentrations in the range from 9.6±6.1 Bq kg$^{-1}$ to 12.4±6.7 Bq kg$^{-1}$ in the investigated period (2008-2013).

The results listed in Table 1 show that the activity concentrations of $^{40}$K in the analysed mosses from eastern Serbia varied from 64 Bq kg$^{-1}$ to 484 Bq kg$^{-1}$, while the annual average activity concentration of $^{40}$K in 2008-2013 was in the range from 184±44 Bq kg$^{-1}$ to 275±82 Bq kg$^{-1}$. These results are in good agreement with the results obtained within a study on natural radionuclides in the mosses from eastern Serbia spas where $^{40}$K activity concentration was in a wide interval from 25 Bq kg$^{-1}$ (*Brachythecium rivulare*) to 427 Bq kg$^{-1}$ (*Homalothecium lutescens*) (24). Other research results have shown that the activity concentrations of $^{40}$K in the mosses in Serbia were up to 740 Bq kg$^{-1}$ (21, 26, 27). The explanation of results on $^{40}$K contents in mosses might be complex due to originally present potassium in the plant tissue besides the ongoing physico-chemical processes.

The activity concentration of $^{7}$Be was determined in 90 moss samples from the territory of eastern Serbia collected in 2008-2013 during the summer season. The average activity concentrations of $^{7}$Be in *Hypnum cupressiforme* in the northern areas of Serbia were in the range from 201 to 920 Bq kg$^{-1}$ (25, 28) and our previous results have shown $^{7}$Be activity concentration in the moss samples from eastern Serbia spas varying from 20 Bq kg$^{-1}$ (*Pylaisia polyantha*) to 212 Bq kg$^{-1}$ (*Homalothecium sp.*) (24). In this study, the activity concentrations of $^{7}$Be in mosses were in the range from 88 to 227 Bq kg$^{-1}$ with almost constant annual average activity concentrations from 140±33 Bq kg$^{-1}$ to 152±34 Bq kg$^{-1}$ in 2008-2013.

The median values of annual activity concentrations of terrestrial radionuclides $^{232}$Th and $^{238}$U, the first members of two natural radioactive series and primordial radionuclide $^{40}$K had a similar trend throughout the investigated time period, while the $^{7}$Be median values were almost constant, as presented in Figures 3 and 4.

The results of activity concentrations of natural radionuclides (potassium, uranium, radium, and thorium) in the moss samples from estern Serbia obtained in this study have shown common values characteristic for the territory of Serbia.

## CONCLUSION

The contents of dominant natural radionuclides $^{238}$U, $^{226}$Ra, $^{232}$Th, $^{40}$K and $^{7}$Be in the moss samples from eastern

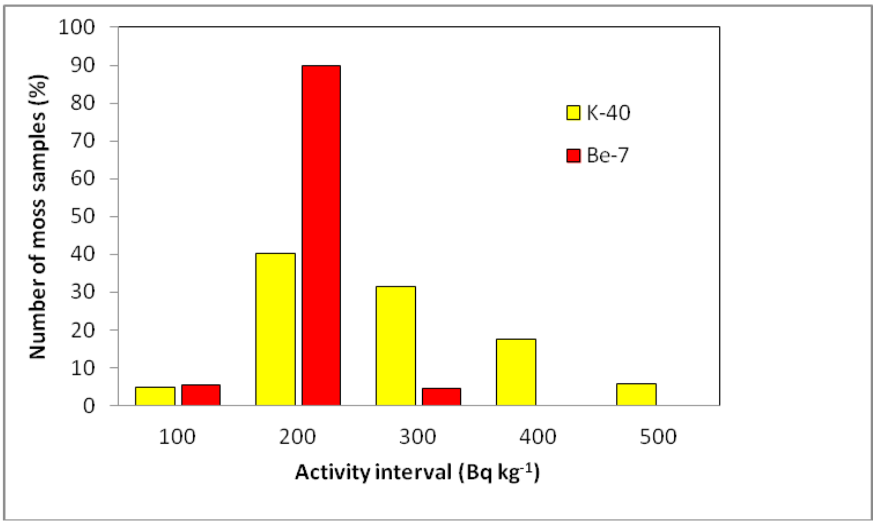


**Figure 2** *The fraction (in %) of analysed moss samples with* $^{40}K$ *and* $^{7}Be$ *activity concentrations up to 500 Bq kg*$^{-1}$

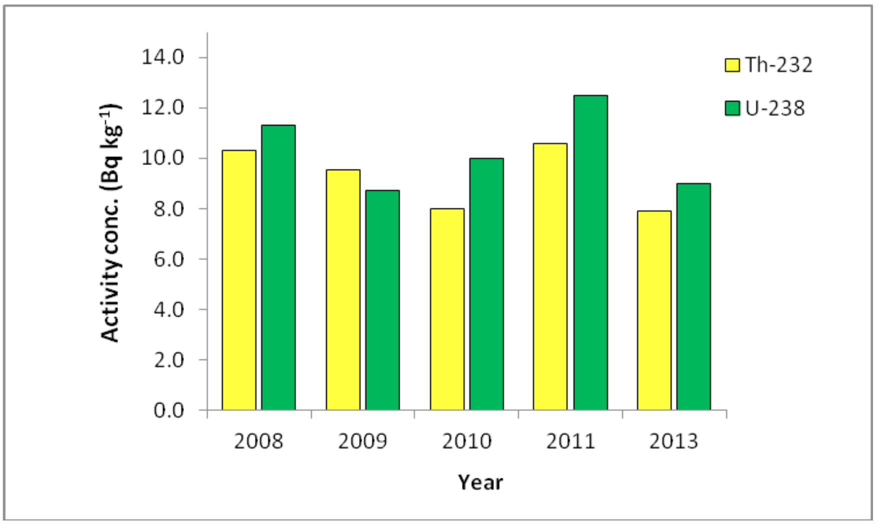


**Figure 3** *Annual median values of* $^{232}Th$ *and* $^{238}U$ *activity concentrations (Bq kg*$^{-1}$*) in analysed mosses*

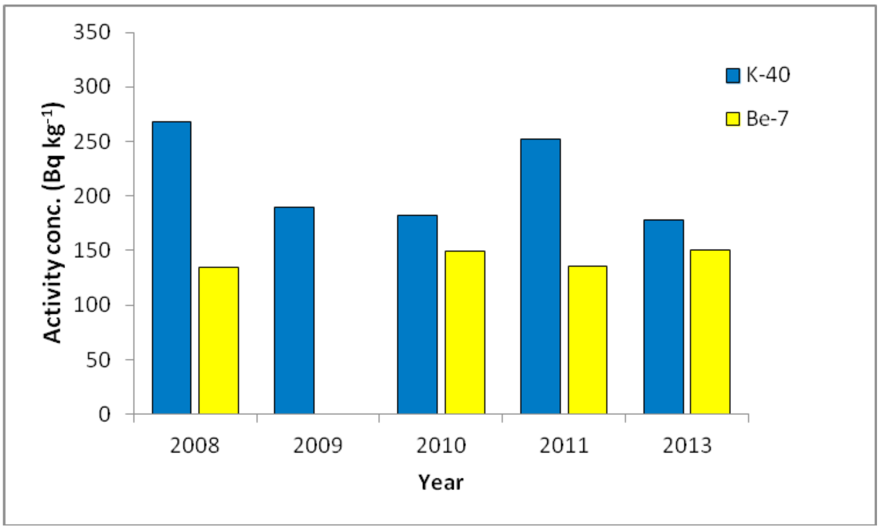


**Figure 4** *Annual median values of* $^{40}K$ *and* $^{7}Be$ *activity concentrations (Bq kg*$^{-1}$*) in analysed mosses*

Serbia in 2008-2013 are within the limits common for this region. The average annual activity concentrations of detected radionuclides do not change significantly in the observed time interval.

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**Prirodni radionuklidi u mahovinama iz istočne Srbije u razdoblju 2008.-2013**.

U radu su prikazani rezultati ispitivanja sadržaja prirodnih radionuklida u 102 uzorka mahovine prikupljene metodom slučajnoga uzorka u razdoblju 2008.-2013. na 30 lokacija u istočnoj Srbiji. Vrijednosti koncentracija (specifične aktivnosti) radionuklida $^{238}U$, $^{226}Ra$, $^{232}Th$, $^{40}K$ i $^{7}Be$, određene primjenom spektrometrije gama zračenja, nalaze se u opsegu: $^{238}U$ (1,1 – 50) Bq $kg^{-1}$, $^{226}Ra$ (1,1 – 41) Bq $kg^{-1}$, $^{232}Th$ (1,4 – 28) Bq $kg^{-1}$, $^{40}K$ (64 – 484) Bq $kg^{-1}$ i $^{7}Be$ (88 – 227) Bq $kg^{-1}$ te ne odstupaju značajno u odnosu na ostale rezultate dobivene za ovu regiju. Distribucija dobivenih podataka o koncentracijama $^{238}U$, $^{226}Ra$ i $^{232}Th$ u analiziranoj mahovini pokazuje vrijednosti do 10 Bq $kg^{-1}$, s učestalošću 54,9 %, 48,0 % i 47,1 %. Dobivene vrijednosti koncentracija primordijalnoga $^{40}K$ i kozmogenoga $^{7}Be$ bile su do 500 Bq $kg^{-1}$, pri čemu je oko 90 % svih rezultata za $^{7}Be$ akumuliranog u mahovinama bilo u opsegu koncentracija 200 – 250 Bq $kg^{-1}$.

KLJUČNE RIJEČI: *$^{238}U$; $^{226}Ra$; $^{232}Th$; $^{40}K$; $^{7}Be$; bioindikatori; gama-spektrometrija; koncentracije aktivnosti; mahovine*