

ELEMENTAL CONCENTRATIONS AND SOIL-TO-MOSS TRANSFER FACTORS OF RADIONUCLIDES IN THE ENVIRONMENT OF NORTH KOSOVO AND METOHIJA

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ABSTRACT

This paper deals with investigations of elemental concentrations and soil-to-moss transfer factors of radionuclides in area of municipalities Kosovska Mitrovica and Zubin Potok. Twelve samples of soil and moss *Hypnum cupressiforme Hedw.* were collected during May 2018. Transfer factors of radionuclides: ²²⁶Ra, ²³²Th, ⁴⁰K and ¹³⁷Cs were calculated with regard to elemental concentrations of radionuclides in soil and moss samples. Analysis was done in order to indicate the different ways of adopting radionuclides by mosses. According to calculated transfer factors and analysis, authors concluded that the soil is dominant source of natural radionuclides and their concentration in moss occurred due to resuspension of soil particles, while artificial ¹³⁷Cs is present in soil and moss samples as a consequence of atmospheric dry and wet deposition.

Keywords: Elemental concentration, Transfer factor, Radionuclide, Moss, Soil.

INTRODUCTION

Primordial radionuclides ²²⁶Ra, ²³²Th and ⁴⁰K are present in all soils and rocks. They are permanent source of irradiation of all biological species in nature, since they have long half-lives. Different chemical forms of these naturally occurring radionuclides emit gamma radiation which is known as terrestrial background. Therewith natural background radiation includes cosmic radiation and radon inhalation.

Besides natural radioactivity, the environment is uneven contaminated with artificial radionuclides, among them the radiologically most important is ¹³⁷Cs (half-life 30.07 y). It occurred in significant amounts particularly after the Chernobyl accident, as well as a consequence of nuclear tests and accidents after II World War. Therefore, exposure to any type of radiation poses some risk (UNSCEAR, 2008).

Radionuclides are incorporated in biological systems through the food chain, because plants rather absorb nutrients from soil than foliar (from particles suspended in air, transported by wind and precipitated by rainfall). On the other hand, the biological species that have ability to accumulate trace elements could serve as bio-indicators of radioecological status of an area.

Mosses are suitable for monitoring the ambient changes in the environment, since they have widespread geographical distribution and "evergreen phase" during the year. They also have high surface in comparison to volume and slowly grow with little morphological changes during the lifespan (Aceto et al., 2003; Ivanić et al., 2019; Fernandez & Carballeira, 2000).

Mosses are sensitive to climatic variations, which affect their growing, physiological activity, uptake and retention of

elements (Zechmeister et al., 2008; Dolegowska & Migaszewski, 2019). Bioaccumulation of trace elements in moss tissue depends on: the atmospheric concentrations and uptake of contaminants, precipitation, elevation, vegetation cover and topography of the sampling site.

Recently, radionuclides in soils and mosses have been investigated in regions of Serbia by many authors (Mitrović et al., 2009; Dragović & Mihailović, 2009; Grdović et al., 2010; Dragović et al., 2010; Čučulović et al., 2012; Mitrović et al., 2016; Krmar et al., 2018).

This is the first environmental study which used mosses as indicators of radiological contamination in area of Kosovska Mitrovica and Zubin Potok municipalities, North Kosovo and Metohija. The aims of this work were to determine elemental concentrations of radionuclides (²²⁶Ra, ²³²Th and ⁴⁰K) and concentrations of radioactive ¹³⁷Cs from atmospheric fallout in soil and moss samples, as well to calculate transfer factors in order to discuss various adopting ways of radionuclides.

MATERIALS AND METHODS

Study area

This study deals with the investigation of elemental concentration of radionuclides in soil and moss samples in the area of municipalities Kosovska Mitrovica and Zubin Potok (Figure 1). Twelve sampling sites of hilly terrain, in the slopes of surrounding mountains (Mokra Gora, Rogozna and Kopaonik) were marked (S1-S12) on the Figure 1.

The relief is characterized by a diverse geological structure with rocks of different origin and age (granite, serpentinite, shale, marble, andesite, limestone).

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The mainly sedimentary, but magmatic and metamorphic rocks consist of shales, gabbro amphibolite, andesites and quartz latites. The part of relief was undergone the karst process in the carbonate rocks with the tectonic movements, which caused numerous fissures and cracks (Dimitrijević, 1997).

The study area belongs to moderately continental climate with cold winter and moderately warm summers. The edges of mountains and valleys between 1000 and 1500 meters of height have sub-mountainous climate (Ivanović et al., 2016). An average amount of precipitation is more than 700 mm and increased with altitude. The measured precipitation in the nearest town Kosovska Mitrovica (510 m a.s.l.) was 657.6 mm and 535.7 mm during 2017 and 2018, respectively (Hydrometeorological Yearbook of Kosovo, 2017-2018).



Figure 1. Map of the study area.

Sample collection and preparation

Moss samples of *Hypnum cupressiforme* Hedw. and soil samples were collected from 12 locations (Figure 1). Sampling was performed in open areas away from trees, and far from towns, single houses and motorways during May 2018. Four samples of moss and soil from area of Kosovska Mitrovica, and eight ones from Zubin Potok were collected.

Hypnum cupressiforme Hedw. (Hypnaceae) is a group of mosses species with a wide variety of habitats in different

climatic zones. It has irregularly branched shoots (older parts are brown color, and younger are green) covered with strongly curved leaves; the stem leaves are concave and sickle-shaped. Capsules with spores are located on the top of the stems. They are rootless, and usually grow 3-5 years on tree trunks, rocks, ground and other surfaces (Frahm, 2009).

The collected moss samples were of carpet-forming growth types. A compound sample was made from five to ten sub-samples collected and mixed in within the same site. Samples were stored in paper bags. After transporting to the laboratory, they have been cleaned from litter and dead leaves. Only green and greenish-brown parts of the moss were used for gamma spectrometric analysis. These moss parts represent 3-5 years of plant growth. Underlying soil samples (0-10 cm) were also collected, cleaned from stones and roots, dried to constant weight, pulverized and sieved. All samples were packed in Marinelli beakers (450 mL), sealed, and left aside for a month to ensure equilibrium between ^{226}Ra and its progeny.

Gamma spectrometry

Gamma spectrometric measurements of soil and moss samples were performed with coaxial HPGe detector (GEM30-70 ORTEC) of 30% relative efficiency and 1.65 keV FWHM at 1.33 MeV (^{60}Co) and 717 eV at 122 keV (^{57}Co). Detector was shielded with 10 cm of lead to reduce the background.

Specific activities of ^{226}Ra , ^{232}Th , ^{40}K and ^{137}Cs in soil and moss samples were measured for 21600 s and 172800 s, respectively. System calibration was done with standard mixture MBSS 2 of gamma-emitting isotopes (^{241}Am , ^{109}Cd , ^{139}Ce , ^{57}Co , ^{60}Co , ^{137}Cs , ^{113}Sn , ^{85}Sr , ^{88}Y , ^{203}Hg , ^{152}Eu) provided by the Czech Metrology Institute. Maestro 32 was used for peak readings. The intensities and gamma lines of considering radionuclides are presented in Table 1.

Table 1. Intensities and gamma lines of radionuclides.

Radionuclide	Progeny	Gamma energy (keV)	Intensity (%)
^{226}Ra	^{214}Pb	351.9	37.1
	^{214}Bi	609.3	46.1
	^{214}Bi	1764.5	15.9
^{232}Th	^{228}Ac	338.3	12
	^{228}Ac	911.1	29
	^{228}Ac	968.9	17.5
	^{208}Tl	583.0	86
	^{208}Tl	860.6	12
^{40}K		1460.7	10.7
^{137}Cs		661.6	84.6

The total uncertainty of the activity measurements which includes the uncertainty of calibration source activity, efficiency of calibration and counting statistical errors were in the range of 3-10%.

Elemental concentrations of radionuclides

Elemental concentrations of ^{226}Ra , ^{232}Th and ^{40}K were obtained by converting their specific activities according the following equation (Tzortzis & Tsertos, 2004; Dragović et al., 2006):

$$F_E = \frac{M_E C}{\lambda_{E,i} N_A f_{E,i}} A_E \quad (1)$$

where F_E is the fraction of element E in the sample, M_E , $\lambda_{E,i}$, $f_{E,i}$ and $A_{E,i}$ are the atomic mass (kg/mol), the decay constant of the measured isotope i of the element E (1/s), the fractional atomic abundance in nature and measured specific activity of the element E (Bq/kg), respectively, N_A is the Avogadro's number ($6.023 \cdot 10^{23}$ 1/mol), and C is a constant with a value of 10^6 for radium/thorium, and 10^2 for potassium. Hence, elemental concentrations F_E are reported in units of ppm (equivalent to mg/kg) for ^{226}Ra and ^{232}Th , and as a percentage (%) for ^{40}K . Thus, if it is assumed that in uranium series radioactive equilibrium exists, then the concentrations of ^{226}Ra , ^{232}Th and ^{40}K can be calculated using the conversion factors:

- 1 Bq/kg of ^{226}Ra = 0.08097 ppm of U;
- 1 Bq/kg of ^{232}Th = 0.246305 ppm of Th;
- 1 Bq/kg of ^{40}K = 0.003195 % of K.

Transfer factor and discrimination factor

The ability of plants to uptake elements, expressed as the transfer factor, is similar for trees, grasses, and mosses (Zolotareva et al., 1983; Kabata-Pendias & Pendias, 2001). The parameters needed for quantification of radionuclide transfer to biota are continually updated and are used for reconsideration of transfer factors recommended by the International Atomic Energy Agency (IAEA, 1994). Transfer factors (TF) from soil to moss were calculated as follows:

$$TF = \frac{A_{moss}}{A_{soil}} \quad (2)$$

Where A_{moss} and A_{soil} are concentrations of radionuclides in moss and soil, respectively.

It has been accepted that caesium enters the plant mainly via potassium transport system. ^{40}K is a part of natural potassium which is the most abundant essential element in soil and plants. Since caesium and potassium belong to the same group of the periodic table and have similar chemical properties, mosses can discriminate between ^{40}K and ^{137}Cs in the process of uptake of these radionuclides. Discrimination factor (DF) is defined as:

$$DF = \frac{TF_{Cs}}{TF_K} \quad (3)$$

where TF_{Cs} and TF_K are transfer factors of ^{137}Cs and ^{40}K , respectively. DF value less than unity means that ^{40}K is more

efficiently absorbed by the plant than ^{137}Cs (Zhu & Smolders, 2000).

RESULTS AND DISCUSSION

Elemental concentrations of ^{226}Ra , ^{232}Th and ^{40}K in soil and moss samples were calculated and presented in Table 2. Mean values of elemental concentrations are in good agreement with results reported for surface soils in Serbia (Dragović et al., 2014). The values of elemental concentration in moss are lower than in soil, which is expected, since the main source of primordial radionuclides is soil. The mean values of ^{232}Th elemental concentration for all the soil and moss samples were higher than for ^{226}Ra . ^{226}Ra easily enters plants from soil and due to chemical activity it behaves similarly to calcium in an organism (Čučulović, 2016). ^{226}Ra elemental concentrations in mosses is related to direct deposition of ^{222}Rn progenies attached to the dust particles, which are accumulated by mosses from soil resuspension, through dry and/or wet atmospheric deposition of aerosols (Kiliç et al., 2019). Due to originally present potassium in the moss tissue, and ongoing physicochemical processes uptake pathways of ^{40}K might be complex. Only sample S5 has higher percentage of ^{40}K in moss than in soil.

Table 2. Elemental concentrations of radionuclides in soil and moss and descriptive statistics.

Sample	^{226}Ra (ppm)		^{232}Th (ppm)		^{40}K (%)	
	soil	moss	soil	moss	soil	moss
S1	9.59	1.06	35.32	6.67	3.95	0.54
S2	1.17	0.15	3.65	0.84	0.51	0.38
S3	3.00	0.53	16.03	2.49	3.61	0.75
S4	2.28	0.15	12.22	1.11	1.44	0.36
S5	0.15	<MDA*	0.39	<MDA*	0.07	0.25
S6	1.40	0.08	5.49	0.20	0.51	0.38
S7	1.45	0.40	4.68	1.55	0.68	0.66
S8	2.74	0.23	11.58	1.08	1.89	0.51
S9	2.53	0.22	9.68	0.39	1.16	0.38
S10	1.68	0.09	7.86	0.71	0.77	0.34
S11	3.29	0.15	16.21	0.57	3.30	0.43
S12	2.84	0.29	13.50	1.50	2.43	0.48
Min	0.15	<MDA*	0.39	<MDA*	0.07	0.25
Max	9.59	1.06	35.32	6.67	3.95	0.66
Mean	2.68	0.30	11.38	1.56	1.69	0.45
Median	2.04	0.22	10.63	1.08	1.30	0.40
SD	2.36	0.28	9.04	1.81	1.33	0.14
Skewness	2.54	2.17	1.71	2.63	0.62	0.91

MDA*- 0.05 for ^{226}Ra ; 0.17 for ^{232}Th

Figure 2 presents concentration of ^{137}Cs in soil and moss samples; the highest concentration in soil was noted in duff/mull soil (S5). Values of ^{137}Cs concentration in soil were higher than concentration in moss in six samples. Since ^{137}Cs is present more

than thirty years in environment mainly from atmospheric Chernobyl's fallout, this is expectable. At the other side, the reasons of higher ^{137}Cs concentrations in moss than in soil samples could be related with altitude and pine forest. It could be considered in terms of features of localities: this occurred in coniferous forests (mosses in pine and spruce forests more readily uptake radionuclides than oak forests), and it could be related with acidic conifer needles, its retaining the soil moisture and making different substrate. Džoljić et al. (2017) reported very low TF_{Cs} for spruce needles (range from not detected to 0.02), which could confirm washing out effect and deposition in substrate by precipitation.

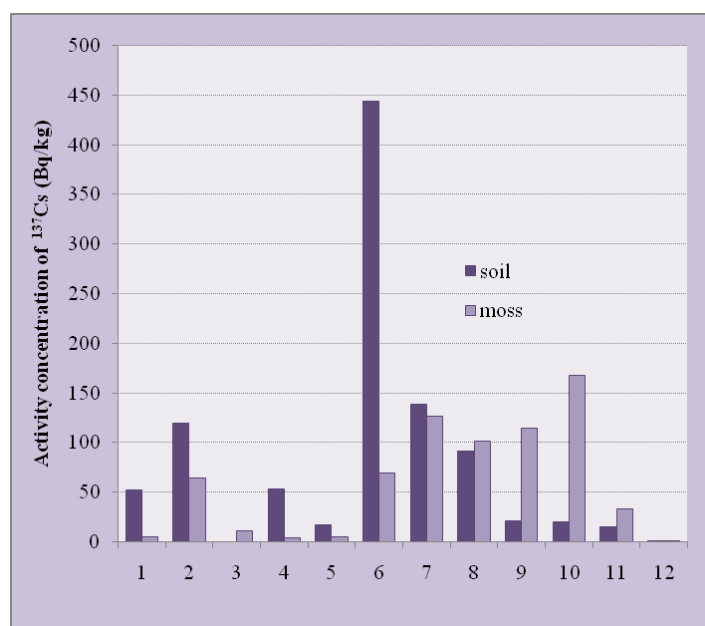


Figure 2. Concentrations of ^{137}Cs in soil and moss samples.

The most important way of ^{137}Cs deposition in moss tissue is possibility that ^{137}Cs remains on the surface, which influences soil resuspension and transport to mosses (Ioannidou & Papastefanou, 2006, Krmar et al., 2018); also, ^{137}Cs can be transported during growing period from older sections to newer sections of moss tissue (Krmar et al., 2018). Furthermore, physiological and morphological features of the same moss species may vary among localities; it includes different growing ages of moss tissue, different dynamic processes of biosorption by different sections of mosses) and hence, the accumulation of atmospheric fallout may differ (Kilić et al., 2019). In addition, variability of radionuclide activities might be attributed to local climatic conditions (amount of rainfall, humidity and wind direction). The results indicate that these moss species absorb water and nutrients as well as other trace elements primarily through wet and dry deposition rather than from soil.

Transfer factors of ^{226}Ra , ^{232}Th , ^{40}K and ^{137}Cs were calculated and presented in Table 3. TF for available data was in the range of 0.04-0.28 for ^{226}Ra , 0.03-0.33 for ^{232}Th and 0.13-3.63 for ^{40}K . A similar range for TF_{Ra} (0.05-0.57) and TF_{Th}

(0.06-0.48) was obtained by Dragović et al. (2010) in the region of Zlatibor Mt. Moss capacity for absorption and retention of ^{226}Ra is much higher than in vascular plants (Tsikritzis et al., 2003). The TF_{K} is found to be more than unity for samples S5. With this exception, the values of TF_{K} mainly fall into the range of results obtained from the Zlatibor mountain area, 0.15-0.96 (Dragović, 2010) and non-urban area in Southern Serbia, 0.19-0.90 (Popović, 2008). A range of soil-to-moss transfer factors of natural radionuclides ^{226}Ra and ^{232}Th can be explained by similar geochemical behavior which influences radionuclide distribution based on the topography and environmental processes such as weathering. Some researchers confirmed the existence of a synergistic and antagonistic relationship between individual elements (Kabata-Pendias & Pendias, 2001).

Table 3. Transfer factors and discrimination factors.

Sample	TF_{Ra}	TF_{Th}	TF_{K}	TF_{Cs}	DF
S1	0.11	0.19	0.14	0.10	0.73
S2	0.13	0.23	0.74	0.53	0.72
S3	0.18	0.16	0.21	-	-
S4	0.07	0.09	0.25	0.09	0.34
S5	-	-	3.63	0.30	0.08
S6	0.06	0.04	0.74	0.16	0.21
S7	0.28	0.33	0.96	0.91	0.95
S8	0.09	0.09	0.27	1.11	4.14
S9	0.09	0.04	0.32	5.58	17.22
S10	0.05	0.09	0.44	8.50	19.39
S11	0.04	0.03	0.13	2.28	17.72
S12	0.10	0.11	0.20	1.22	6.21

On the other hand transfer factors are spanning from 0.02-8.50 for ^{137}Cs (for available data). The values of TF_{Cs} are lower than those reported by Dragović et al. (2010) for Zlatibor Mt. (1.01-13.1). The TF_{Cs} are found to be more than unity for five of eight locations in Zubin Potok which could be attributed to mainly hilly terrain and higher precipitation rate. Some authors reported that the average transfer factor for ^{137}Cs in *Hypnum cupressiforme* was up to two-fold higher than for other moss species (Dragović et al., 2010).

The DF ranges from 0.08 to 19.39 for sampling locations (Table 3). For certain location (mosses in coniferous and mixed forests), DF is found to be more than unity which indicated that ^{137}Cs uptake by moss tissue is more readily than ^{40}K . Also, some authors report that certain synergistic effects have been observed for antagonist pairs of elements, which largely depend on the corresponding reaction of the plant species (Kabata-Pendias & Pendias, 2001).

CONCLUSION

The mean values of ^{226}Ra , ^{232}Th and ^{40}K elemental concentration and ^{137}Cs activity concentrations measured in soil

samples were comparable to the worldwide averages. Elemental concentrations in soil samples were in the range of 0.15-9.59 ppm for ^{226}Ra , 0.39-35.32 ppm for ^{232}Th , and 0.07-3.95% for ^{40}K . Mean elemental concentration of ^{232}Th in soil and moss samples was higher than ^{226}Ra . Wide ranges of values were observed; the values of natural radionuclides in moss are lower than in soil, since the main source of primordial radionuclides is soil. However, concentration of artificial ^{137}Cs does not show such a trend; it was higher in half of soil samples. ^{137}Cs activity concentrations in soil samples are spanning from 0.39-443.4 Bq/kg, while activity of this isotope in moss samples ranged from 1-168.1 Bq/kg. The variability of radionuclide activities might be attributed to local climatic conditions (amount of rainfall, humidity and wind direction). The results indicate that these moss species absorb water and nutrients as well as other trace elements primarily through wet and dry deposition rather than from soil. It was concluded that this occurred mainly in broad leaved, deciduous forest, while concentration of Cs in another six samples (coniferous and mixed forests) is higher in moss samples than in soil. Transfer factors of ^{226}Ra , ^{232}Th , and ^{40}K were in the range of 0.04-0.28, 0.03-0.33 and 0.13-3.63, respectively. A range of soil-to-moss transfer factors of natural radionuclides ^{226}Ra and ^{232}Th can be explained by similar geochemical behavior which influences radionuclide distribution based on the topography and environmental processes such as weathering. The TF_{Cs} are found to be more than unity for five of eight locations in Zubin Potok which could be attributed to mainly hilly terrain and higher precipitation rate. DF is found to be more than unity which indicated that ^{137}Cs uptake by moss tissue is more readily than ^{40}K .

Based on results and discussion it can be pointed out that significant radiological contamination in terms of natural and artificial radionuclides in area of Kosovska Mitrovica and Zubin Potok municipalities does not exist.

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