RADIONUCLIDES' CONTENT IN FOREST ECOSYSTEM LOCATED IN SOUTH-WESTERN PART OF SERBIA

by

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The results of the gamma-spectrometric measurements in a 16500 ha large region of south-western Serbia, are presented. Activity concentrations of ^{40}K, ^{137}Cs, and ^{210}Pb in different deciduous and evergreen trees in the region are investigated. For all the investigated isotopes, there is a tendency that, on average, the lowest activity concentrations were found in tree stems, then in leaves, while the highest ones were in the soil. Statistical analysis did not show any differences between activity concentrations of leaves and needles, showing that both leaves and needles could be equally well used as a biomonitors.

Key words: forest ecosystem, evergreen tree, deciduous tree, gamma-ray spectrometry, specific activity, ^{40}K, ^{137}Cs, ^{210}Pb

INTRODUCTION

As a consequence of uncontrolled usage of Earth's resources, regeneration and sustainable development of different ecosystems is becoming one of the mankind's main priorities. Forests represent a very important ecosystem, as they cover around one third of the land area of Europe [1]. In Serbia, 29.1 % of the territory is covered by forests [2]. Forests are more prone to atmospheric pollution as they have greater ability to absorb it, compared to other vegetation types [3].

An increased concentration of radionuclides in a forest ecosystem can lead to an increase of external exposure of hunters, rangers, mushroom pickers, and other groups of people that spend some time in forests as well as, to an increase of internal exposure to those using medical herbs, or eating wild berries [4].

Some contaminants of forest ecosystems are radionuclides that can be of natural or artificial origin. Among the natural ones, primordial long-lived radionuclides ^{40}K, and radionuclides from uranium and thorium series are present in soil, while their presence in the air is usually negligible [5]. On the other hand, progenies of ^{222}Rn and ^{220}Rn isotopes, from uranium and thorium decay series, can be found in the air as well. Being a good tracer, ^{210}Pb, from uranium series, with a half-life of 22.3 years, is interesting to monitor.

The first significant contamination of forest by ^{137}Cs, ^{90}Sr and other artificial radionuclides in Europe, was in the period of nuclear testing in 50's and 60's and the highest one due to Chernobyl accident in 1986 [6]. On the other hand, contribution of airborne radionuclides from the damaged Fukushima Daiichi Nuclear Reactors to European forests was insignificant [7]. Due to its half-life of 30.07 years and high mobility, ^{137}Cs is present in the forest ecosystem for decades and can become part of the food chain and thus, represents a health risk that is interesting to monitor.

Since forests are efficient absorbers of pollutants, it is a matter of concern to what extent radionuclides stay in forest ecosystems and consequently, could have impact on humans as they exploit forests' resources. Circulation of radioactive contaminants in forest ecosystems can be simplified in a few processes. The first process is interception of contaminants by leaves, needles and branches of the canopy. Contaminants could be either absorbed by a tree canopy or further transferred to the forest floor by processes such as weathering (rainfall and wind) or leaf fall. Radionuclide contaminants from the forest floor further migrate to soil. In soil, they can migrate to dif-

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ferent layers of soil or can be absorbed by roots and consequently transferred to leaves or needles through woody steams [8].

All these processes vary in different for different types of forests, deciduous or evergreen, with different sizes of tree canopy, and dependent on the time of the year [8]. In order to understand the distribution of radionuclides in different parts of woods (trees), radionuclide content in tree stems of different trees, leaves or needles, detritus (tree floor) and soil in the forest ecosystem of south- western Serbia has been investigated. Moss and bark are known as a good biomonitors [9, 10] and it is of interest to determine whether some other parts of trees could be used as biomonitors as well.

MATERALS AND METHODS

Investigation was conducted in south-western Serbia, covering the region of 16500 ha at an altitude between 750 m and 1150 m, rich in natural resources. It is a part of Dinaric Alps (Dinarides), mainly composed of Mesozoic limestones and dolomite, deposited on the top of carbonate platform [11]. The most dominant geological formations are Golija and Rogozna mountains, and the Pešter plateau. It is a vast mountainous territory, with mild and sharp ascents, river cuts and valleys, plateaus, large complexes of deciduous and coniferous forests. The region is rich in flora and fauna, clean water with numerous mineral and thermal springs. Hardwood trees cover 86.9 % and conifers 13.1 % of the total volume of all trees. The most common species are: sessile with 17.7 %, cerris with 8.6 %, black pine with 6.3 %, spruce with 4.6 %, hornbeam with 2.1 %, while other tree species participate with less than 1.0 % [2].

An overview of the collected samples of tree stems, leaves or needles and detritus taken from different deciduous and evergreen trees as well as the number of soil samples taken in close vicinity of those trees are given in tab. 1.

All environmental samples were collected and prepared according to an appropriate guidebook [12]. Soil samples were collected from depths of up to 20 cm, adjacent to the trees from which the other samples were collected. In total, two soil samples near deciduous and eight soil samples close to evergreen trees were collected. The soil samples were dried, sieved and packed in Marinelli beakers. Samples of biota were dried either at a room temperature or at 105 °C, mineralised at 400 °C, and packed in cylindrical boxes of 120 cm^3 and 250 cm^3 volumes, depending on the sample size.

The measurements were conducted in the Department of Radiation and Environmental Protection, Vinča Institute of Nuclear Sciences, on three HPGe detectors, models Canberra GC 2018-7500, Canberra 7229N-7500-1818, and Canberra GC5019-7500SL

Table 1. An overview of collected biological samples taken from different deciduous and evergreen trees and soil samples taken in vicinity of these trees

Name of tree		Number of samples			
Common name	Latin name	Tree stem	Leaves/ needles	Detritus	Soil
Deciduous trees					
European beech	*Fagus sylvatica*	2	1	1	
Turkey oak	*Quercus cerris*				
Wild cherry	*Prunus avium*	1			
Willow	*Salix alba*	1	1		
Alder	*Alnus glutinosa*	1			
Common hornbeam	*Carpinus betulus*	1	1	2	1
European crab apple	*Malus sylvestris*	1			1
Field maple	*Acer campestre*	2			
Common hazel	*Corylus avellana*	1			
Sessile oak	*Quercus petraea*	2		1	
Evergreen trees					
Black pine	*Pinus nigra*	2	1	1	2
Douglas-fir	*Pseudotsuga menziesii*	1		1	1
Scots pine	*Pinus sylvestris*	1	2	1	2
Spruce	*Picea abies*	3	2	1	2
Blueberry	*Vaccinium myrtillus*	1			1
Silver fir	*Abis alba*		1	1	

with relative efficiency of 20 %, 18 %, and 50 %, respectively. The duration of the measurements ranged from 12000 seconds to 237000 seconds. The measurement geometries were Marinelli beaker and cylindrical geometries of 120 cm^3 and 250 cm^3, depending on the matrix and the volume of the samples.

The detectors efficiency calibration was performed by measuring certified radioactive standard, Marinelli beaker filled with epoxy resin containing gamma emitting radionuclides (product 9031-OL-419/12 issued by Czech Metrological Institute) and a set of laboratory standards produced by spiking the chosen matrix with the certified radioactive mixture solution ER X 9031-OL-426/12 issued by Czech Metrological Institute, Inspectorate for Ionizing Radiation. The radioactive solution contained the following radionuclides: ^{241}Am, ^{109}Cd, ^{139}Ce, ^{57}Co, ^{60}Co, ^{137}Cs, ^{203}Hg, ^{113}Sn, ^{85}Sr, and ^{88}Y, with the energies that span from 59 keV to 1898 keV. The matrices and geometries were chosen in such a way to match best the geometry and matrix of the measured samples. Namely, Marinelli beaker filled with coal and cylindrical geometry filled with ashed grass and coal were used. The preparation of the secondary reference material was conducted as described in [13].

Net count was corrected for the background and coincidence summing via coincidence summing correction factor calculated using EFFTRAN [14] for cylindrical geometry and MEFFFTRAN [15] for Marinelli beaker. The combined measurement uncertainty of the activity concentration was calculated by using standard error propagation formula and is given at the 95 % level of confidence. Minimum detectable activity (MDA) was calculated using standard Currie method [16].

Although the transport of radionuclides from soil to a plant depends on many parameters, such as physicochemical properties of soil and radionuclides, structural and biochemical features of plants, transfer factors are used to estimate the amount of radioactive contamination in investigated organisms from the ambient environment. Transfer factor, *TF*: *soil-to-leaves*, and *soil-to-tree stem*, were calculated by using [17, 18]

$$TF \quad \frac{A_i}{A_j} \tag{1}$$

where A_i (Bqkg^{-1} dry weight) is the activity concentration of leaves and tree stem, respectively and A_j (Bqkg^{-1} dry weight) – the activity concentration of soil.

RESULTS AND DISCUSSION

Activity concentrations of ^{40}K, ^{137}Cs, and ^{210}Pb for all the samples are given in Becquerel per kilogram of dry weight. Descriptive statistics (maximum, minimum, and mean value with standard deviation) of activity concentrations of radionuclides in tree stem, leaves and needles, detritus, and soil are given in tab. 2, while the graphical representation of mean radionuclide concentration of these samples is shown in fig. 1. Activity concentrations of ^{40}K range from 4.3 0.6 Bqkg^{-1} (*Pinus silvestris*) to 82 5 Bqkg^{-1} (*Quercus cerris*) for tree stem, from 59 7 Bqkg^{-1} (*Pinus nigra*) to 160 20 Bqkg^{-1} (*Carpinus betulus*) for leaves; 44 7 Bqkg^{-1} (*Fagus sylvatica*) to 160 10 Bqkg^{-1} (*Carpinus betulus*) for detritus, and from 62 5 Bqkg^{-1} to 970 60 Bqkg^{-1} for soil.

Activity concentrations of ^{137}Cs range from <0.009 Bqkg^{-1} (*Malus sylvestris*) to 2.5 0.2 Bqkg^{-1} (*Fagus sylvatica*) for tree stem, from <0.1 Bqkg^{-1} (*Pinus nigra*) to 13 0.8 Bqkg^{-1} (*Picea abies*) for leaves; 2.2 0.1 Bqkg^{-1} (*Picea abies*) to 96 6 Bqkg^{-1} (*Picea abies*) for detritus and from < 0.1 Bqkg^{-1} to 410 20 Bqkg^{-1} for soil.

Table 2. Descriptive statistics of ^{40}K, ^{137}Cs, and ^{210}Pb activity concentrations in tree stem, leaves and needles, detritus, and soil. Mean values are given in the form of mean standard deviation while in the case of measurements below detection limit mean values are given as an upper limit

Radionuclide (Bqkg^{-1} dry weight)		Tree stem	Leaves /Needles	Detritus	Soil
^{40}K	Min	4.3 0.6	59 7	44 7	62 5
	Max	82 5	160 20	160 10	970 60
	Mean	21 18	114 30	89 45	500 270
^{137}Cs	Min	<0.009	<0.1	2.2 0.1	<0.1
	Max	2.5 0.2	13 0.8	96 6	410 20
	Mean	0.52	5.6	22 29	95
^{210}Pb	Min	<0.3	24 1.5	32 6	<2
	Max	3.2 0.5	150 20	380 50	450 40
	Mean	1.6	62 40	170 105	100

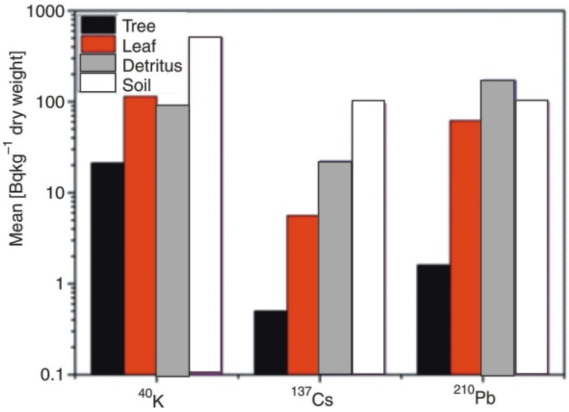

Figure 1. Mean value of activity concentrations of ^{40}K, ^{137}Cs, and ^{210}Pb in tree stem, leaves/needles, detritus and soil

Activity concentrations of ^{210}Pb range from <0.3 Bqkg^{-1} (*Pinus nigra*) to <5 Bqkg^{-1} (*Corylus avellana*), for tree stem, from 24 1.5 Bqkg^{-1} (*Picea abies*) to 150 20 Bqkg^{-1} (*Fagus sylvatica*) for leaves; 32 6 Bqkg^{-1} (*Quercus petraea*) to 380 50 Bqkg^{-1} (*Picea abies*) for detritus and from <2 Bqkg^{-1} to 450 40 Bqkg^{-1} for soil.

The obtained results are in agreement with the results reported in the region and worldwide [17, 19-23]. A wide range of radionuclides' content was found in all groups of samples, which leads to a large spread of values around mean value (large standard deviation). On average, tendency could be observed for all the isotopes, that the lowest activity concentration is detected in the tree stem, medium in the leaves and the highest in soil. The highest concentrations of ^{40}K and ^{137}Cs were found in soil, which is not surprising since ^{40}K generally origins from soil and most of airborne ^{137}Cs has migrated to soil, since last larger generation of ^{137}Cs that could influence the investigated region was during the Chernobyl accident. The translocation of potassium goes from older plant parts to developing parts, from tree stem to branches and leaves, during the vegetative growth [24], therefore, the lowest concentration was found in tree stems and higher in leaves. Since caesium, as potassium, is a group I alkali metal, it could be roughly expected that Cs+ uptake a follows similar mechanism as for K+ uptake, and therefore it is not surprising that ^{137}Cs concentration was also lowest in tree stem as for ^{40}K.

In the case of ^{210}Pb, the highest concentration, on average, was found in detritus. The origin of the ^{210}Pb in detritus as well as in leaves is twofold: one is from the soil, and another is the deposition from the atmosphere [17]. Investigation of stable lead isotopes ^{206,207,208}Pb, has shown that Pb uptake from mor layer and mineral soils is 0.03 and 0.02 mg m^{-2} per year respectively, while the Pb uptake directly from the atmosphere is 0.05 mg m^{-2} per year [25].

The amount of foliage in the tree canopy, known as leaf area index (LAI) is an important parameter in the description of its interaction with the atmosphere.

The higher the LAI, the higher would be the ability for the wet deposition of radionuclides in the foliage. The worldwide average value of LAI for forest temperate evergreen needle is slightly higher than LAI for the deciduous leaf, although within the measurement uncertainty, while its maximum is twice the maximum of LAI for deciduous trees [26]. Authors have investigated whether there is a systematic difference in radionuclide concentration in leaves and needles in our investigated region. The obtained means of the activity concentration of ^{40}K, ^{137}Cs, and ^{210}Pb for deciduous trees are: 140 26 Bqkg^{-1}, 4.9 7.1 Bqkg^{-1}, and 85 57 Bqkg^{-1}, respectively, while for evergreen trees are: 102 25 Bqkg^{-1}, 5.9 4.8 Bqkg^{-1}, and 52 31 Bqkg^{-1}, respectively. To check whether there is a difference in the mean activity concentrations of ^{40}K, ^{137}Cs and ^{210}Pb in leaves and needles, a two sample Student's *t*-test with significance level of $\alpha = 0.05$ was applied. The obtained *p*-values of 0.09, 0.58, and 0.22 for ^{40}K, ^{137}Cs, and ^{210}Pb, respectively indicate that there are no statistical differences of measured activity concentrations between leaves of deciduous and needles of evergreen trees.

Soil-to-leaves and *soil-to-tree stem* transfer factors for ^{40}K, ^{137}Cs, and ^{210}Pb, calculated only for samples with activity concentrations above MDA are given in tab. 3.

The obtained results for soil to leaves transfer factors are within the range of the worldwide results [17, 19-21, 23, 27].

Soil to leaves transfer factor of ^{40}K is systematically higher than transfer factor from soil to the tree stem, which is expected since K^+ translocation goes from older tree parts to the developing ones, as already mentioned [24]. Although only two measurable results were obtained, soil to leaves transfer factor higher than one for ^{210}Pb, indicate that an important source of ^{210}Pb in leaves is from atmospheric deposition. Also, only two results for transfer factors were found for soil to leaves and soil to tree stem and they differ by two orders of magnitude. To understand the results, larger statistics is required.

Table 3. *Soil-to-leaves* and *soil-to-tree stem* transfer factors for ^{40}K, ^{137}Cs, and ^{210}Pb

Tree name	Transfer	Transfer factor		
		^{40}K	^{137}Cs	^{210}Pb
Fagus sylvatica	Soil to tree stem			
	Soil to leaves	0.22	5.2	3.0
Malus sylvestris	Soil to tree stem	0.022		
	Soil to leaves			
Pseudotsuga menziesii	Soil to tree stem	0.049	0.021	
	Soil to leaves			
Pinus sylvestris	Soil to tree stem	0.007		
	Soil to leaves	0.19	0.038	
Picea abies	Soil to tree stem	0.006	2.1	
	Soil to leaves	0.103		
Vaccinium myrtillus	Soil to tree stem			
	Soil to leaves	0.14		1.0

CONCLUSIONS

In this manuscript, radionuclide content in different parts of typical deciduous and evergreen trees of forest ecosystem located in south-western part of Serbia was investigated. For the investigated radionuclides ^{40}K, ^{127}Cs, and ^{210}Pb, a trend was observed that, on average, the lowest activity concentrations were found in tree stems, then in leaves, while the highest ones were in soil.

Although worldwide data indicate that LAI is slightly higher for forest temperate evergreen needles than for deciduous leaves, Student's *t*-test did not indicate any statistical differences of measured ^{40}K, ^{127}Cs, and ^{210}Pb activity concentrations between leaves and needles.

Derived transfer factors of ^{40}K from soil to tree stem and from soil to leaves confirm that the transport of potassium ions goes from the tree stem to the developing parts of plant. Higher activity concentrations of ^{210}Pb in leaves than in soil indicate that important contribution to ^{210}Pb activity concentration is from the atmosphere which is also confirmed by Todorović and co-authors [17] underlying effect of ^{210}Pb accumulation in leaves. Therefore, the abovementioned results indicate that leaves could be used as good biomonitors.

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AUTHORS' CONTRIBUTIONS

S. H. Hadrović performed field work and collected samples. J. D. Krneta Nikolić, M. M. Rajačić, and D. J. Todorović conducted the measurements and data analysis. I. T. Čeliković performed a data analysis and served as a corresponding author. All authors contributed to the writing of the manuscript.

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САДРЖАЈ РАДИОНУКЛИДА У ШУМСКОМ ЕКОСИСТЕМУ У ЈУГОЗАПАДНОМ ДЕЛУ СРБИЈЕ

У овом раду представљена су гама спектрометријска мерења у региону величине 16500 ha, који се налази у југозападном делу Србије. Испитиване су специфичне активности ^{40}K, ^{137}Cs и ^{210}Pb у различитим врстама листопадног и зимзеленог дрвећа, присутног у региону. За све испитиване изотопе је уочен тренд да су најниже специфичне активности у просеку измерене у стаблу, потом у лишћу, док су највише у земљишту. Анализа је показала да не постоји статистички значајна разлика у специфичним активностима лишћа и иглица, што указује на чињеницу да се лишће и иглице могу подједнако добро користити као биомонитори.

Кључне речи: шумски екосистем, зимзелено дрво, листопадно дрво, гама спектрометрија, специфична активност, ^{40}K, ^{137}Cs, ^{210}Pb