SEASONAL VARIATIONS OF NATURALLY OCCURRING RADIONUCLIDES AND ^{137}Cs IN THE LEAVES OF DECIDUOUS TREE SPECIES AT SITES OF BACKGROUND RADIOACTIVITY LEVELS

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Abstract. Activity concentration of natural radionuclides and ^{137}Cs were studied in leaves of the deciduous trees. In the spring and autumn season, leaves were collected in the area of normal background radiation levels represented by city parks in a multi-year period (2002–2012). Measurements by means of gamma-ray spectrometry showed ^{226}Ra and ^{210}Pb seasonal accumulation in leaves, while ^{238}U and ^{235}U could be detected only in autumn. Difference between seasons was not found significant for ^{40}K and ^{137}Cs. The study of radionuclides transfer factors was conducted by analyzing its relationships with basic soil properties at the beginning and the end of the vegetation period.

Key words: Natural radionuclides, ^{137}Cs, Soil-to-leaves transfer factors, Background sites.

1. INTRODUCTION

The main contribution to the background radiation arises from the naturally occurring radionuclides members of the uranium, thorium and actinium decay series and the long-lived primordial potassium isotope, ^{40}K. Radiological risk assessments in terrestrial environments are based on the evidence of these radionuclides’ migrations, distribution pathways and transfers between natural compartments. The migration from soil to plants provides one important pathway since long-lived radionuclides present in soil are taken up and translocated by plants regardless of their biological necessity [1]. Plant species have a different ability to absorb radionuclides as non-essential elements depending on their biological properties using the mechanism of “mimicry” or chemical similarity with essential elements necessary for nutrition, growth and development [2]. Radionuclides are taken up by plants, if present in their available form through the root system from soil or from the air through the pathway of foliar deposition [3].

Concerning higher plants, the anthropogenic radioisotope ^{137}Cs has predominantly been studied in the parts of the trees, especially after the Fukushima nuclear accident which caused a majority of radioactive fallout to be captured by forests [4]. Uptake of natural radionuclides by trees was mainly studied in areas affected by uranium mining where higher levels of uranium and its decay products are available [5, 6]. On the other hand, there are scarce data of primordial radionuclides background activity concentrations in tissues of higher plants due to their very low and often undetectable levels. Although, if sufficient material is available for analysis, background activities could be measurable [7]. At a few background sites, ^{238}U, ^{226}Ra and ^{232}Th were detected in the organs of the trees [7, 8, 9] and activity levels in the leaves showed to be of the same order of magnitude as in the tree barks and one order of magnitude higher than in the trunk wood. Distribution like that within the tree is expected because a leaf, as an actively growing tissue, is enriched with both essential and nonessential elements [10]. Comparing uranium plant/soil concentration ratios between sites uncontaminated and contaminated by uranium, some differences were observed for the deciduous tree leaves, but those values were not found to be significantly different [6]. In order to examine further such a difference, more data about the effects of background levels of natural radioactivity on plants are needed [1].

Plant uptake is a complex process affected by characteristics such as soil type, physical and chemical properties of the soil, the type of plants and the physico-chemical form of the radionuclides which is why a large variability in measured *transfer factors* (TFs) values are produced [3]. Recent studies have suggested that TF variability could be reduced if mobile or plant available fraction of radionuclides and not its total concentration in soil is considered [2, 4]. Those fractions are found to be strongly affected by the soil's parameters, especially textural characteristics [5, 11].

The present study was carried out in the area of normal background radioactivity levels represented by three Belgrade city parks. Leaves from selected deciduous tree species *Tilia* spp. (linden) and *Aesculus hippocastanum* L. (horse chestnut) commonly found in the parks were sampled each year from 2002 to 2005 and again from 2008 to 2012 in the spring, at the begining, and in autumn, at the end of the vegetation season. After improving detection efficiency, ^{238}U, ^{235}U, ^{226}Ra, ^{210}Pb, ^{40}K and ^{137}Cs activities became measurable in the samples of leaves and enabled the study of the associated changes of their vegetation cycle. The root uptake from the soil to the leaves was considered the main pathway of the investigated radionuclides translocation and in the case of ^{210}Pb, pathways of foliar deposition and root uptake were taken into consideration. The study of radionuclides transfer from soil to the leaves is done by analyzing transfer factors relationships with some basic soil properties.

2. MATERIALS AND METHODS

The study area was located in the city of Belgrade, the capital of Serbia (44° 49′ 14" N, 20° 27′ 44'' E; 117 m a.s.l; population around 2 million). The climate in Belgrade is moderate continental with fairly cold winters and warm summers. The annual precipitation pattern in Belgrade has an increase in rainfall from spring to summer with June as the wettest month of the year. The sampling was conducted in the three city parks in the central zone of the city: The *Botanical Garden* (BG), *Students' Park* (SP) and *Karadjordjev Park* (KP). Samples of leaves were collected from deciduous tree species *Tilia* spp. (linden: *Tilia tomentosa* L. and *Tilia cordata* Mill.) and *Aesculus hippocastanum* L. (horse chestnut). Collection took place from 2002 to 2005 and again from 2008 to 2012. In the entire period (2002–2012), seven sampling events in spring and eight sampling events in autumn were performed at the beginning (May) and at the end (October) of the vegetation season, respectively. At each park, five subsamples (10–15 fully developed leaves) were taken randomly from the tree crowns 2 m above the ground and a composite sample was made. In the laboratory the leaves were dried at room temperature or at 105°C and then mineralized by dry ashing at 430°C. The ashes of leaf samples were packed in 125 cm^3 plastic cylinder containers, sealed with a film of beeswax and measured 4 weeks later after the radioactive secular equilibrium within the uranium and thorium decay chains is assured.

Measurement results of radionuclides activity concentration in individual samples of leaves each year and season showed the presence of radionuclides ^{40}K, ^{210}Pb and to some extent ^{137}Cs. In order to detect radionuclides other than that, improvement of detection efficiency was achieved by increasing the available sample mass. This was done by joining together the individual mineralized samples of leaves in such a way to obtain integral samples ($n = 12$) that would be representative for each species (chestnut, linden), season (spring, autumn) and park (BG, SP, KP) in the investigated period (2002–2012). Since integral leaf samples consisted of subsamples of different initial mass, the concentration factor F_c had to be determined. It was calculated based on the activity of long-lived radionuclide ^{40}K ($t_{1/2} = 1.28 \cdot 10^9$ y) measured before and after integration of the leaf samples:

$$F_c = \frac{A'_{\text{int}} \cdot m'_{\text{int}}}{\sum_{i=1}^{n} A_i^o \cdot m_i^0} \tag{1}$$

In the equation (1), A'_{int} and m'_{int} are ^{40}K activity concentration (Bq kg^{-1} ash weight) and mass (kg ash weight), respectively of the given integral sample and A_i^0 and m_i^0 are the initial ^{40}K activity concentration (Bq kg^{-1} dry weight) and mass (kg dry weight), respectively of each individual leaf sample from which the integral sample consisted. The number n in the sum is the number of the terms being added with values 7 or 8 depending on the season. Activity concentration (Bq kg^{-1} dry weight) of radionuclides identified in the integral samples of leaves was determined using the *concentration factor* (F_c).

Radionuclide activity measurements were conducted by the standard gamma spectrometry method using HPGe detectors (Canberra Industries, Inc., USA) of 20% and 18% relative efficiency and energy resolution of 1.80 keV and 1.69 keV at the

1332 keV gamma ray energy of ^{60}Co, respectively. The detector was calibrated using standard reference material with mineralized grass matrix in the 125 ml cylindrical geometry. Detector geometric calibration was performed with a secondary reference material produced by spiking the mineralized grass with the standard solution containing homogeneously dispersed radionuclides: ^{241}Am, ^{109}Cd, ^{139}Ce, ^{57}Co, ^{60}Co, ^{203}Hg, ^{88}Y, ^{113}Sn, ^{85}Sr, ^{137}Cs, and ^{210}Pb with 72.40 kBq total activity at 31.08.2012. (9031-OL-427/12, type ERX, Czech Metrology Institute, Prague). The activity of ^{238}U was determined by its decay product ^{234}Th at 63.3 keV. The activity of ^{226}Ra was determined by ^{214}Bi (609.3 keV, 1120.3 keV, and 1764.5 keV) and ^{214}Pb (295.2 keV and 351.9 keV). The activity of ^{235}U was established using its 185.7 keV gamma-energy corrected for ^{226}Ra (186.1 keV). The ^{210}Pb activity was calculated based on its gamma photons of 46.5 keV energy. ^{40}K and man-made ^{137}Cs activities were obtained from their single gamma ray lines of 1460.8 keV and 661.66 keV energy, respectively. The counting time was 60 ks. The spectra were recorded and analyzed using Canberra Genie 2000 software. The uncertainty of the method is expressed as an expanded measurement uncertainty for the factor $k = 2$ that corresponds to a normal distribution with a confidence level of 95%. The combined standard uncertainty of the activity concentration determined in the leaves was 7% for ^{40}K, 15–20% for ^{226}Ra, ^{210}Pb, ^{137}Cs and 25–30% for ^{238}U and ^{235}U.

Adjacent to deciduous trees, soil samples were collected at the same locations at depth of 0–50 cm at BG and SP and the sampling was conducted at a depth of up to 30 cm at KP. All the details of the methods used to determine the basic soil properties and radionuclides activity concentration can be found in our previous work [12]. The soil-to-leaves transfer factor was calculated [3] as the ratio between the radionuclide activity concentration (Bq kg^{-1} dry weight) of the integral leaf sample and the radionuclide mean activity concentration (Bq kg^{-1} dry weight) of soil according to the whole investigated depth.

The normality of data was evaluated using the Shapiro-Wilk's test. Statistically, radionuclides activity concentrations and their TF values were shown to follow a normal distribution at the 95% confidence level. One-way analysis of variance (ANOVA) was performed to specify the significant differences between the examined data groups at the 95% confidence level. To find relationships between radionuclides soil-to-leaves transfer factors and the physical and chemical properties of the soil, Pearson's linear correlation analysis was performed.

3. RESULTS AND DISCUSSION

3.1. ACTIVITY CONCENTRATIONS IN THE SOIL

The results of the determination of radionuclides activity concentrations (Bq kg^{-1} dry weight) and some basic physical and chemical properties of soils in the Belgrade parks are presented in Table 1. All soil profiles from the parks were alkaline and with the lowest $CaCO_3$ content in the topsoil increasing with depth (together with soil pH), indicating soil calcium-carbonate leaching over time. A significantly higher content for the coarse sand fraction was found at site SP, which was slightly deficient with finer soil fractions (silt and clay) compared to BG and KP. The KP soil was of silty clay loam compared to the silt loam at BG and SP. In our previous study of the same area was concluded that soil from BG has kept its original profile differentiation unlike the soil from SP and KP which natural structure has changed due to anthropogenic modifications [12].

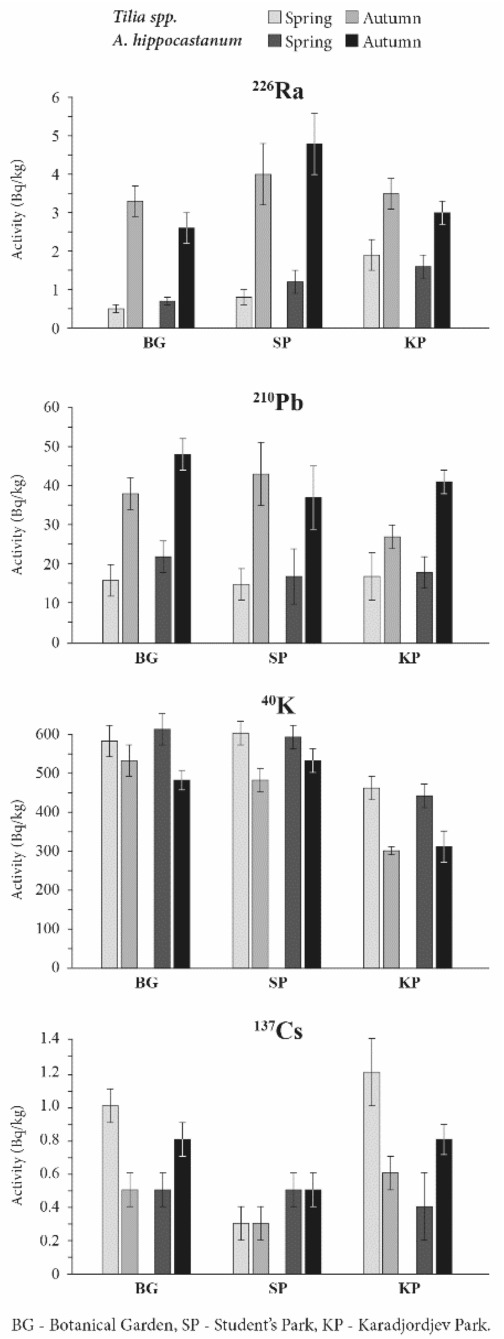
Table 1

The mean values of radionuclides activity concentrations (Bq kg^{-1} dry weight) and basic properties of soil at Belgrade parks BG, SP and KP

Measured parameter	BG	SP	KP	AM (SD)	Range
^{238}U	39	37	22	34.4 (9.4)	14–46
^{226}Ra	38	43	38	40.2 (4.2)	33–50
^{210}Pb	37	49	41	42.8 (9.2)	29–63
^{235}U	2.0	1.6	2.7	2.0 (0.5)	1.3–3.4
^{40}K	485	449	560	488 (48)	424–576
^{137}Cs	2.0	12.2	10.7	7.9 (6.2)	<mdc–19
pH H_2O	7.85	7.86	7.84	7.9 (0.2)	7.4–8.1
pH KCl	7.42	7.19	7.26	7.3 (0.2)	6.9–7.6
$CaCO_3$ (%)	20.4	23.1	6.9	18.3 (7.0)	3.8–26.0
SOM (%)	2.3	3.9	2.7	3.0 (1.2)	1.3–5.1
Coarse sand (%) (2000–200 μm)	4.7	14.8	3.0	8.2 (6.1)	1.6–21.1
Fine sand (%) (200–50 μm)	12.7	11.7	4.5	10.4 (4.2)	4.1–16.1
Coarse silt (%) (50–10 μm)	29.4	23.3	27.5	26.6 (4.5)	22.3–38.7
Fine silt (%) (10–2 μm)	29.0	27.5	32.3	29.2 (3.6)	21.7–34.6
Clay (%) (<2 μm)	24.3	22.8	32.6	25.6 (4.5)	20.4–33.2

3.2. ACTIVITY CONCENTRATION IN THE SAMPLES OF LEAVES

After joining individual samples of *A. hippocastanum* and *Tilia* spp. leaves, the obtained integral samples measurements showed that radionuclides ^{226}Ra, ^{210}Pb, ^{40}K and ^{137}Cs could be detected in each integral sample (n = 12) and their activity concentration variations according to season and park were illustrated in Fig. 1. Radionuclides ^{238}U and ^{235}U were detected in four samples of leaves in autumn. In one season, the activities of each radionuclide showed no differences in the leaves between chestnut and linden at each park, which is why they were grouped together according to season. Activity concentrations (Bq kg^{-1} dry weight) in the integral samples of leaves representative for the entire investigated period (2002–2012), together with their soil-to-leaves transfer factors are presented in Table 2.



BG - Botanical Garden, SP - Student's Park, KP - Karadjordjev Park.

Fig. 1 – Seasonal changes of the activity concentrations of ^{226}Ra, ^{210}Pb, ^{40}K and ^{137}Cs radionuclides in of *A. hippocastanum* (chestnut) and *Tilia spp.* (linden) leaf samples (error bars correspond to expanded uncertainties with factor k = 2).

Table 2

Activity concentrations (Bq kg^{-1} dry weight) of radionuclides in the integral samples of chestnut and linden leaves at Belgrade parks and their soil-to-leaves transfer factors (TF)

	Activity in leaves (Bq kg^{-1} dry weight)			TF (soil-to-leaves)	
		Range	AM (SD)	Range	AM (SD)
^{238}U	Autumn	2.7 – 12.0	6.7 (4.4)	$2.12 \times 10^{-1} – 3.16 \times 10^{-1}$	2.03×10^{-1} (8.91×10^{-2})
^{235}U	Autumn	0.13 – 1.00	0.4 (0.4)	$4.87 \times 10^{-2} – 4.90 \times 10^{-1}$	2.20×10^{-1} (2.11×10^{-1})
^{226}Ra	Spring	0.5 – 1.9	1.1 (0.5)	$1.30 \times 10^{-2} – 4.96 \times 10^{-2}$	2.82×10^{-2} (1.46×10^{-2})
	Autumn	2.6 – 4.8	**3.5** (0.8)	$6.77 \times 10^{-2} – 1.11 \times 10^{-1}$	**8.78×10^{-2}** (1.46×10^{-2})
^{210}Pb	Spring	15 – 22	17.5 (2.4)	$3.05 \times 10^{-1} – 5.88 \times 10^{-1}$	4.20×10^{-1} (0.98×10^{-2})
	Autumn	27 – 48	**39.0** (7.1)	$6.59 \times 10^{-1} – 1.28 \times 10^{0}$	**9.31×10^{-1}** (2.21×10^{-1})
^{40}K	Spring	440 – 610	547 (76)	$7.85 \times 10^{-1} – 1.34 \times 10^{0}$	1.12×10^{0} (2.50×10^{-1})
	Autumn	300 – 530	438 (106)	$5.35 \times 10^{-1} – 1.18 \times 10^{0}$	9.04×10^{-1} (2.85×10^{-1})
^{137}Cs	Spring	0.3 – 1.2	0.7 (0.4)	$2.46 \times 10^{-2} – 4.90 \times 10^{-1}$	1.58×10^{-1} (1.82×10^{-1})
	Autumn	0.3 – 0.8	0.6 (0.2)	$2.46 \times 10^{-2} – 3.92 \times 10^{-1}$	1.39×10^{-1} (1.48×10^{-1})

* Values significantly different compared to spring at the $p < 0.05$ level are given in bold letters

^{226}Ra activity increased during the vegetative period (Fig. 1) and radium is expected to accumulate in leaves because when taken by the tree roots it is moved up through the transporting tissue (xylem) and after interception by some of the tree organ it stays there immobile without further distribution or transportation back [13]. It is known that ^{210}Pb is intercepted by plants through the foliar uptake by dry or wet atmospheric deposition and from the soil through root uptake to some extent, while the contribution from each of these two sources is difficult to assess [3]. In this study, the maximum ^{210}Pb activities in leaves in autumn coincided with ^{210}Pb natural production in the atmosphere from the decay of noble gas radon whose emanation from soil also reaches a maximum in the autumn [14]. So, the ability of the investigated leaves to trap and retain atmospheric particles (aerosols) containing ^{210}Pb of natural origin could be reflected in the ^{210}Pb increase in leaves from spring to autumn. Anthropogenic sources are considered to contribute less than 1% to the total ^{210}Pb in the air [6].

As an essential element for plant growth and development, potassium and its radioactive isotope concentrations in leaves are tree and soil specific, which is why ^{40}K ranges in leaves can vary greatly [8, 15]. In the parks, ^{40}K decreased in leaves from spring to autumn (Fig. 1) since it was most likely leached or translocated back to the roots during the last part of the season, but the decrease was not statistically significant. The source of most ^{137}Cs in the soil of Belgrade city parks at the present time is radioactive fallout of nuclear fission products from the Chernobyl accident. As ^{137}Cs is mainly below the minimum detectable activity in the air [14], it arrived to the chestnut and linden leaves dominantly by the root uptake as a potassium analogue. The lack of specific seasonal pattern is observed for ^{137}Cs (Fig. 1).

In order to compare the present activity measurements with other studies, some of the available activity concentration levels in the leaves of broadleaf species grown in the soils at background sites and corresponding soil-to-leaves transfer factors are given in Table 3. It was found that soil-to-leaf concentration ratio values of stable elements and corresponding radioisotopes in the tree leaves are not significantly affected by climate differences [16].

Table 3

Activity concentration (Bq kg^{-1} dry weight) ranges of ^{238}U, ^{226}Ra, ^{210}Pb, ^{40}K and ^{137}Cs in the leaves of broadleaf species at background sites and its soil-to-leaves transfer factors (in brackets) found in the literature

	Activity concentration in leaves (Bq kg^{-1} dry weight) (TF soil-to-leaves)				
Literature source	^{238}U	^{226}Ra	^{210}Pb	^{40}K	^{137}Cs
[7][1]	1.68–13.0 –	7.4–37.3 –	10.3–20.3 –		
[8][2]	8.5–11.9 (0.303–0.354)			163–219 (0.801–0.945)	
[9][3]		2.0–13.2 (0.06–0.37)		50.0–369.8 (0.35–2.60)	
[15][4]		0.34–8.21 (0.03–0.65)		59.2–1207.7 (0.32–5.27)	0.2–6.6 (0.05–1.53)
[17][5]	– (0.017)		– (0.71)	– (1.4)	

[1] Oak tree, Eucalyptus tree, Acacia tree

[2] *Evodia roxburghiana, Turpinia pomifera, Eugenia arnottiana, Elaeocarpus oblongus, Glochidion neilgherrense*

[3] *Terminalia paniculata* Roth., *Careya arborea* Roxb., *Diospyros malabarica*

[4] *Terminalia catappa* L., *Anacardium occidentale* Linn., *Magnolia champaca*, *Syzygium cumini* L., *Mangifera indica* L., *Tectona grandis* L. f.

[5] Apple tree

3.3. SOIL-TO-LEAVES TRANSFER FACTORS

Soil-to-plant TF values for radium (+II) are usually found to be higher than that for uranium (+IV) [2], but in this study ^{226}Ra TF means from each season were significantly lower than that for uranium ($p < 0.05$). Even though ^{226}Ra was continuously available, uranium translocation and accumulation in the leaves starting from the moment of its higher availability in the late summer exceeded radium's activity build up in the leaves during the entire vegetation period. This could be explained by uranium's TF high and positive correlation with Ca-carbonates ($p < 0.01$) and pH ($p < 0.001$) in autumn. The high $CaCO_3$ content in the park's alkaline soils is the main condition for higher uranium availability [6], but conditions for its transfer from soil to the leaves were met at the end of the vegetation period, most probably as a result of soil pH increase. The pH values of soils also increased from July to October at a rehabilitated uranium mine and favoured ^{238}U and ^{226}Ra solubilization and availability [18].

For uranium, the main positive correlation was found between the fractions of sand (2 – 0.05 mm) and TF values of ^{238}U ($p < 0.01$) and between fine sand (0.2 – 0.05 mm) and ^{235}U ($p < 0.05$). Sand particles could have influenced uranium soil-to-leaves transfer because a greater percentage of uranium's labile fraction is found to originate from sand (coarse or fine) than from other fractions of soil [5]. ^{226}Ra soil-to-leaves TFs showed significant positive correlations with fine silt fractions (0.01 – 0.002 mm) in the spring ($r = 0.82$, $p < 0.05$) and with the coarse sand (2 – 0.2 mm) in the autumn ($r = 0.83$, $p < 0.05$). Comparably, sand and labile fractions extracted from silt were identified as the main fractions with the capacity to liberate radium when considered as a substrate for plant growth instead of the bulk soil [2]. This suggests conditions for ^{226}Ra availability and uptake by roots of higher plants have changed during the vegetation season and at the end of it, radium transfer to the leaves became connected to the coarsest soil particles as it was in the case of uranium uptake. It seems that in autumn, with pH rise, the source of both radionuclides availability are the coarsest particle sizes which might be associated with the mechanism of reversible (physical) sorption by soil and not the finest particle sizes which are rather the cause of radionuclides retention due to irreversible (chemical) soil sorption [11].

There might be contributions from the chestnut and linden root uptake in spring based on the ^{210}Pb TFs positive correlation with coarse silt ($r = 0.83$, $p < 0.05$) and in the autumn correlations were not found. Uneven cumulative effect of ^{210}Pb deposited from the air to the leaves could mask potential correlations with the soil properties, but it is more likely that root uptake is totally suppressed because of the strong lead sorption in the presence of higher Ca-carbonates in the park's alkaline soil [6]. ^{137}Cs and ^{40}K TF means couldn't be distinguished between seasons in the parks. In each season, the main positive correlation was found between the fractions of coarse silt (0.5 – 0.1 mm) and TFs of ^{137}Cs ($p < 0.05$) and negative one between fine silt (0.1 – 0.002 mm) and ^{40}K ($p < 0.01$). At the SP site, which was the site of highest cesium activities in the soil, the lowest cesium TF values are found and it was opposite for ^{40}K transfer. This behavior could be explained by most probable potassium-based fertilization of soil usually used to provide larger available potassium fraction which consequently suppresses ^{137}Cs uptake and reduces its transfer to plants [4].

4. CONCLUSIONS

In one season, the activities of natural radionuclides including fall-out ^{137}Cs, showed no differences between chestnut and linden leaves. The TF values means of ^{226}Ra and ^{210}Pb were significantly lower in spring than in autumn, while no differences were found for ^{40}K and ^{137}Cs between the seasons. In the alkaline park's soils, according to the present analysis, the availability of radionuclides appeared to be affected by the granulometric distribution of the soil. Fractions of silt were positively correlated to the TFs of ^{210}Pb and ^{226}Ra in spring and ^{137}Cs during the entire vegetative period. Fractions of sand (coarse and/or fine) were positively associated to the TFs of ^{238}U, ^{235}U and ^{226}Ra in autumn and ^{40}K in spring and autumn.

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