ANALYSIS OF ^{238}U, ^{226}Ra, AND ^{210}Pb TRANSFER FACTORS FROM SOIL TO THE LEAVES OF BROADLEAF TREE SPECIES

by

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This analysis of ^{238}U, ^{226}Ra and ^{210}Pb transfer factors from the soil to the leaves of different native broadleaf trees at sites previously modified by uranium presence and at the site of background radioactivity levels, was conducted using data from a few available studies from the literature. The broadleaf tree species *Quercus ilex, Quercus suber, Eucalyptus camaldulensis, Quercus pyrenaica, Quercus ilex rotundifolia, Populus sp. and Eucalyptus botryoides Sm*. **at the affected sites and** *Tilia spp*. **and** *Aesculus hippocastanum L*. **at the background site were included in the study regardless of the deciduous or evergreen origins of the leaves. In the papers cited here, data about basic soil parameters: pH, total Ca [gkg^{-1}], sand [%], and silt + clay [%] fractions were also available. All the collected data of activity concentration [Bqkg^{-1}] dry weight in the soil ($n=14$) which was in the range: 22-6606 for ^{238}U, 38-7700 for ^{226}Ra, and 37-7500 for ^{210}Pb, and the tree leaves in the range: 2.7-137.6 for ^{238}U ($n=10$), 2.6-134.2 for ^{226}Ra ($n=14$), and 27-77.2 for ^{210}Pb ($n=14$), indicated that it was normally distributed after log-transformation. The present study was conducted under the hypothesis that biological differences between the examined broadleaf tree species have a lesser influence on the transfer factors of the investigated radionuclides from soil to tree leaves compared to the impact of the soil parameters and radionuclides activity concentrations in the soil. Consequently, it was examined whether ^{238}U, ^{226}Ra, and ^{210}Pb soil-to-leaves transfer factor values for average broadleaf species could be predicted statistically in the first approximation based on their activity concentration in the soil and at least one basic soil parameter using multiple linear regression.**

Key words: natural radionuclide, soil-to-leaves transfer factor, root uptake, foliar uptake

INTRODUCTION

The naturally occurring radionuclides ^{238}U, ^{226}Ra, and ^{210}Pb are non-essential elements, but may enter the trees the same pathways as an essential element: via the root uptake or foliar deposition. The radionuclides ^{238}U, ^{226}Ra, and ^{210}Pb, present in soil in the form available for uptake, may be taken by the tree roots, translocated to the xylem and then to the tree leaves promoted by the transpiration of water via the leaves [1]. Additionally, the airborne part of naturally occurring ^{210}Pb may be directly deposited on the outer cell layer of the leaf surfaces and absorbed into the leaf tissue through the pathway of foliar uptake [1, 2]. After uptake, radionuclides may be translocated, leached or accumulated within parts of the tree (the roots, leaves, twigs, among others) [2-5]. The study of elements detected in the trees of natural boreal vegetation showed that an element (and its isotope) would exhibit a different leaf-to-twig concentration ratio depending on its role and origin in the tree [3]. In the case of the deciduous *Betula pubescens* tree, leaf-to-twig ratio values of 1.5-2 for potassium (an essential-nutrient) >0.6-1.0 for uranium (non-essential-soil derived) >0.3-0.6 for lead (nonessential-atmospherically derived), were obtained. Natural radioactive isotopes that are transferred from the soil to the roots and then to the leaves, may be deposited in the soil during the decomposition of fallen leaves and be continuously recycled by the trees due to their long half-life.

Radionuclides belonging to the uranium decay series, were detected in the samples of tree leaves collected at Canadian background sites using various methods (alpha spectroscopy, gamma spectroscopy, beta counting, gas flow proportional counting) [2]. However, the method detection limit was not sufficiently sensitive to detect those radionuclides in all of the *background* samples, despite the collection of very large plant samples which allowed ashing as a pre-concentration step in the analysis. So, it is more difficult to detect ^{238}U, ^{226}Ra, and ^{210}Pb isotopes in the tree leaves at background sites, unlike at sites in the vi-

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cinity of uranium mining facilities, where levels of uranium and its decay products can be relatively high in the samples of soils and plants [6-8]. For example, an effect of radionuclide accumulation in leaves was clearly evident at a former uranium mine, with activity concentrations much higher in the old leaves of *Quercus suber* (for ^{238}U and ^{210}Pb) and *Eucalyptus camaldulensis* (for ^{226}Ra) compared to those in the young leaves [9].

The soil-to-plant transfer factor (TF) is expressed as the ratio between the radionuclide activity concentration in the plant tissue, or the plant part such as the leaf [Bqkg^{-1}] (dry weight plant tissue), to that in the soil [Bqkg^{-1}] (dry weight soil). The TF is commonly used to quantify radionuclide uptake from the soil by plants and it is assumed that transfer primarily depends on root uptake within the soil layer of a standardized thickness, compared to the less significant plant contamination (such as foliar uptake and soil adhesion by resuspension). A standardized depth of 20 cm for all crops and the fruit trees is adopted, assuming that the bulk root density is usually found in that surface layer of soil [5].

By definition, transfer from soil to plant is considered as an equilibrium process when the naturally occurring radionuclide activity concentration in the plant is linearly related to its concentration in the soil, implying a constant TF independent of soil concentration [10-13]. But, because plant uptake is a complex process affected by characteristics such as the radionuclides' physicochemical form, soil type, the physical and chemical properties of the soil, the type and the biological properties of plants and climate conditions, a deviation from linearity and a large variability of measured TF values is produced [12-15]. To reduce this variability, soil-to-plant TF values for the natural radionuclides (uranium, thorium, radium, lead, polonium) were grouped according to major crop type and to soil group, as defined by their texture and organic matter content (sand, loam, clay and organic) and the generic TF values were recommended [12].

The TF deviates from linearity because of the characteristic uptake of radionuclides by plants which is greater at their low concentrations in soil, asymptotically reaching constant (saturation) value at high soil concentrations that is very well described by a non-linear function [13]. This non-linearity of the transfer of a radioactive isotope was found to be equally explained by its total as well as mobile soil concentrations [16, 17]. Radionuclides are usually tolerated by the tree roots on their low activity concentrations in the soil. Based on their response to increasing concentrations in the soil, there are species whose root uptake would differ from that of an average tree species and may be categorised as an excluder species (the uptake of radionuclides is restricted by an exclusion mechanism), an indicator species (concentrations in leaves linearly reflect concentrations in the soil) or an accumulator species (radionuclides accumulate in tree leaves without symptoms of toxicity until a critical threshold level in soil is reached). In any case, the TF behaviour of both tolerant and non-tolerant species is described by the non-linear function in the wide range of concentrations in the soil [1, 13, 16, 17].

In this work, it was examined whether there were any differences in ^{238}U, ^{226}Ra, and ^{210}Pb TF values from the soil to the leaves of broadleaf trees between the sites affected by uranium mining and the sites with the background radioactivity level, by using available data from a few studies found in the literature (see Materials and methods). Natural radionuclides were assumed to be in the *steady-state* between the soil and the trees because the radionuclide activity concentration in the soil is not changing over time (after a sufficient time after contamination at affected sites). That degree of contamination of the trees is determined by root uptake for ^{238}U and ^{226}Ra and by root uptake and foliar deposition in the case of ^{210}Pb [5]. It was hypothesized that the biological differences between the examined broadleaf tree species would have less influence on the transfer of the investigated radionuclides from the soil to the tree leaves, compared to the impact of their activity concentrations in the soil and site-specific soil parameters. Since the cited papers contained data about basic soil parameters (pH, total Ca, percentages of sand and silt + clay), it was examined whether ^{238}U, ^{226}Ra and ^{210}Pb soil-to-leaves TF values, for average broadleaf species, could be predicted by their activity concentration in the soil and at least one basic soil parameter using multiple linear regression.

MATERIALS AND METHODS

The broadleaf tree species which soil-to-leaves TF values are investigated in this work were: *Quercus ilex, Quercus suber, Eucalyptus camaldulensis* [9], *Quercus pyrenaica, Quercus ilex rotundifolia, Populus sp.* [18] affected by uranium mining; and *Eucalyptus botryoides* Sm. affected by uranium-rich coal mining [15]. At the site considered as background, the broadleaf tree species were *Tilia* spp. (linden) and *Aesculus hippocastanum* L. (horse chestnut) [19]. The collected activity concentration data of ^{238}U, ^{226}Ra and ^{210}Pb, determined in soils and leaves, are presented in tab. 1. When all the available data are grouped together, the activity concentrations [Bqkg^{-1}] dry weight in the soil were in the range (n = 14): 22-6606 for ^{238}U, 38-7700 for ^{226}Ra and 37-7500 for ^{210}Pb and in the tree leaves: 2.7-137.6 for ^{238}U (n = 10), 2.6-134.2 for ^{226}Ra (n = 14) and 27-77.2 for ^{210}Pb (n = = 14). It was estimated that the measurement uncertainties of TF values, obtained in the cited works, were in the range of 15 % to 30 %, except where [9] they were in the range of 2 % to 10 %. Data about basic soil properties were also available in these cited works and are included in tab. 1. The ranges of their values were: 4.40-7.86 for

Table 1. Retrieved data of ^{238}U, ^{226}Ra, and ^{210}Pb dry weight activity concentration [Bqkg^{-1}] in the samples of the soil and the leaves and the mean values of basic soil properties

	^{238}U [Bqkg^{-1}]		^{226}Ra [Bqkg^{-1}]		^{210}Pb [Bqkg^{-1}]		Soil pH	Ca [gkg^{-1}]	Sand (2-0.05 mm)	Silt + clay (<0.05 mm)	
	Soil	Leaves	Soil	Leaves	Soil	Leaves					
Uranium mine [18]	64-81 1038-2200	<mdc <mdc-5	81-113 4155-7700	8-16 19-20	157-169 4793-7900	29-75 54-56	5.8^{a} 6.1^{b}	2.2 2.3	79.6	9.8	10.6
Uranium mine rehabilitated [9]	52-344 6606	17-138 38-70	216-305 354	35-134 17-27	321-739 227	38-69 50-77	6.5^{c} 4.6^{d}	7.0 1.7	87.2*	12.8*	
U-rich coal mine [15]	31.5	6.3	–	–	–	–	4.4^{e}	–	27.3	50.2	22.5
Background site [19]	22-39	<mdc-12	38-43	2.6-4.8	37-49	27-48	7.8^{f}	6.7	17.1	56.8	26.6

Note: *– Sand: 2 – 0.067 mm, Silt + clay: <0.067 mm
a – *Quercus pyrenaica, Quercus ilex rotundifolia*, b – *Populus sp. 1 and 2*, c – *Eucalyptus camaldulensis, Quercus suber 1*, d – *Quercus suber 2*, *Quercus ilex*, e – *Eucalyptus botryoides Sm*, f – *Tilia spp.1, 2, 3 and A. hippocastanum 1, 2, 3*

pH, 1.7-11 gkg^{-1} for total Ca, 7.4 %-91 % for sand fraction and 9 %-92.5 % for the sum of silt + clay fractions.

The normality of the datasets was evaluated using the Shapiro-Wilk's test and statistical analysis was conducted at the 95 % confidence level. Due to the scarcity of some data, it was accepted that datasets indicated normal distribution if the values of standardised skewness and kurtosis were within the range of –2 to +2. One-way analysis of variance (ANOVA) was performed to specify the significant differences at the 95 % confidence level between the examined data groups from the sites considered as affected and as background. In order to predict TF values based on the available data, multiple linear regression was performed because this might explain how a single response variable (TF) depends linearly on a number of predictor variables (soil activity concentrations and basic soil properties). To interpret results of regression, apart from the F-test statistic and p-value, which are reported in the regression (ANOVA table), the value of R^2 (the proportion of variation in the response that is explained by the regression of all the predictors in the model) and the adjusted R^2 was used [20]. To test the null hypothesis that the standard deviations of TF values within each site (affected and background) is the same, Levene's test was used in which the p-value suggests that there is not a statistically significant difference amongst the standard deviations at the 95 % confidence level if p 0.05.

RESULTS AND DISCUSSION

Activity concentration in leaves *vs.* in the soil

Datasets of all the collected ^{238}U, ^{226}Ra, and ^{210}Pb activity concentration values in the soil were normally distributed after log-transformations. When divided into two groups coming from an affected and the background site, respectively, the soil activity concentration geometric mean (GM) values and ranges were [Bqkg^{-1}]: 404.6 (31.5-6606) and 31.8 (22.3-39.0) for ^{238}U, 471.9 (81.0-7700) and 39.9 (38.3-43.2) for ^{226}Ra, and 577 (157.0-7900) and 42 (37.4-49.2) for ^{210}Pb. The activity concentrations in tree leaves also followed log-normal distribution. At the affected and background site, respectively, their GM values and ranges were [Bqkg^{-1}]: 24.0 (5.0-137.6) and 5.5 (2.7-11.7) for ^{238}U, 23.8 (8.1-134.2) and 3.5 (2.6-4.8) for ^{226}Ra and 53.4 (29.0-77.2) and 38.4 (27-48) for ^{210}Pb.

When significance of differences was tested, only means of ^{210}Pb could be distinguished between background (39 Bqkg^{-1}) and affected (56 Bqkg^{-1}) sites ($p < 0.05$). The ^{238}U and ^{226}Ra mean activity concentrations, respectively, were higher at the affected (45.5 Bqkg^{-1} and 23.8 Bqkg^{-1} than at the background site (6.7 Bqkg^{-1} and 3.5 Bqkg^{-1}), but the difference was not found to be significant at the 5.0 % significance level. Statistically, none of the activity concentration values in leaves showed to be a significant outlier. It could be said that natural radionuclides inclined to be saturated in leaves because the activity concentration in leaves (C_{leaves}) could have been fitted well enough by the non-linear function of concentrations in soil (C_{soil}) which was approaching one saturation value in the leaves at higher values for ^{238}U ($r = 0.92$, $p < 0.05$, $n = 8$), ^{226}Ra ($r = 0.78$, $p < 0.01$, $n = 12$) and ^{210}Pb ($r = 0.62$, $p < 0.05$, $n = 12$).

Furthermore, in the lower, narrow interval of soil activity concentrations of 20-350 Bqkg^{-1} there was evidence of a significant ($p < 0.0001$) linear relationship: $\ln C_{\text{leaves}} = \alpha (\ln C_{\text{soil}}) + \beta$ in which the slope was found to be approximately 1 for ^{238}U (α 1.4) and ^{226}Ra (α 1.1). Based on these statistical results, differences between the investigated broadleaf species could not be observed since there was a great similarity between the uptake responses of leaves on increasing ^{238}U, ^{226}Ra, and ^{210}Pb activity concentration in soil.

Activity concentration in leaves *vs.* soil parameters

Datasets of soil property values of pH, total Ca, sand and silt + clay fractions were normally distributed as well. It can be seen in tab. 1 that in soil affected by uranium mining, the predominant soil particle size fraction was sand (>80 %) and in the soil affected by uranium-rich coal mining and the background site it was a fraction of silt + clay (>70 %). Hence, the soil texture of the investigated samples varied widely from sandy to silty clay loam. The main result obtained was a statistically significant exponential increase of activity concentration in leaves with percentages of sand fractions of soil for ^{238}U ($r = 0.74$, $p < 0.05$, $n = 9$), ^{226}Ra ($r = 0.94$, $p < 0.0001$, $n = 13$) and ^{210}Pb ($r = 0.70$, $p < 0.01$, $n = 13$), which suggest that radionuclides have a tendency to be more accumulated in leaves where there is an increase of the coarsest soil fraction. This result can be explained knowing that sorption-desorption processes influence radionuclide concentration in soil solution and consequently its mobility and availability to plants [21]. One of the ways to describe radionuclide interaction in soils (accumulation in soil *vs* leaching from soil) is the solid-liquid distribution coefficient (Kd) determined as the ratio of the concentration of a radionuclide in the soil solid phase to the concentration in the soil solution (Lkg^{-1}) [5, 12]. Consisting of larger grain sizes with small specific surface areas, coarse textured soils have a low capacity for radionuclides retention that may lead to their longer presence in the soil solution. Accordingly, coarse sand (0.5-2 mm) and fine sand (0.067-0.5 mm), respectively, were the two main soil structural fractions which were found to have an effect on the distribution coefficient of ^{238}U and ^{226}Ra in the natural soil investigated [22], since the smallest Kd values were obtained exactly for those two fractions of soil. Hegazy *et al*. [23] also observed significant positive correlations between the uranium content in guava and mango leaves and the coarse sand fractions of soil and reported that uranium was more highly accumulated in the plants in coastal black sand soils with a higher percentage of coarse sand (>65 %) than in cultivated inland clayey soils.

Concerning the ^{210}Pb, study by Prakash *et al*. [24] of ten vertical soil profiles of 0-30 cm depth (containing 40-80 % particles of sand) demonstrated a significant positive correlation between the ^{210}Pb activity concentration in soil and the percentages of particles of sand (2-0.02 mm), while correlation coefficients were found to be stronger in each deeper layer of soil (0-10 cm *vs* 10-20 cm *vs* 20-30 cm). It was observed there that the percentage of sand particles of soil was decreasing lengthwise (0-30 cm) and hence concluded that the porosity of soil changed according to depth along with the number of micropores and their dimensions. So, the positive relationship between ^{210}Pb and the percentages of sand have been explained by the pore spaces distribution in the soil. In the upper layer, with higher ^{210}Pb activity concentration and higher sand content, larger pore spaces enabled grater diffusion of ^{222}Rn atoms (into and out of the soil surface) which subsequently decayed to ^{210}Pb. Oppositely, in the deeper soil layer with the decrease of coarse soil particles, ^{222}Rn atoms stayed blocked in the smaller pore spaces of soil which resulted in lower ^{210}Pb activity concentration in the soil. In the study of Girault *et al*. [25] (and references therein), there were no significant systematic differences of the ^{222}Rn emanation coefficients for soils between different soil types (clay, silt, sand, gravel and till) (mean varied from 0.17 to 0.24) and especially between sands (with a mean 0.242) and even between plants that included leaves of multiple broadleaf tree species (range of variation 0.82-0.95). This could lead to the conclusion that small ^{210}Pb activity concentration differences can be expected in the leaves of average broadleaf trees (if not known to be hyperaccumulator or excluder species) grown in soils of different textures. In the present study, in the soils from silty clay loam to sandy texture (with 64 % of sand variation), the total ^{210}Pb activity in the tree leaves (27-77 Bqkg^{-1}) varied only 33 %. A very similar overall variability was noted for the ^{210}Pb activity concentration (35 %) in the samples of leaves of oak (*Quercus robur*) grown in the environment influenced by the waste disposal from the aluminium industry and the samples of leaves of the same species outside of its influence [26].

Transfer factor *vs* soil activity concentration

Shapiro-Wilk's test of normality of a given TF values datasets for ^{238}U, ^{226}Ra, and ^{210}Pb revealed a normal frequency distribution. The results of the examination of whether there were any significant differences between radionuclides TF values from the affected and background site are presented in tab. 2. The ^{238}U and ^{226}Ra TF means of 0.156 and 0.125, respectively, from affected

Table 2. ANOVA table for soil-to-leaves TF values for broadleaf species

Parameter	TF ^{238}U		TF ^{226}Ra		TF ^{210}Pb	
	Bqkg^{-1} per Bqkg^{-1} of dry weight					
	Affected ($n = 6$)	Background ($n = 4$)	Affected ($n = 8$)	Background ($n = 6$)	Affected ($n = 8$)	Background ($n = 6$)
Minimum	0.0002	0.121	0.0026	0.068	0.0071	0.658
Maximum	0.4	0.316	0.44	0.111	0.478	1.283
AM (ASD)	0.156 (0.177)	0.203 (0.089)	0.125 (0.145)	0.088 (0.015)	0.180 (0.163)	0.931 (0.222)
GM (GSD)	0.026 (0.050)	0.189 (0.644)	0.051 (0.165)	0.089 (0.846)	0.093 (0.214)	0.909 (0.789)
F-ratio (p-value)	0.23 ($p > 0.05$)		0.39 ($p > 0.05$)		53.73 ($p < 0.0001$)	
Levene's test (p-value)	3.06 ($p > 0.05$)		2.88 ($p > 0.05$)		0.60 ($p > 0.05$)	

sites modified by uranium presence could not be distinguished from that of 0.203 and 0.088, respectively, at the background site, tab. 2. When comparing uranium plant-soil concentration ratios between sites uncontaminated and contaminated by uranium, some differences for the deciduous trees and shrubs leaves were observed, but were not significantly different either [5]. Ranges of ^{226}Ra TF values, from the present study, of 0.0026-0.44 at the affected site and 0.068-0.111 at the background site, could be compared with that obtained for the tree leaves (*Quercus robur, Castanea sativa, Tilia ×europaea, Quercus ilex*) collected exclusively at the background sites: 0.071-0.231 of granitic and metamorphic origin and 0.0022-0.137 of calcareous and sedimentary origin at sites characterized as not contaminated sandy soils [25].

In the present study, only the mean ^{210}Pb TF value was found to be significantly different at the background site (0.931) compared to the affected site (0.180) which implies that transfer was higher in the soils of silty clay loam then that of sandy texture. As suggested by Tagami and Uchida [14], soil-to-tree leaves TF values can be evaluated using TF values for leafy vegetables. The TF data for the same genus of trees would be preferable to use since differences in the leaves of the same genus are smaller, however, data for leafy vegetables might be used for the tree leaves since the function of leaves is the same amongst plants. TF values of U, Ra, and Pb, according to the soil group and their arithmetic mean (AM) values, [12] were used here for comparison. The ^{238}U TF AM of 0.175, in this study, was found to be comparable (same order of magnitude) to the AM of 0.221 for leafy vegetables given for all types of soil and ^{226}Ra TF mean of 0.109 with the AM value of 0.161 was given for the Loam group of soils (no data for Sand soils). Also, the lower ^{210}Pb TF AM value of 0.078 for sand soils and higher of 0.817 for loam soils for leafy vegetables were found to be analogous to the AM values of 0.180, in this study at the site with sandy texture (affected) and of 0.931 at the site with loamy texture (background), tab. 2. The origin of the differences between ^{210}Pb soil-to-leaves TF appears to be more associated to the soil type *i. e.*, its texture, then to the type of broadleaf tree species in the wide range of activity concentrations in the soil (10^1-10^3 Bqkg^{-1}).

Prediction of soil-to-leaves transfer factor by soil parameters

Further analysis was done after excluding the soil-to-leaves TF value for *Eucalyptus camaldulensis* as a statistically significant outlier for ^{226}Ra (0.44) and ^{238}U (0.4), but not in the case of ^{210}Pb (0.093). The uptake of elements by Eucalyptus camaldulensis is considerably high because it is a tree species well known for developing a deep root system capable of accumulating considerable levels of metals and with a high ability for their translocation from roots to the aboveground parts of the tree [15]. At a uranium mill tailings site, the median TF value of *Eucalyptu*s *sp*. leaves was found to be higher for ^{226}Ra (0.032) compared to ^{210}Pb (0.0087) [6] and this deficit of ^{210}Pb in the leaves is most likely due to lower mobility and transfer of ^{210}Pb from the substrate to the roots and from the substrate to the leaves when compared to ^{226}Ra.

In order to examine first to what extent soil-to-leaves TF values depend on the activity concentration in soil, due to log-normal distributions of activity concentrations in the soil, the TF values were log-transformed before the regression. Logarithms of TF values could be fitted well by log-activity concentrations of soil ($p < 0.01$ or higher) for ^{238}U ($R^2 = 67.0$ %, F-ratio = = 14.24), ^{226}Ra ($R^2 = 75.2$ %, F-ratio = 33.42), and ^{210}Pb ($R^2 = 96.9$ %, F-ratio = 372.46), tab. 3. This indicated that more than 65 % of U, Ra, and Pb TF variations might be explained only by the decreasing trend of corresponding activity concentration in the soil, regardless of biological differences between broadleaf species. That the soil concentration is one of the main factors causing TF variation (and decrease) is also shown for uranium and lead TF for the parts of the tree (leaf, needle, root), at two boreal forest sites [11]. Similarly, Noskova *et al*. [27] reported that for the aboveground plant parts of *Sorbus aucuparia* and *Betula pubescens* trees ^{238}U uptake deviated less from the mentioned decreasing trend compared to ^{226}Ra because radium uptake by plants might be more sensitive to changes in the physicochemical characteristics of soil.

It was examined whether the results would be improved if one more predictor (*i. e.*, the known soil parameter) in addition to the soil activity concentration was included in the analysis. The simplest statistically significant model showed to be the one which included soil activity concentration and the percentage of sand fraction of the soil, tab. 3. Sand percentage (which varied from 7.4 % to 91 %) could be an important predictor because natural radionuclides TF values are associated with the soil texture (percent proportion of sand, silt and clay) given that the highest soil-to-plant TF values were found in coarse textured soils and the lowest in fine textured soils [12]. Based on the F-ratio and R^2 values, tab. 3, now more than 90 % of TF variations have been explained for ^{238}U ($R^2 = 93.7$ %, F-ratio = 37.45), ^{226}Ra ($R^2 = 97.8$ %, F-ratio = 200.51), and ^{210}Pb ($R^2 = 98.3$ %, F-ratio = 290.48). The fit of the data was better if the TF value of *Quercus pyrenaica* for ^{226}Ra and ^{210}Pb and of *Populus sp. 2* in the case of ^{238}U was excluded. The sand percentage coefficient estimate was more significant in the case of ^{226}Ra TF ($p < 0.001$) compared to the ^{238}U ($p < 0.1$) and ^{210}Pb TF ($p < 0.1$). In conclusion, in the case of ^{210}Pb, the site-specific percentage of sand is probably not an important predictor since its addition only improved regression results by ~1 % and the F-ratio value decreased (from 372 to 290).

Table 3. Results of simple and multiple linear regression

	X_i	$\hat{\beta}$		R^2 (R^{2*})	F-ratio (p – value)
ln (TF ^{238}U) (n = 9)	ln (^{238}Usoil) Const.	–0.8207** 1.1386	0.1127 0.2175	67.046 (62.338)	14.24 (0.01)
ln (TF ^{238}U) (n = 8) ^{o}	ln (^{238}Usoil) Sand (%) Const.	–0.7730***	0.1127 0.0073 0.3904	93.7418 (91.2386)	37.45 (0.001)
ln (TF ^{238}U) (n = 8) ^{x}	ln (^{238}Usoil) Silt+clay (%) pH Ca (gkg^{-1}) Const.	–1.8285** 0.0790** –4.4713** ** **	0.1674 0.0087 0.3664 0.0537 2.9378	99.4545 (98.7272)	136.74 (0.001)
ln (TF ^{226}Ra) (n = 13)	ln ($^{226}Ra_{soil}$) Const.	–0.6176*** 0.0258	0.1068 0.5703	75.238 (72.987)	33.42 (0.000)
ln (TF ^{226}Ra) (n = 12) □	ln ($^{226}Ra_{soil}$) Sand (%) Const.	–0.9678*** 0.0258*** **	0.0506 0.0027 0.1957	97.805 (97.3172)	200.51 (0.000)
ln (TF ^{226}Ra) (n = 13) □	ln ($^{226}Ra_{soil}$) pH Ca (gkg^{-1}) Const.	–0.7776*** –0.6067** 0.1065* 4.5575**	0.0832 0.1382 0.0434 1.1232	92.1367 (89.5156)	35.15 (0.000)
ln (TF ^{210}Pb) (n = 14)	ln (2^{10}Pbs$_{oil}$) Const.	–0.9153*** 3.3941***	0.0474 0.2611	96.8788 (96.6187)	372.46 (0.000)
ln (TF ^{210}Pb) (n = 13)□	ln ($^{210}Pb_{soil}$) Sand (%) Const.	–1.0064*** 0.0063 3.5722***	0.0565 0.0029 0.2204	98.3079 (97.9694)	290.48 (0.000)
ln (TF ^{210}Pb) (n = 14)	ln ($^{226}Ra_{soil}$) Silt+clay (%) pH Ca (gkg^{-1}) Const.	–0.8091*** 0.0308*** –1.0170*** 0.1076** 7.5800***	0.0577 0.0058 0.1763 0.0315 0.8395	98.4851 (97.8118)	146.28 (0.000)

Without mark $p < 0.1$, *$p < 0.05$, **$p < 0.01$, ***$p < 0.001$
TF values excluded in the regression: – *E. camaldulensis*, º – *Populus sp. 2*, – *E. botryoides Sm.*, □ – *Q. pyrenaica*

Finally, multiple linear regression was done for ^{238}U, ^{226}Ra, and ^{210}Pb TF if all known predictors (soil parameters) were included in the analysis besides the soil activity concentrations. These results are presented in tab. 3. In the case of ^{238}U TF, the coefficient estimates of silt + clay fraction, the pH and the total Ca in soil were significant ($p < 0.01$) and their inclusion considerably improved regression results ($R^2 = 99.45$ %, F-ratio = 136.74). In the case of ^{226}Ra TF, the pH ($p < 0.01$) and the total soil Ca ($p < 0.05$) were significant predictors, but grain size showed to be not important at all, while according to the F-ratio the regression quality decreased to a great extent (from 200.51 to 35.15). Al-Masri *et al*. [21] reported that lower TF values of ^{226}Ra then the ^{238}U from soil to the leaves of coriander and parsley were probably a result of the chemical similarity between Ca and ^{226}Ra that compete for the same absorption sites in the plants. The influence of Ca content in soil is well known because ^{238}U and ^{226}Ra concentrations in soil solutions and their availability to plants may be enhanced if exchangeable Ca concentration levels in soil solutions are low, which is why a negative relationship should be expected, but here the relationship was positive. Soil Ca level was actually lower at the affected site (~3 gkg^{-1}) compared to the background site (~7 gkg^{-1}). In contrast, the GM activity concentrations in leaves for ^{238}U and ^{226}Ra were higher at the affected (24.0 Bqkg^{-1} and 23.8 Bqkg^{-1}) compared to the background site (5.5 Bqkg^{-1} and 3.5 Bqkg^{-1}), but this relationship couldn't be reflected on TF distributions. Concerning grain size in this study, it could be observed that the percentage of silt + clay has been an important predictor of ^{238}U TF due to the significant and positive relation between them ($p < 0.01$) and in the case of ^{226}Ra TF it was the percentage of sand ($p < 0.001$), as mentioned before. This is in agreement with the study of Blanco Rodriguez *et al*. [10] who stated that the linearity assumption may be considered valid and that uranium soil-to-plant TF showed to be constant and independent of the substrate concentration if the fines and clays fraction (<0.067 mm) was used as the substrate instead of the bulk soil, and the same was valid in the case of radium when the coarse sand fraction (0.5-2 mm) played the role of substrate.

Regarding the radioisotope of lead, when all available parameters were included in the regression, ^{210}Pb TF was not found to be statistically related to its soil concentrations, but to the activity concentrations of its precursor in soil, ^{226}Ra and the parameters that influenced ^{226}Ra transfer to the leaves (pH and total soil Ca), including silt + clay percentage, tab. 3. The relation between ^{210}Pb TF and ^{226}Ra activity concentration in soil may be explained if atmospheric deposition is considered as the main pathway of ^{210}Pb uptake in the leaves since the fractions of radon ^{222}Rn (decay product of ^{226}Ra) emanated from soil are known to be dominant source of ^{210}Pb in the air [2, 28]. It is known that the radon emanation coefficient is proportional directly to the ^{226}Ra concentrations in soil and inversely to the soil grain size, and if grain size decreases (increasing the particles specific surface area) radon emanation rate increases [29]. This may explain higher (and above 1) soil-to-leaves ^{210}Pb TF values at the background site where investigated soil samples contained the highest percentages of clay fraction (<0.002 mm), from 22 % to 32 %. It was reported that the clay content influenced the Pb isotope transfer from soil into the native boreal plant species and actually enhanced the uptake of Pb, but this opposite effect was difficult to explain [17]. The radon flux into the atmosphere is a complex function of ^{226}Ra content, characteristics of soil (mineral grain size, porosity, water content) and meteorological conditions [29]. In addition, the different morphological characteristics of leaves such as leaf surface area, leaf structure (roughness or smoothness) and the trapping of aerosols in a different manner by the leaf surfaces could be reflected in ^{210}Pb activity variations in leaves [30] and its TF. A positive relationship between ^{210}Pb TF and silt + clay percentage may also be explained knowing that the number of blocked ^{222}Rn atoms in the pore spaces of soil is proportional to the percentage of finer particles of soil, as previously mentioned. It could be assumed that, apart from the atmospheric deposition of ^{210}Pb on the leaf surfaces, there is an important transport pathway within the tree that should be taken into account since it is recognised that plant roots take up radon gas from soils and that ^{222}Rn (and ^{226}Ra) can be extracted from soil via the transpiration stream of the trees [31].

Transfer factor values verification

All the obtained relationships presented in the tab. 3 implied that activity concentrations in tree leaves (C_{leaves}) are a non-linear function of activity concentrations in the soil (C_{soil}) of the type: $C_{\text{leaves}} = a\, C_{\text{soil}}^{\ b}$ with parameter values $a > 0$ and $1 < b < 1$. This is a regularly observed power function that reflects the non-linearity in the soil to plant transfer of the essential and non-essential elements [11, 13, 25, 27]. To extrapolate these obtained results, it was examined whether the concentration in leaves would be zero when the concentration in the soil is zero. This was correct ($C_{\text{soil}} = 0 \quad C_{\text{leaves}} = 0$) for all the relationships from the tab. 3, except the one for ^{238}U TF predicted by pH, soil Ca and silt + clay percentage which shouldn't be used for the soil concentrations lower then ~1 Bqkg^{-1}, meaning that more data are needed to improve the regression coefficient estimates. Regression results for ^{238}U, ^{226}Ra, and ^{210}Pb soil-to-leaves TF of the investigated broadleaf trees which explained more than 97 % of their variations are illustrated in fig. 1.

In this section, the soil-to-leaves TF values from the literature and those predicted in this study were compared. When using a number of predictor variables, regression results might reflect interrelationships of the included parameters among each other [20] which is why simpler models are probably more suitable. This also explains why ^{238}U and ^{226}Ra TF values were calculated in the first approximation using their activity concentration in the soil and the percentage of sand fraction of soil (the coefficients are given in tab. 3). It was noted that differences between reported and calculated values of about 2 % to 30 % were within the range of uncertainty of TF values determination.

Using values for coarse sand (73.8-82.2 %) from [23], there was an agreement between the TF values reported and those predicted in this study, respectively: 0.18 *vs.* 0.20 (for 81.86 Bqkg^{-1} ^{238}U$_{\text{soil}}$) and 0.97 *vs.* 1.00 (for 9.75 Bqkg^{-1} ^{238}U$_{\text{soil}}$) for mango leaves at two different sites and for guava leaves: 0.19 *vs.* 0.20 (for 86.79 Bqkg^{-1} ^{238}U$_{\text{soil}}$) and 0.19 *vs.* 0.12 (25.66 % of sand and 60.88 Bqkg^{-1} ^{238}U$_{\text{soil}}$). Similarly, ^{226}Ra TF values for the leaves of woody species, used as medicinal plants [32], grown in the soil with the mean value of 56.7 % fraction of sand were compared with the values predicted in this study: 0.25 *vs.* 0.31 (for 30.8 Bqkg^{-1} ^{226}Ra$_{\text{soil}}$) for *Fagus sylvatica L.*; 0.52 *vs.* 0.54 (for 17.3 Bqkg^{-1} ^{226}Ra$_{\text{soil}}$) for *Prunus spinosa L.* and 0.522 *vs.* 0.41 (for 23 Bqkg^{-1} ^{226}Ra$_{\text{soil}}$) for *Sambucus nigra L.* There was also an agreement for ^{226}Ra TF for shrubs *Atriplex halimus* (0.33 *vs.* 0.25) and *Atriplex Leucoclada Bioss* (0.27 *vs.* 0.25) at the background site (51.57 % of sand and 33 Bqkg^{-1} ^{226}Ra$_{\text{soil}}$) [33]. However, discrepancy (one order of magnitude difference) was found for the same species grown in the soil recently highly contaminated by ^{226}Ra (~15 10^3 Bqkg^{-1}) [33]. In the case of ^{210}Pb, TF values are calculated using only its activity concentrations in the soil (the coefficients are given in tab. 3). For the leaves of *Quercus robur* grown at the site affected by aluminium industry (using mean values 32.2 Bqkg^{-1} ^{210}Pb$_{\text{soil}}$ and 42.2 Bqkg^{-1} ^{210}Pb$_{\text{leaves}}$), consistence is found between ^{210}Pb TF value of 1.30 for oak leaves [26] and the value of 1.24 calculated in this study. Moderate agreement is found between the predicted and reported ^{210}Pb TF values for the leaves of woody species used as medicinal plants in India: 0.7 *vs.* 0.89 (for 46.1 Bqkg^{-1} ^{210}Pb$_{\text{soil}}$) for *Justica adhatoda* and 1 *vs.* 0.72 (for 58.5 Bqkg^{-1} ^{210}Pb$_{\text{soil}}$) for *Calycopteris floribunda* [30]. There was no consistency for ^{210}Pb TF for the leaves of eucalyptus (0.005 *vs.* 0.025 for 13800 Bqkg^{-1} ^{210}Pb$_{\text{UMT}}$) [6] and sycamore maple (0.008 *vs.* 0.023 for 7610 Bqkg^{-1} ^{210}Pb$_{\text{UMT}}$) [7] trees grown at uranium mill tailings (UMT). The results of comparison between TF values indicate that current calculations cannot be used for the substrates different from soil (such as tailings) and highly contaminated soil which significantly deviate from the radionuclides "steady state" between the soil and the trees.

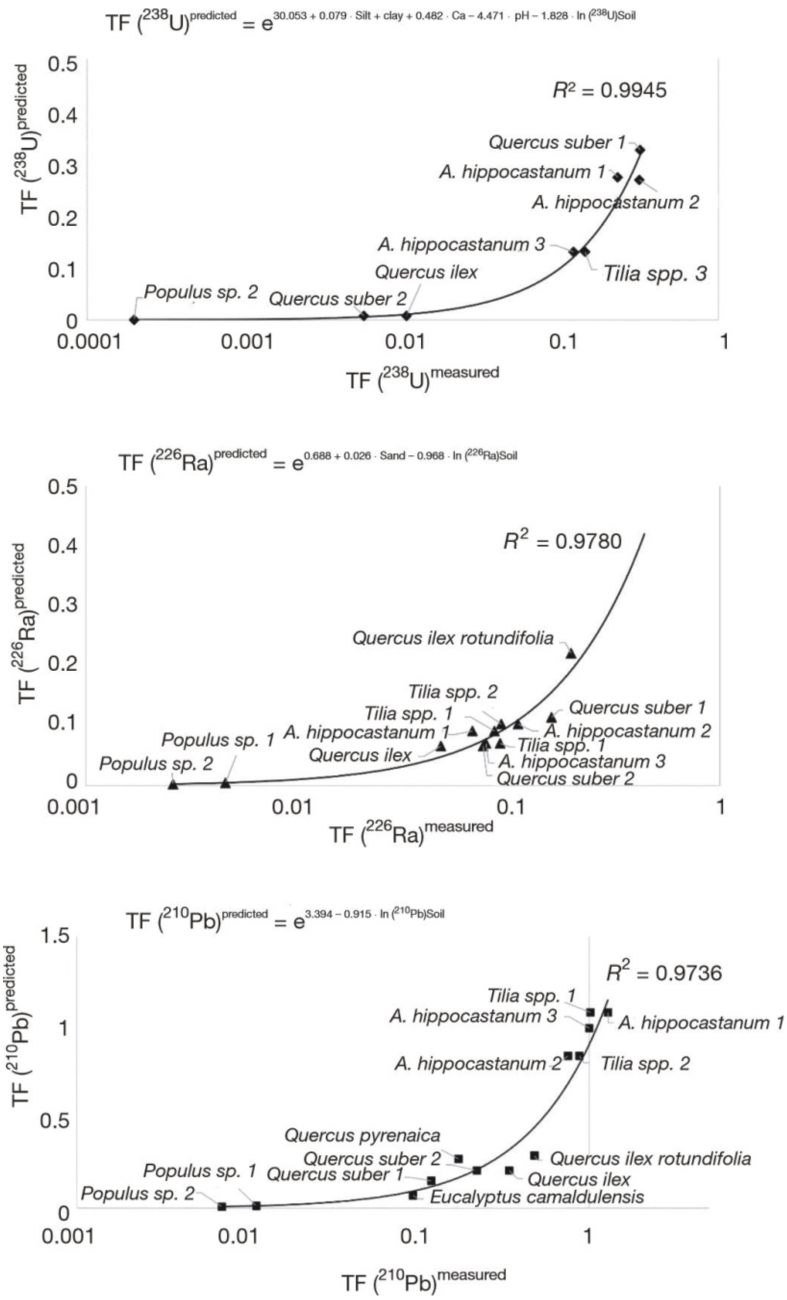

Figure 1. The predicted TF values (TF^{predicted}) *vs* measured TF values (TF^{measured}) for ^{238}U, ^{226}Ra, and ^{210}Pb from soil to the leaves of the examined broadleaf tree species

CONCLUSION

Soil-to-leaves TF values for ^{238}U, ^{226}Ra, and ^{210}Pb of different broadleaf tree species were collected from the literature from a few studies performed at sites affected by uranium presence and at the background site. The range of overall TF values was: 0.0002-0.4 for ^{238}U ($n = 10$), 0.0026-0.44 for ^{226}Ra ($n = 14$) and 0.007-1.283 for ^{210}Pb ($n = 14$). When significant differences were tested, TF of ^{238}U and ^{226}Ra, respectively, of 0.156 and 0.125 from affected sites couldn't be distinguished from that of 0.203 and 0.088 at the background site at the 95 % confidence level. Only mean ^{210}Pb TF of 0.931 was found to be significantly higher at the background site compared to the value of 0.180 at the affected site. There was a great similarity between uptake responses of the leaves on increasing activity concentration in the soil (from ~20 Bqkg^{-1} to ~8000 Bqkg^{-1}), so that biological differences between investigated broadleaf species could not be observed. The analysis indicated that ^{238}U, ^{226}Ra and ^{210}Pb activity concentrations and basic soil parameters (soil pH, total Ca, sand and silt + clay percentages) had the main impact on natural radionuclides TF values from soil to tree leaves. All the investigated broadleaf trees could be considered as an average species except *Eucalyptus camaldulensis* whose soil-to-leaves TF value was a statistically significant outlier ($p < 0.05$) for ^{226}Ra (0.44) and ^{238}U (0.4), but not in the case of ^{210}Pb (0.093). It was concluded that the expected TF values for average broadleaf tree could be calculated in the first approximation for ^{238}U and ^{226}Ra using their activity concentration in the soil and the percentage of sand fraction of soil, as well as in the case of ^{210}Pb using only its activity concentration in the soil.

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АНАЛИЗА ТРАНСФЕР ФАКТОРА ^{238}U, ^{226}Ra И ^{210}Pb ИЗ ЗЕМЉИШТА У ЛИШЋЕ ШИРОКОЛИСНИХ ВРСТА ДРВЕЋА

Анализа трансфер фактора ^{238}U, ^{226}Ra и ^{210}Pb из земљишта у лишће различитих врста широколисних стабала затечених, са једне стране, на локацијама претходно модификованим присуством уранијума и, са друге стране, на месту основног нивоа природног гама зрачења, урађена је коришћењем доступних података преузетих из неколико студија из литературе. Широколисне врсте дрвећа *Quercus ilex, Quercus suber, Eucalyptus camaldulensis, Quercus pyrenaica, Quercus ilex rotundifolia*, *Populus sp*. и *Eucalyptus botryoides Sm*. са модификованих локација и *Tilia spp*. и *Aesculus hippocastanum L*. на месту основног гама зрачења анализиране су заједно без обзира на листопадно или зимзелено порекло њихових листова. Подаци о основним параметрима земљишта, односно вредности pH, укупног садржаја Ca [gkg^{-1}], фракције песка [%] и суме фракција праха и глине [%] били су такође доступни у цитираним радовима. Прикупљени подаци о концентрацијама активности [Bqkg^{-1}] суве масе које су се кретале у земљишту у интервалу ($n = 14$): 22-6606 за ^{238}U, 38-7700 за ^{226}Ra и 37-7500 за ^{210}Pb и у узорцима лишћа у интревалу: 2.7-137.6 за ^{238}U ($n = 10$), 2.6-134.2 за ^{226}Ra ($n = 14$) и 27-77.2 за ^{210}Pb ($n = 14$) указивали су да прате нормалну расподелу након логаритамске трансформације. Студија је спроведена под хипотезом да биолошке разлике између различитих врста широколисног дрвећа имају мањи утицај на вредности трансфер фактора у поређењу са утицајем основних параметара земљишта и концентрација активности у земљишту, испитиваних природних радионуклида. Сходно томе, испитано је да ли се ^{238}U, ^{226}Ra и ^{210}Pb вредности трансфер фактора из земљишта у лишће просечне широколисне врсте могу предвидети статистички у првој апроксимацији на основу специфичне активности датог радионуклида у земљишту и најмање једног основног параметра земљишта коришћењем вишеструке линеарне регресије.

Кључне речи: природни радионуклид, трансфер фактор земљиште-лишће, упијање кореном, фолијарно упијање