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Distribution of natural radionuclides in anthrosol-type soil

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Abstract: Taking into account the importance of distribution and transfer of radionuclides in the soil-water-plant system, especially in agricultural fields, in this study, natural radionuclide determination in regosol-type soil was performed. The correlations between main soil properties and the contents of natural isotopes ^{238}U, ^{226}Ra, ^{232}Th, and ^{40}K in agricultural soil and their distribution through the soil depth was studied. For determination of natural radionuclide activity concentration, we used the CANBERRA HPGe detector, applying the gamma spectrometry method. The investigation was performed on an experimental peach tree field characterized by a anthrosol type of soil, derived from the chernozem type of natural soil. The effect of measured soil properties on activity concentration levels was analyzed by simple and multiple linear regression analysis. Considering the linear model, in our soil profiles of 0-80 cm depth, clay content was positively related with ^{238}U, ^{226}Ra, and ^{40}K, and about 30% of the variation of those radionuclides was explained. Sand fraction was negatively related with ^{238}U, ^{226}Ra, ^{232}Th, and ^{40}K and the correlation was medium, except for ^{226}Ra (r = -0.68). The joint effect of pH, carbonates, humus, clay, and sand contents on natural radionuclide distribution in the 0-80 cm layer was found by multiple linear regression analysis. Radionuclide behavior was explained by the R-squared statistic. The investigations of uranium distribution showed that when the soil layers of 0-20 cm and 60-80 cm are considered together, uranium behavior is affected by soil pH and the content of humus, carbonates, and clay. Multiple regression analysis of ^{226}Ra, ^{232}Th, and ^{40}K activities in soil layers of 0-20 cm and 60-80 cm showed that their lower mobility and uniform depth distribution are associated with small variations of carbonates along soil depth and mineral composition of the parent materials, while the soil pH had no effect on their behavior.

Key words: Distribution of natural radionuclides, anthrosol-type soil

Introduction

It is well known that the production of fertilizers from natural phosphate ore can lead to the redistribution of uranium and other radionuclides in the environment and to increased radiation exposure (Mortvedt 1994; IAEA 2003; Falck et al. 2006). Long-term application of phosphate fertilizers in agricultural fields could elevate natural radionuclide levels in the soil, and enrichment was observed in the surface soil layers (Mortvedt 1994; Takeda et al. 2006). The contribution to the uptake of radionuclides by plants grown in fertilized fields has also been established (Pulhani et al. 2005; Nasim-Akhtar et al. 2007). Although according to some authors the input

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of radionuclides of natural origin into the food chain via phosphate fertilizers appears to be of lesser concern because the radionuclide activity concentrations in most processed materials are only slightly higher than the levels in soil (Bolivar et al. 1995; Ioannides et al. 1997; Saueia et al. 2006), it has been concluded that knowledge about the distribution and transfer of radionuclides in the soil-plant-man system is necessary (IAEA 2003).

Naturally occurring radionuclides of terrestrial origin are members of the ^{238}U and ^{232}Th series, together with ^{40}K, and their concentrations in soil are related to the nature of the parent rock during soil genesis. Their natural radioactivity levels vary depending on the geological and geographical structure (UNSCEAR 2000; Belivermis et al. 2009). Artificial fertilizers applied to agricultural soil may change the natural level of terrestrial gamma radiation, since they are a product of phosphate rock containing an elevated level of natural radionuclides, especially ^{238}U (UNSCEAR 1988; IAEA 2003). Radionuclides in soils either occur in the soil mineral phase or are adsorbed onto soil components. Their behavior in soil is influenced by the mineral and organic inventory and elemental properties of the radionuclides, as well as biological fixation and transmutation (Koch-Steindl et al. 2001). It should be noted that the main soil properties have an important influence on the transport and evolution of radionuclides in soil (Golmakani et al. 2008). The major factor that affects geochemical reactions associated with radionuclides and explains their mobility in soil is the pH value (Echevarria et al. 2001; Koch-Steindl et al. 2001). Sorption and complexation on clay minerals, oxides, and organic matter are the main processes that have an impact on radionuclide mobility in soil (IAEA 2003). It was found that organic substances and noncrystalline clay minerals were responsible for accumulation of uranium derived from fertilizers (Takeda et al. 2006). Some studies have indicated that bioavailability of radionuclides is inversely related to soil sorption (Morton et al. 2002 and references therein) and radionuclides, which are soluble and thus present in soil solution, are most readily sorbed by plants (Mortvedt 1994; Golmakani et al. 2008). Taking into account the importance of distribution and transfer of radionuclides in the soil-water-plant system, especially in agricultural fields, the aim of this article is to investigate relations between main soil properties and the contents of natural isotopes ^{238}U, ^{226}Ra, ^{232}Th, and ^{40}K in agricultural soil and their distribution through the soil depth.

Materials and methods

For this study, a peach tree field in the experimental field "Radmilovac" (property of the Faculty of Agriculture, University of Belgrade), located near Belgrade, was selected. The covered area was 2 ha. The soil of this field is of the anthrosol type that was derived in 1992 from a natural chernozem type of soil by a special anthropogenic treatment that changed the natural structure and build of original soil. It was treated with fertilizers for about 10-12 years, when fertilization and other treatments like ploughing and irrigation of soil and trees were stopped; that was the moment of our sample collection. Taking into account that under normal radiological circumstances any part of studied soil area would be representative for the whole study area, the soil samples were collected at 4 randomly selected points from the Ap horizon depth of 80 cm with a step of 20 cm (upper layer of 0-20 cm, layers from the root zone of 20-40 cm and 40-60 cm, and layer below the root zone of 60-80 cm). Three tree soil profiles (P_1, P_2, P_3) were taken under the peach trees and a fourth (C_1) was taken from a soil area covered with grass within the same field.

In order to prepare 16 soil samples for measurement, the soil was air-dried and sieved through a 2 mm sieve and its physical and chemical properties were analyzed by standard methods: potentiometric method (in water and KCl) for soil pH, volumetric method by Scheibler's calcimeter for $CaCO_3$ content, and Tjurin's method modified by Simakov for the content of humus and mechanical composition were conducted by pipette method.

Soil samples were packed in 500 cm^3 Marinelli beakers, sealed, and left for 4 weeks to attain radioactive equilibrium between ^{226}Ra and ^{222}Rn and their progenies. For determination of natural radionuclide activity concentration, we used the CANBERRA HPGe detector, applying the gamma

spectrometry method. The activity of ^{238}U was determined by ^{234}Th (63 keV) or by ^{234}Pa (1000 keV). The activities of ^{226}Ra were determined by their decay products: ^{214}Bi (609, 1120, and 1764 keV) and ^{214}Pb (295 and 352 keV). The activities of ^{232}Th were determined by their decay product, ^{228}Ac (338 and 911 keV), and ^{40}K from its 1460 keV γ-line (Janković et al. 2008). The analytical precision of our measurements was approximately 10%.

Results

Soil characteristics

In order to investigate whether there were any relations along the soil profile depth between radionuclide activity concentration and main soil characteristics, several physical and chemical characteristics of the studied soil were observed. The results for the soil characteristics of the studied soil profiles (P_1, P_2, P_3, C_1) at each of the 20 cm depth intervals are presented in Table 1.

It was noticed that, belonging to the same anthrosol type, the soil profiles did not deviate much in their characteristics, except for the higher carbonate content in profile P_2. They had the same silty clay texture, with clay content variation from 33.08% to 43.33% and sand content variation from 2.18% to 16.12%. The soil profile P_1 (with the highest average radionuclide activity concentration) contained the highest amount of clay and the lowest sand percentage among all soil profiles. In profile P_2, there was a much higher carbonate content, 7%-10%, than in other profiles, where the content was less than 1%. This difference comes from the parent material that was mixed during deep tillage while preparing the soil for the planting of the peaches.

The variation in carbonates affected the pH level, as well; P_2 was alkaline and other soil profiles varied from weakly acidic to neutral. However, it should be noted that the variation of pH within the profiles was rather small (CV: 1%-3%). All profiles exhibited the same decreasing trend of humus percentages, and this characteristic varied the most of all soil characteristics (the humus percentages varied about 40% with the depth).

The leaching of the clay (particle sizes of less than 0.002 mm) out of the 20-60 cm layer and its accumulation in the deeper layers was observed. Since the particle size reduction was probably favored by the presence of abundant water in the root zone layer, the percentages of more coarse particles as sand (particle sizes greater than 0.05 mm) were higher in the root zone layers (20-60 cm).

Table 1. Physical and chemical characteristics of soil profiles P_1, P_2, P_3, and C_1.

Soil depth (cm)	pH_{KCl}	pH_{H2O}	Humus (%)	Carbonates (%)	Sand (%)	Silt (%)	Clay (%)
0-20	6.61	7.47	2.12	0.25	2.18	54.8	43.02
20-40	6.50	7.64	1.36	0.29	6.62	51.75	41.63
40-60	6.43	7.59	1.19	0.38	6.01	52.37	41.62
60-80	6.26	7.57	0.92	0.24	4.54	52.13	43.33
0-20	7.25	8.16	1.27	9.04	12.31	52.98	34.71
20-40	7.28	8.27	1.1	7.98	12.46	54.39	33.15
40-60	7.38	8.32	0.75	9.04	16.12	50.69	33.19
60-80	7.41	8.29	0.48	10.3	13.19	53.45	33.36
0-20	6.67	7.45	1.42	0.17	13.32	52.01	34.67
20-40	5.70	7.18	1.06	0.21	14.56	52.36	33.08
40-60	5.27	6.85	0.67	0.21	13.89	52.83	33.28
60-80	5.54	7.04	0.46	0.25	12.89	53.6	33.51
0-20	6.71	7.71	2.8	1.05	5.33	55.92	38.75
20-40	6.76	7.82	1.97	0.98	9.61	52.6	37.79
40-60	6.69	7.95	1.64	0.63	5.97	55.71	38.32
60-80	6.70	7.94	1.05	0.46	8.22	56.3	35.58

Radionuclide distribution

The distribution of natural radionuclides in the 20 cm depth intervals of the 4 soil profiles is presented in Table 2. It can be noticed that radioisotope activities (Bq kg^{-1}) lie in the range of 50-90 for ^{238}U, 39-59 for ^{226}Ra, 46-62 for ^{232}Th, and 565-755 for ^{40}K. The obtained values of activity concentration are in good agreement with the recommended values for background gamma radiation reported for soils worldwide (UNSCEAR 2000): 16-110 for ^{238}U, 17-60 for ^{226}Ra, 16-64 for ^{232}Th, and 140-850 for ^{40}K.

It should be mentioned that all radionuclides are inclined to accumulate in the upper (0-20 cm) and the lower soil layers. Average activity concentration of ^{238}U was the highest in P_1 (75.5 Bq kg^{-1}) and the lowest in P_2 (58 Bq kg^{-1}). As this is the area of homogeneous lithology, a high correlation among natural radionuclide activity concentration is expected (Navas et al. 2002a). The investigations showed that a high and significant ($P < 0.001$) correlation could be established at the confidence level of 95% between 3 radionuclides, ^{226}Ra, ^{232}Th, and ^{40}K, but not between ^{238}U and other natural radionuclides. The correlation values were: $r = 0.86$ between ^{226}Ra and ^{232}Th (R^2 = 73%), $r = 0.80$ for ^{226}Ra and ^{40}K (R^2 = 64%), and $r = 0.93$ for ^{232}Th and ^{40}K (R^2 = 86%).

Discussion

According to our analysis, the activity concentration variation of natural radionuclides with soil depth is different because ^{238}U varies more with depth (Figure 1). The coefficient of variation (CV) for uranium was 17%-24% with respect to ^{226}Ra, ^{232}Th, and ^{40}K, which exhibit more homogeneous depth distribution, and their variation (CV: about 10%) is comparable with the experimental uncertainty of activity determination.

Table 2. Depth distribution of natural radionuclides in the 20 cm intervals of the studied soil profiles P_1, P_2, P_3, and C_1.

	^{238}U (Bq kg^{-1})	^{235}U (Bq kg^{-1})	^{226}Ra (Bq kg^{-1})	^{232}Th (Bq kg^{-1})	^{40}K (Bq kg^{-1})
Profile 1 (P_1)					
0-20 cm	90 ± 18	4.0 ± 0.6	59 ± 6	58 ± 6	683 ± 68
20-40 cm	79 ± 24	4.5 ± 0.7	44 ± 4	49 ± 5	615 ± 62
40-60 cm	52 ± 12	3.5 ± 0.4	53 ± 6	56 ± 6	689 ± 79
60-80 cm	81 ± 24	4.6 ± 0.7	55 ± 7	62 ± 6	755 ± 76
Profile 2 (P_2)					
0-20 cm	68 ± 15	2.7 ± 0.3	40 ± 4	47 ± 5	565 ± 56
20-40 cm	50 ± 16	3.7 ± 0.6	40 ± 5	45 ± 5	579 ± 58
40-60 cm	66 ± 14	3.9 ± 0.5	39 ± 4	50 ± 5	614 ± 61
60-80 cm	49 ± 16	3.0 ± 0.4	43 ± 4	46 ± 5	571 ± 58
Profile 3 (P_3)					
0-20 cm	75 ± 20	3.4 ± 0.5	45 ± 5	50 ± 5	617 ± 62
20-40 cm	54 ± 19	3.5 ± 0.5	49 ± 5	50 ± 5	641 ± 64
40-60 cm	51 ± 10	3.3 ± 0.4	53 ± 6	62 ± 7	705 ± 71
60-80 cm	84 ± 17	3.4 ± 0.4	48 ± 10	52 ± 5	624 ± 62
Control 1 (C_1)					
0-20 cm	84 ± 17	3.6 ± 0.5	49 ± 5	56 ± 7	692 ± 69
20-40 cm	69 ± 21	3.5 ± 0.4	45 ± 5	52 ± 5	623 ± 62
40-60 cm	72 ± 22	3.5 ± 0.5	54 ± 6	55 ± 6	627 ± 63
60-80 cm	54 ± 13	3.6 ± 0.4	53 ± 5	60 ± 6	673 ± 67

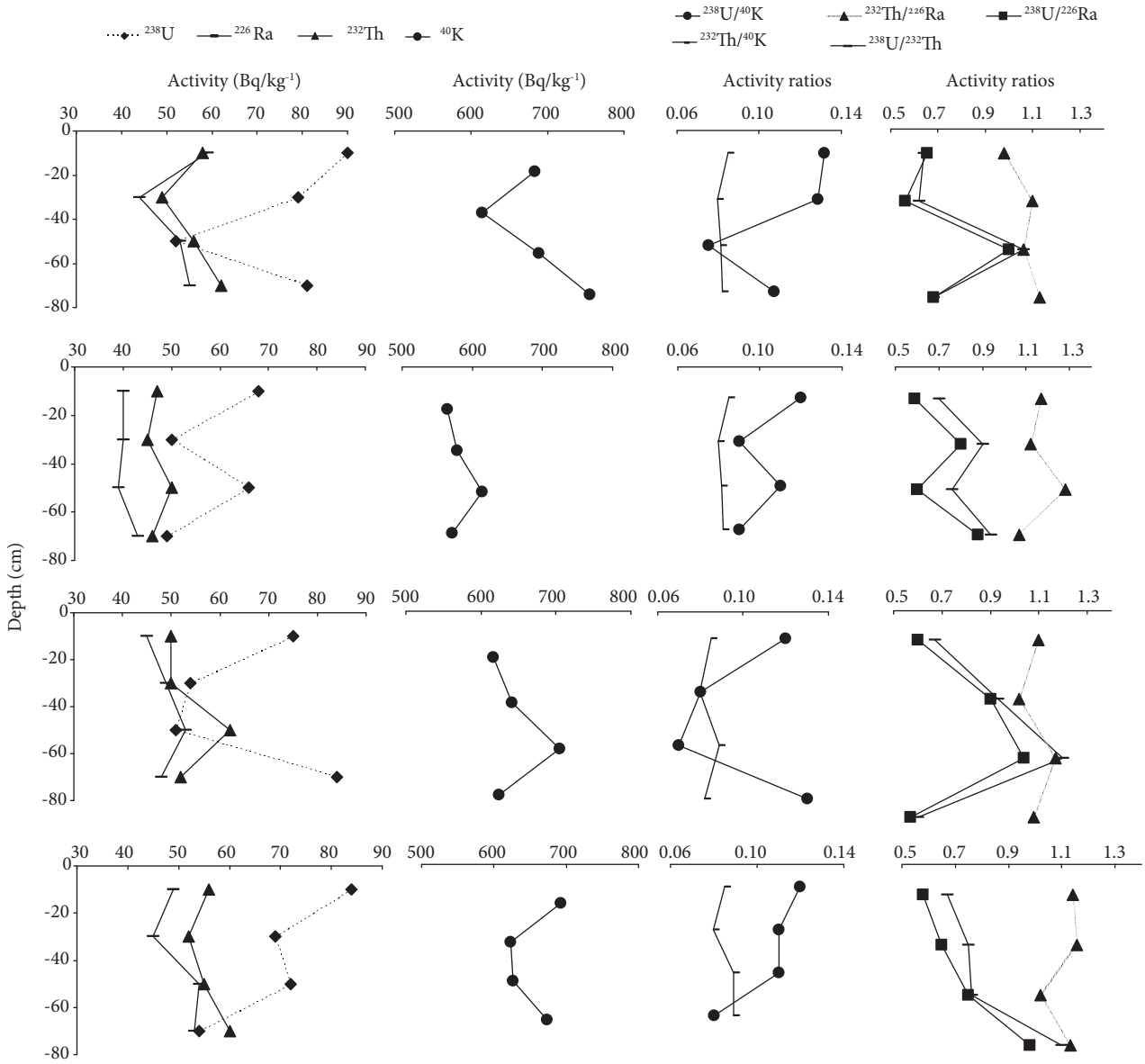

Figure 1. The activity concentration variation and activity ratios of natural radionuclides.

If parents of the ^{238}U and ^{232}Th decay series in soil are in secular equilibrium with their progenies, the activity ratios of parent to progeny, such as ^{238}U to ^{226}Ra, are equal to unity. According to Evans et al. (1997), in most environmental samples, the ratio of activities for members of the ^{238}U and ^{232}Th decay chain, like ^{232}Th to ^{238}U and ^{232}Th to ^{226}Ra, are typically equivalent to 1.1. Activity ratios for natural radionuclides ^{40}K and ^{232}Th could be calculated, and the ^{232}Th to ^{40}K ratio was estimated to be about 0.085, taking the median value of activity concentration for ^{40}K and ^{232}Th reported by UNSCEAR (2000).

In Figure 1, activity ratios are presented graphically. In every soil layer, in all profiles along the depth of 0-80 cm, ^{232}Th to ^{40}K ratios were approximately equal to 0.085, and activity ratios for ^{226}Ra to ^{232}Th were approximately equal to 1.1. Since high and significant correlation coefficients were found between ^{226}Ra, ^{232}Th, and ^{40}K, their constant activity ratios along the soil depth indicates that those

radionuclides originate from the same geochemical source (Navas et al. 2002b).

However, the activity ratios of ^{226}Ra to ^{238}U and ^{232}Th to ^{238}U values are less or equal to 1 or 1.1, respectively, in different soil layers. It should be noted that as a result of the accumulation of anthropogenically introduced uranium by use of phosphate fertilizers in this field (although in the short period of time of 10-12 years) (IAEA 2003), the ratios have been disturbed in favor of ^{238}U. The natural level of uranium ($U_{natural}$) in this soil could be calculated using the following equation (Takeda et al. 2006): $U_{natural} = 1.1 \times Th_{soil}$, where Th_{soil} is the observed activity concentration (Bq kg^{-1}) of ^{232}Th. According to this, the total uranium enhancement above the natural level in every soil layer with a step of 20 cm is estimated to be less than 0.5%. It has also been established that the accumulation of uranium in all soil profiles, P_1, P_2, P_3, and C_1, (Table 2) occurs especially in the upper 0-20 cm soil layer. This is in agreement with the results of other authors, who have found that the uranium level elevation by fertilization remains in the upper soil layer (Mortvedt 1994; Takeda et al. 2006).

Further investigations of soil profiles revealed the existence of proportionality of ^{226}Ra to ^{238}U, about 1, and ^{232}Th to ^{238}U, about 1.1, in the root zone layers (20-40 cm and 40-60 cm) in P_1, P_2, and P_3. This can be explained by the high mobility of the anthropogenic uranium, since in the migrative physico-chemical form, uranium can probably be partially absorbed by the root system of the peach trees and partially transferred to the deepest soil layer (60-80 cm) where it is accumulated. It should also be mentioned that the uranium remaining in the soil is fixed, coming from the geochemical source in a form less mobile and less available or totally unavailable for plant uptake (Stojanovic et al. 2006).

The effect of measured soil properties on activity concentration levels was analyzed by simple and multiple linear regression analysis. Linear models of the regression analysis between pH, humus, carbonate content, particle size, and radionuclide activities in the studied soil profiles with 0-80 cm depth are presented in Table 3. Marked correlations are significant at the $P < 0.05$ level unless otherwise noted.

Considering the linear model, in our soil profiles along 0-80 cm depth, clay content was positively related with ^{238}U, ^{226}Ra, and ^{40}K, and about 30% of the variation of those radionuclides was explained. Sand fraction was negatively related with ^{238}U, ^{226}Ra, ^{232}Th, and ^{40}K and the correlation was medium, except for ^{226}Ra ($r = -0.68$). Compared to ^{232}Th, which is associated only with sand fraction, the other natural radionuclides are associated with clay and sand fractions, as well. Since the sand fraction enhances the vertical mobility of radionuclides (Golmakani et al. 2008), the accumulation of radionuclides in the 60-80 cm layers was observed. At these depths, they are adsorbed onto clay surfaces or fixed within a lattice structure (Navas et al. 2002).

Humus content is correlated only with ^{238}U and explains 25% of its variability. The soil pH appears to be unrelated to the ^{238}U concentration along 0-80 cm depth, but it is inversely related to ^{226}Ra, ^{232}Th, and ^{40}K with similar medium correlations. Carbonates are not related to ^{238}U, but their percentages are inversely and significantly related to ^{226}Ra, ^{232}Th, and ^{40}K, describing more than 43% of their variation. It was noticed that carbonates varied little within the profiles, which is connected with the uniform depth distribution of ^{226}Ra, ^{232}Th, and ^{40}K. It can be concluded that within

Table 3. Correlations between clay, sand, humus, carbonates, and pH and ^{238}U, ^{226}Ra, ^{232}Th, and ^{40}K activity concentrations.

	^{238}U (Bq kg^{-1})	^{226}Ra (Bq kg^{-1})	^{232}Th (Bq kg^{-1})	^{40}K (Bq kg^{-1})
Clay (%)	0.5	0.58	-	0.57
Sand (%)	-0.51	-0.68 **	-0.53	-0.55
Humus (%)	0.54	-	-	-
Carbonates (%)	-	-0.74 ***	-0.66 **	-0.68 **
pH	-	-0.55	-0.5	-0.54

Significance level: *** $P < 0.001$, ** $P < 0.01$.

those 4 profiles, ^{226}Ra, ^{232}Th, and ^{40}K form a group of natural radionuclides with low mobility and similar constant depth distribution, but different from ^{238}U and its mobility.

The effect of the investigated soil properties on the natural radioactivity was also assessed through multiple linear regression analysis. The joint effect of pH, carbonates, humus, clay, and sand contents on natural radionuclide distribution in the 0-80 cm layer was determined. Radionuclide behavior was explained by the R-squared statistic. The model explains 38% of uranium behavior, 45% of ^{40}K, 51.5% of ^{232}Th, and 76.5% of ^{226}Ra at the $P < 0.05$ level. However, if the soil layers of 0-20 cm and 60-80 cm are considered together, the joint effect of soil properties explains a much higher percentage of radionuclide variation: 97% for ^{238}U, 95% for ^{226}Ra, 91% for ^{232}Th, and 86% for ^{40}K at the $P < 0.01$ level. In Table 4, the results of multiple linear regression analysis are presented.

The investigation of the natural radioactivity by multiple linear regressions in the soil layer of 20-60 cm, in which the root system zone exists, showed that no similar equation could be found. This result is in accordance with those of Hinsinger (2001), who found that plant roots can alter the chemical conditions in the soil solution in their immediate vicinity. Those changes in ionic and ligand concentrations and pH can have dramatic effects on the chemical reactions that occur in the soil solution and soil solid interface that determine the bioavailability of radionuclides to plants. Therefore, in the root zone, as a result of the biological activity of the root, to some extent all natural radionuclides and especially anthropogenically introduced uranium could take part in chemical interactions between plant roots' soil solution and soil solid constituents (Hinsinger 2001).

The investigations of uranium distribution showed that in our soil profiles, when the soil layers of 0-20 cm and 60-80 cm are considered together (Table 4), uranium behavior is affected by soil pH and the content of humus, carbonates, and clay. It is well known that, in soil, 80%-90% of uranium is present in a +VI oxidation state, and the chemical speciation of U(VI) is dependent on soil composition and on soil pH while complexation reactions rule its evolution in soil (Echevarria et al. 2001). According to our investigations, since there is an absence of large amounts of humus, the uranium is considered mobile. It occurs as a uranyl ion (UO_2^{2+}), which is the predominant uranium species in soil, and in neutral solutions it is transported as a $UO_2(CO_3)_2^{2-}$ complex (Mortvedt 1994). According to Navas et al. (2002a), as pH increases and humus content decreases down the soil profiles, uranium may also form organo-complexes and migrate downward in the soil profiles. Apart from uranium being partially absorbed by the plant roots in the so-called rhizosphere (soil layer of 20-60 cm), it seems that changed soil conditions favor the leaching of uranyl complexes to the deeper soil sections and their further binding to clay particles where they are more abundant. Moreover, the retention of uranium in the upper and the lowest layers is connected with sorption of bivalent uranyl ions to the negatively charged surfaces of clay minerals (Vandenhove et al. 2007).

Multiple regression analysis of ^{226}Ra, ^{232}Th, and ^{40}K activities in the soil layers 0-20 cm and 60-80 cm

Table 4. Coefficients in multiple regression equations for predicting ^{238}U, ^{226}Ra, ^{232}Th, and ^{40}K activities (Bq kg^{-1}) in the soil layers 0-20 cm and 60-80 cm, considered together with pH and percentages of clay, sand, humus, and carbonates.

	^{238}U (Bq kg^{-1})	^{226}Ra (Bq kg^{-1})	^{232}Th (Bq kg^{-1})	^{40}K (Bq kg^{-1})
Constant	326	86	69	282
Clay (%)	1.4	0.43	-	10.7
Sand (%)	-	1.7	1.1	-
Humus (%)	8	-	2.6	9.7
Carbonates (%)	2.1	0.58	0.64	7
pH	42	-	-	-

Significance level: ** $P < 0.01$.

showed that their lower mobility and uniform depth distribution are associated with small variations of carbonates along soil depth and mineral composition of the parent materials, while the soil pH had no effect on their behavior (Navas et al. 2002b). It has been noticed that radionuclide distribution is connected with particle size distribution, which is related to water infiltration in the 20-60 cm layer. It has been observed that ^{226}Ra and ^{232}Th are related via sand fraction, ^{226}Ra and ^{40}K via clay fraction of parent materials, and ^{232}Th and ^{40}K via humus fraction, so those radionuclides are moving through the soil column along with the soil particles with which they are connected.

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