# NATURAL ISOTOPES $^{238}$ U I $^{40}$ K CONTENT IN RIGOSOL FROM THE AREA OF SCHOOL ESTATE GOOD "RADMILOVAC" OF FACULTY OF AGRICULTURE, ZEMUN

Ivana Ž. Vukašinović <sup>1</sup>, Dragana J. Todorović <sup>2</sup>, A.R. Đorđević <sup>1</sup>, M.B. Rajković <sup>1</sup>, Mirjana D. Stojanović <sup>3</sup> and V.B. Pavlović <sup>1</sup>\*

Abstract: Distribution of natural gamma-emiting radionuclides <sup>238</sup>U and <sup>40</sup>K were determined in the soil profiles from the peach-trees field on experimental farm Radmilovac, southeast Belgrade. Internal soil morfolgy has been changed in 1992. when soil *rigosol* type with deep Ap-horizon (0-80cm) has been formed by special treatment of parent soil, *chernozem* type. Gamma-spectrometry method is applied in measurement of radionuclide activities in soil samples by using hyperpure coaxial gamma-ray detector, Canberra type. Investigation results has been shown that the natural activity contents obtained in the experiment are within the range of normal backround activity according to UNSCEAR (2000) and that radionuclide activity decreased in the plant root zone.

**Key words:** radionuclide, soil *rigosol* type, isotopes <sup>238</sup>U and <sup>40</sup>K, Ap-horizon.

-

<sup>\*1</sup> Ivana Ž. Vukašinović, Aleksandar R. Đorđević, Miloš B. Rajković, Vladimir B. Pavlović, Faculty of Agriculture, P.O.Box 14, 11081 Belgrade-Zemun, Nemanjina 6, Serbia <sup>2</sup> Dragana J. Todorović, Laboratory of Radiation and Environmental Protection, Institute of Nuclear Science "Vinča", Belgrade, Serbia

<sup>&</sup>lt;sup>3</sup> Mirjana D. Stojanović, Head of System Quality and Characterization Laboratory, Institute for Technology of Nuclear and Other Mineral Raw Materials (ITNMS), 11000 Belgrade, 86 Franchet d'Eperey, Serbia

#### Introduction

Natural radioactivity arises mainly from primordial radionuclides of terrestrial origin that are members of <sup>238</sup>U and <sup>232</sup>Th series together with <sup>40</sup>K and their concentrations and distribution in soil provides useful information in the monitoring of environmental radioactivity (Stojanović, 2000). Artificial fertilizers application on agricultural soil may change the natural level of terrestrial gamma-radiation since they are product of phosphate rock containing elevated level of natural radionuclides, especially <sup>238</sup>U (Stojanović et al., 2006; Mortvedt, 1994). Concentrations of <sup>40</sup>K in soils varies considerably from one country to another depending on the origin of fertilizers components and according to some outhors thay don't deviate much from natural levels found in soil (Bolivar et al., 1995; Saueia et al., 2006). In soils, radionuclides occur in minerals or are adsorbed onto soil components and main influence to their concentration and behavior in soil has soil parameters such as pH, organic matter, clay or carbonates contents (Navas et al., 2002).

The paper aims to investigate contents of natural isotopes <sup>238</sup>U and <sup>40</sup>K in agricultural soil and it's relation with main soil properties that affects their distribution with soil depth.

#### **Materials and Methods**

From the area of experimental farm "Radmilovac" property of Faculty of Agriculture (Southeast Belgrade), under the peach-trees field four soil profiles *rigosol* type has been taken. In 1992, *rigosol* type soil is derived from natural soil type *chernozem* by special anthropogenic treatment that changed natural structure and build of original soil. Peach-trees field has been treated with fertilizers about 10-12 years when fertilization stopped and tree years after that was the moment of collection of our samples (Rajković et al., 2008). Soil samples collected from Ap-horizon of 80 cm depth within soil layers 0-20 cm, 20-40 cm, 40-60 cm and 60-80 cm. Three soil profile (P<sub>1</sub>, P<sub>2</sub>, P<sub>3</sub>) were opened near the peach-tree root zone and fourth one (C<sub>1</sub>) from the soil area covered with grass but of the same field.

To prepare 16 soil samples for measurement, soil were air-dried and sieved through 2 mm sieve and their physical and chemical properties were analyzed by standard methods. Soil samples were packed in 500 cm<sup>3</sup> Marinelli beakers and kept sealed for 4 weeks to attain radioactive equilibrium. Gamma-spectrometry method is applied in order to determine

natural radionuclides activity concentration. The gamma-ray activities of soil samples were measured using p-type coaxial HPGe detector (Canberra type) computer controlled by conventional electronic with relative efficiency of 20 % end energy resolution of 1.8 keV for the 1332 keV gamma ray energy of <sup>60</sup>Co. A reference soil material (National Office of Measures OMH, Budapest) spiked with a series of radionuclides (<sup>22</sup>Na, <sup>57</sup>Co, <sup>60</sup>Co, <sup>89</sup>Y, <sup>133</sup>Ba and <sup>137</sup>Cs) with total activity of 1.5 kBq kg<sup>-1</sup> on 01.07.1991. were used for efficiency calibration. The geometry of the counting samples was the same as that of the standard sample and the counting time for all the samples was 70.000 s. The spectra were analyzed using the program GENIE 2000. The background spectrum was recorded immediately after or before the sample counting and subtracted from each sample spectrum. Activity of <sup>238</sup>U was determined by <sup>234</sup>Th (63 keV) or by <sup>234</sup>Pa (1000 keV) and activity of <sup>40</sup>K from its 1460 keV γ-line (Janković et al., 2008). Analytical precision of measurements was approximately 10 %.

#### **Results and Discussion**

The distribution of natural radionuclides in the 20 cm depth intervals, in four soil profiles ( $P_1$ ,  $P_2$ ,  $P_3$  and  $C_1$ ) is presented in Table 1 together with values of their main soil properties pH value, humus, carbonates, clay and send contents.

Soil profiles characteristics do not differ much from one another (Table 1) since profiles belong to the same soil type. Their common property is the silty clay texture. The clay content varied from 46.51 to 56.45 % and send content 2.18 to 16.12 %. Soil profile **P**<sub>1</sub> with highest average radionuclide activity concentration contained the highest clay and the lowest sand percentage among all soil profiles. In the profile **P**<sub>2</sub> there is much higher carbonate content 7-10 % referred to carbonate content in other profiles that is less than 1%. Variation in carbonates has affected on change of pH, **P**<sub>2</sub> is alkaline and in other profiles pH varied from weakly acidic to neutral but variation of pH within profiles is small, CV was 1-3 %. All profiles exhibit the same decreasing trend of humus percentages and it varied most of all other soil characteristics about 40 % with the depth. In the so-called rhizosphere zone (20-60 cm) there is a slightly lesser clay content while the sand content is slightly enhanced.

Radioisotope activities (Bq/kg) lie in the range of 52-90 for <sup>238</sup>U and 615-755 for <sup>40</sup>K. These values agree with recommended values for background gamma-radiation reported for soils worldwide (UNSCEAR,

2000) 16-110 for <sup>238</sup>U and 140-850 for <sup>40</sup>K, so radioactive impact of fertilization could be considered negligible.

Tab. 1. – Depth distribution of natural radionuclides (Bq/kg) and values of pH, carbonate, clay and sand contents in the 20 cm intervals in the studied soil profiles  $P_1$ ,  $P_2$ ,  $P_3$  and  $C_1$ 

	<sup>238</sup> U (Bq/kg)	<sup>40</sup> K (Bq/kg)	$ m p_{H_{20}}$	Humus (%)	Carbonates (%)	Sand (%)	Clay (%)	
Profile $l(\mathbf{P_1})$								
0-20 cm	90	683	7.47	2.12	0.25	2.18	43.02	
20-40 cm	79	615	7.64	1.36	0.29	6.62	41.63	
40-60 cm	52	689	7.59	1.19	0.38	6.01	41.62	
60-80 cm	81	755	7.57	0.92	0.24	4.54	43.33	
Profile 2 (P <sub>2</sub> )								
0-20 cm	68	565	8.16	1.27	9.04	12.31	34.71	
20-40 cm	50	579	8.27	1.10	7.98	12.46	33.15	
40-60 cm	66	614	8.32	0.75	9.04	16.12	33.19	
60-80 cm	49	571	8.29	0.48	10.3	13.19	33.36	
Profile 3 (P <sub>3</sub> )								
0-20 cm	75	617	7.45	1.42	0.17	13.32	34.67	
20-40 cm	54	641	7.18	1.06	0.21	14.56	33.08	
40-60 cm	51	705	6.85	0.67	0.21	13.89	33.28	
60-80 cm	84	624	7.04	0.46	0.25	12.89	33.51	
Control $l\left(\mathbf{C_{1}}\right)$								
0-20 cm	84	692	7.71	2.8	1.05	5.33	38.75	
20-40 cm	69	623	7.82	1.97	0.98	9.61	37.79	
40-60 cm	72	627	7.95	1.64	0.63	5.97	38.32	
60-80 cm	54	673	7.94	1.05	0.46	8.22	35.58	

Activity concentration (Bq/kg) variation of natural radionuclides  $^{238}$ U and  $^{40}$ K with 0-80 cm soil depth in the 20 cm intervals in the studied soil profiles  $P_1$ ,  $P_2$ ,  $P_3$  and  $C_1$  is presented in Figure 1.

From Figure 1, it could be seen that activity concentration variation of those two natural radionuclides with soil depth is different. Uranium incline to accumulate in the upper soil layer (0-20 cm) and  $^{40}$ K intend to accumulate in the lower layers.  $^{238}$ U varies more with depth, variation coefficient (CV) is 17-24 %, with respect to  $^{40}$ K that exhibit more homogeneous depth distribution, CV is 4-8 %.

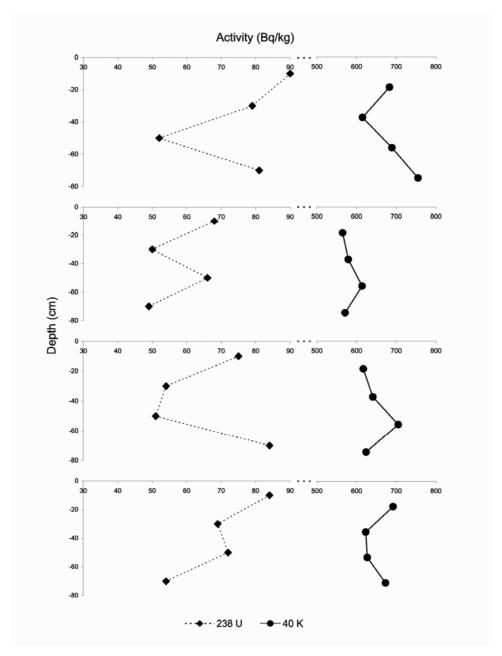


Fig. 1. – Activity concentration (Bq/kg) variation of natural radionuclides  $^{238}$ U and  $^{40}$ K with 0-80 cm soil depth in the 20 cm intervals in the studied soil profiles  $P_1$ ,  $P_2$ ,  $P_3$  and  $C_1$ 

Couse of higher uranium variation is enhanced mobility of anthropogenically introduced uranium (Potpora, et al., 2000; Vukašinovic et al., 2008). At pH values higher than 6.50, uranium is considered mobile and being in the migrative phisico-chemical form, it was partially transferred to the deeper soil layers and partially absorbed by the root system of the peach-trees (Stojanović et al., 2006). Differently from <sup>238</sup>U, low mobility and constant depth distribution of <sup>40</sup>K indicates that its content in soil is related to the mineral comosition of parent materials (Navas et al., 2002).

In the studied profiles, the effect of soil properties on levels of activity concentrations was analyzed through simple linear regression. 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 is presented in the Table 2. Marked correlations are significant at  $p \ge 0.05$  level unless otherwise mentioned.

Tab. 2. – Correlations between clay, physical clay, sand, humus, carbonates and pH and  $^{238}$ U and  $^{40}$ K activity concentrations

	<sup>238</sup> U (Bq/kg)	<sup>40</sup> K (Bq/kg)	R <sup>2</sup> (%)
Clay (%)	0.50	0.57	≈ 30 %
Sand (%)	- 0.51	- 0.55	$\approx 30 \%$
Humus (%)	0.54	-	$\approx 25 \%$
Carbonates (%)	-	- 0.68 **	$\approx 40~\%$
pH value	-	- 0.54	$\approx 30 \%$

Significance level: \*\*  $\geq 0.01$ 

Considering linear model, in our soil profiles along 0-80 cm depth, clay content is positively related with <sup>238</sup>U and <sup>40</sup>K and about 30 % variation of both radionuclides were explained. Sand fraction is negatively related with <sup>238</sup>U and <sup>40</sup>K with medium correlation. As sand fraction raises vertical mobility of radionuclides (Stojanović et al., 2006), in our soil profiles downward movement of radionuclides occurs in the 20-60 cm layer and it is

followed by accumulation to the lower soil layers where they are adsorbed onto clay surfaces or fixed within its lattice structure (Navas et al., 2002).

Humus content is correlated only with <sup>238</sup>U and explains 25 % of its variability. Soil properties that affects <sup>238</sup>U behaviour is suggesting that uranium may form complex ions such as stable uranyl carbonate and uranium organo-oxides complexes and migrate downword (Navas et al., 2002).

Carbonate percentage is inversely and significantly related with <sup>40</sup>K describing more than 40 % of its variation and indicated by high correlation coefficients seems to be important predictor for this radionuclide: small variation of carbonates within profiles is connected with uniform depth distribution of <sup>40</sup>K (Navas et al., 2002). The soil pH appears to be unrelated to <sup>238</sup>U concentration but it is inversely related with <sup>40</sup>K with medium correlation.

#### Conclusion

It can be said that within those four soil profiles, <sup>40</sup>K is natural radionuclide with low mobility and constant depth distribution which is different from <sup>238</sup>U enhanced mobility. Soil properties are differently affecting natural radionuclides mobility: accumulation of both radionuclides to the deeper soil layer is influenced by clay and sand content while carbonate content is the main predictor of restricted mobility of <sup>40</sup>K. Favored by the humus content in the root zone, anthropogenically introduced uranium being in the migrative phisico–chemical form was partially transferred to the deeper soil layers and partially absorbed by the root system of the peach-trees.

#### REFERENCES

- Mortvedt, J.J. (1994): Plant and Soil Relationships of Uranium and Thorium Decay Series Radionuclides - a review. Journal of Environmental Quality, 23, p. 643-650.
- 2. Bolivar, J. P., Garcia Tenorio, R. and Garcia-Leon, M. (1995): Fluxes and Distribution of Natural Radionuclides In The Production of Phosphate Fertilizers. Appl. Rad. Isot., 46, p. 717-718.
- 3. Navas, A., Soto, J. and Machin, J. (2002): <sup>238</sup>U, <sup>226</sup>Ra, <sup>210</sup>Pb, <sup>232</sup>Th and <sup>40</sup>K Activities In Soil Profiles of the Flysch Sector (Central Spanish Pyrenees). App.Radiat.Isot., 57, p. 579-589.

- Potpora, D., Stojanović, M., Ileš, D., Tešmanović, Lj. i Zildžović, S. (2000): Metode određivanja urana antropogenog i geohemijskog porekla, Međunarodna konferencija Otpadne vode, komunalni otpad i opasan otpad, Kopaonik, 23-26.05.2000.god., Zbornik radova, s. 531-535.
- 5. Rajković, M.B., Vukašinović, I., Đorđević, A.R., Todorović, D. and Pavlović, V. (2008): <sup>238</sup>U, <sup>226</sup>Ra, <sup>232</sup>Th and <sup>40</sup>K Distribution with Soil Depth in Agricultural Soil *Rigosol* Type and Its Relation with Main Soil Properties", Physical Chemistry 2008, *Proceedings* of the 9<sup>th</sup> International Conference on Fundamental and Applied Aspects of Physical Chemistry, A. Antić-Jovanović (Ed.), The Society of Physical Chemists of Serbia, September 24-26, 2008, Belgrade, Serbia, Biophysical Chemistry, Photochemistry, Radiation Chemistry (F), Volume I, F-23-P, p. 418-420.
- 6. Saueia, C.H.R. and Mazzilli, B.P. (2006): Distribution of Natural Radionuclides In The Production and Use of Phosphate Fertilizers In Brazil. Journal of Environmental Radioactivity, 89, p. 229-239.
- Stojanović, M. (2000): Utvrđivanje zavisnosti između sadržaja urana i fosfora u različitim zemljištima Srbije, Doktorska disertacija, Poljoprivredni fakultet, Zemun.
- 8. Stojanović, M., Babić, M., Stevanović, D., Martinović, Lj. (2006): Efekat višegodišnje primene fosfornih đubriva na kontaminaciju zemljišta Srbije. In: "Kontaminacija zemljišta Srbije radionukleidima i mogućnost njihove remedijacije", M. Stojanović (Ed.), Institute for Technology of Nuclear and Other Mineral Raw Materials (ITNMS), Belgrade: s. 67-115. (in Serbian)
- 9. United Nations Science Committee on the Effects of Atomic Radiation Sources (2000): UNSCEAR 2000 Report to the General Assembly with Scientific Annexes. New York: United Nations, p. 111-125.
- Vukasinovic, I., Todorovic, D., Đorđevic, A., Rajkovic, M., Stojanovic, M. and Pavlovic, V. (2008): Distribution of Radinuclides In Agricultural Soil Samples of Southeast Belgrade, Serbia. 2008 International Year of Planet Earth, Eurosoil 2008, Soil-Society- Environment, 25-29 August 2008, Vienna, Austria, Book of Abstracts, Winfried E. H. Blum, M. H. Gerzabek and M. Vodrazka (Eds.), Session S10: Advances in Soil monitoring, P527, p. 310.
- 11. Janković, M., Todorović, D., Savanović, M. (2008): Radioactivity measurements in soil samples collected in the Republic of Srpska. Rad. Measur., 43, p. 1448-52.

Received: March 06, 2009 Accepted: June 22, 2009

## SADRŽAJ PRIRODNIH IZOTOPA <sup>238</sup>U I <sup>40</sup>K U *RIGOSOLU* SA PODRUČJA OGLEDNOG ŠKOLSKOG DOBRA "RADMILOVAC", POLJOPRIVREDNOG FAKULTETA U ZEMUNU

### Ivana Ž. Vukašinović <sup>1</sup>, Dragana J. Todorović <sup>2</sup>, A.R. Đorđević <sup>1</sup>, M.B. Rajković <sup>1</sup>, Mirjana D. Stojanovic <sup>3</sup> i V.B. Pavlović <sup>1</sup>\*

#### Rezime

Distribucija prirodnih radionuklida gama-emitera <sup>238</sup>U i <sup>40</sup>K je određena u profilima zemljišta sakupljenih sa voćnjaka pod zasadom breskvi na Oglednom školskom poljoprivrednom dobru "Radmilovac", Poljoprivrednog fakulteta, Beograd-Zemun.

Unutrašnja morfologija zemljišta je izmenjena 1992. godine kada je zemljište *rigosol* tipa sa dubokim Ap-horizontom (0-80 cm) formirano rigolovanjem osnovnog zemljišta *černozem* tipa.

Aktivnost radionuklida u uzorcima zemljišta određena je metodom gama-spektrometrije korišćenjem koaksijalnog HPGe-detektora (Canberra).

Rezultati ispitivanja su pokazala da je eksperimentalno određena prirodna aktivnost u granicama normalne osnovne aktivnosti preporučene od strane UNSCEAR (2000) i da aktivnost radionuklida opada u zoni korenovog sistema.

Primljeno: 06 mart 2009 Odobreno: 22 jun 2009

\_

<sup>\* &</sup>lt;sup>1</sup> Ivana Ž. Vukašinović, <u>ivvu@EUnet.rs</u>, Aleksandar R. Đorđević, Miloš B. Rajković, Vladimir B. Pavlović, Poljoprivredni fakultet, P.O.Box 14, 11081 Beograd-Zemun, Nemanjina 6

<sup>&</sup>lt;sup>2</sup> Dragana J. Todorović, Laboratorija za zaštitu od zračenja i zaštitu životne sredine, Naučni institut "Vinča", Beograd

<sup>&</sup>lt;sup>3</sup> Mirjana D. Stojanović, Institut za tehnologiju nuklearnih i drugih mineralnih sirovina (ITNMS), 11000 Beograd, 86 Franše d'Esperea 86, Srbija.