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²³⁸U, ²²⁶Ra, ²³²Th AND ⁴⁰K DISTRIBUTION WITH SOIL DEPTH IN AGRICULTURAL SOIL *RIGOSOL* TYPE AND ITS RELATION WITH MAIN SOIL PROPERTIES

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Abstract

Distributions of natural gamma emitting radionuclides ²³⁸U, ²²⁶Ra, ²³²Th and ⁴⁰K were determined in two soil profiles *rigosol* type under the peach-tree field: one from and second out of the peach-tree root zone. Radioisotope activities (Bq/kg) lie in the range of normal terrestrial gamma-radiation in soil [1]. It is found that variation in natural radionuclides activities along a soil depth of 0-80 cm is due to differences in main soil properties: pH-reaction, carbonates, humus, clay and sand contents confirmed by high correlation between them.

Introduction

Naturally occurring radionuclides of terrestrial origin are members of ²³⁸U and ²³²Th series together with ⁴⁰K and their concentrations in soil are related to the nature of parent rock during soil genesis. 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 ²³⁸U [2,3]. 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 [2,4]. The paper aims to investigate contents of natural isotopes ²³⁸U, ²²⁶Ra, ²³²Th and ⁴⁰K in agricultural soil and its relation with main soil properties that affects their distribution with soil depth.

Materials and Methods

From the area of school estate good "Radmilovac" of Faculty of Agriculture, under the peach-trees field two 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. 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. First soil profile (**P1**) were taken from the peach-tree root zone and second one (**C1**) from the soil area covered with grass but of the same, previously fertilized, field.

To prepare 8 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 cm³ Marinelli beakers and kept sealed for 4 weeks to attain radioactive equilibrium. Gamma-spectrometry method applied using HPGe detector (Canberra type) in order to determine natural radionuclides activity concentration. Activity of ²³⁸U was determined by ²³⁴Th (63 keV) or by ²³⁴Pa (1000 keV). The activities of ²²⁶Ra and ²³²Th were determined by their decay products: ²¹⁴Bi (609 keV), ²¹⁴Pb (352 keV) and ²²⁸Ac (911 keV), respectively and ⁴⁰K from its 1460 keV γ -line. Analytical precision of measurements was approximately 10%.

Results and Discussion

The distribution of natural radionuclides in the 20 cm depth intervals, in two soil profiles (**P1** and **C1**) is presented in Table 1. together with values of their main soil properties pH (KCl) reaction, humus, carbonates, clay and sand contents.

Table 1. Depth distribution of natural radionuclides and values of pH, humus, carbonate, clay and sand contents in the studied profiles **P1** and **C1**

| | ²³⁸ U (Bq/kg) | ²²⁶ Ra (Bq/kg) | ²³² Th (Bq/kg) | ⁴⁰ K (Bq/kg) | pH | Humus (%) | CaCO ₃ (%) | Clay (%) | Sand (%) |
|-----------------------|-----------------------------|------------------------------|------------------------------|----------------------------|------|--------------|--------------------------|-------------|-------------|
| <i>Profile 1 (P1)</i> | | | | | | | | | |
| 0-20 cm | 90 | 59 | 58 | 683 | 6.61 | 2.12 | 0.25 | 43.02 | 2.18 |
| 20-40 cm | 79 | 44 | 49 | 615 | 6.50 | 1.36 | 0.29 | 41.63 | 6.62 |
| 40-60 cm | 52 | 53 | 56 | 689 | 6.43 | 1.19 | 0.38 | 41.62 | 6.01 |
| 60-80 cm | 81 | 55 | 62 | 755 | 6.26 | 0.92 | 0.24 | 43.33 | 4.54 |
| <i>Control 1 (C1)</i> | | | | | | | | | |
| 0-20 cm | 84 | 49 | 56 | 692 | 6.71 | 2.80 | 1.05 | 38.75 | 5.33 |
| 20-40 cm | 69 | 45 | 52 | 623 | 6.76 | 1.97 | 0.98 | 37.79 | 9.61 |
| 40-60 cm | 72 | 54 | 55 | 627 | 6.69 | 1.64 | 0.63 | 38.32 | 5.97 |
| 60-80 cm | 54 | 53 | 60 | 673 | 6.70 | 1.05 | 0.46 | 35.48 | 8.22 |

Radioisotope activities (Bq/kg) lie in the range of 52-90 for ²³⁸U, 44-59 for ²²⁶Ra, 49-62 for ²³²Th and 615-755 for ⁴⁰K. These values agree with recommended values for background gamma-radiation reported for soils worldwide [1]: 16-110 for ²³⁸U, 17-60 for ²²⁶Ra, 16-64 for ²³²Th and 140-850 for ⁴⁰K. Total uranium enhancement above natural level is estimated to be less than 0,5%, amount so small that radioactive impact of fertilizer is considered negligible [5].

Activity concentration variation of ²³⁸U with soil depth is higher (18-22 %) with respect to ²²⁶Ra, ²³²Th and ⁴⁰K that exhibit more homogeneous depth distribution. Their variation with depth (approximately 10%) is comparable with experimental uncertainty of activity determination. All radionuclides incline to accumulate in the upper (0-20 cm) and the deepest layer (60-80 cm), except ²³⁸U in **C1**. In order to investigate is there any relation along a soil profile depth between radionuclides activity concentration and main soil properties, linear correlation coefficients were found.

For ^{238}U , in **P1**, highest correlation found with carbonates contents ($r = -0.95$). Since there is absence of large amounts of humus uranium is considered mobile, occurring as a uranyl ion (UO_2^{2+}) and in neutral solutions transported as a $\text{UO}_2(\text{CO}_3)_2^{2-}$ complex [3]. In **P1**, it becomes available for uptake in the plant root zone (40-60 cm). In **C1**, out of the root zone, uranium is decreasing with depth gradually where high correlation with clay is found ($r = 0.95$). Natural radioisotopes are adsorbed onto clay surface or fixed within the lattice structure [2,4] that is connected with reduced downward movement of ^{238}U in **C1**.

Group of ^{226}Ra , ^{232}Th and ^{40}K in **P1** exhibits similar behavior as their main correlations are with clay ($r = 0.73$, $r = 0.83$ and $r = 0.74$ for ^{226}Ra , ^{232}Th and ^{40}K respectively) and sand ($r = 0.87$, $r = 0.61$ and $r = 0.40$). In general, increased sand content in soil enables radioisotopes to move along soil layers [2]. In **P1**, as sand content is low, ^{226}Ra , ^{232}Th and ^{40}K stayed fixed within clay minerals and accumulated in the upper or the deepest soil layer being in the form unavailable for plant root uptake. In the **C1** profile, behavior of ^{226}Ra and ^{232}Th is connected with pH, that is more constant down the **C1**. However, for ^{40}K in **C1** important correlation with soil properties is not found.

Conclusion

Soil properties differently affects ^{238}U , ^{226}Ra , ^{232}Th and ^{40}K radionuclides mobility: accumulation of all radionuclides to the deepest soil layer and restricted mobility of ^{226}Ra , ^{232}Th and ^{40}K in soil is influenced by clay and sand content while carbonate content is the main predictor for ^{238}U mobility. In the root zone, anthropogenically introduced uranium, being in the migrative physico-chemical form was partially transferred to the deeper soil layers and partially absorbed by the root system of the peach-trees. Low mobility and constant depth distribution of ^{226}Ra , ^{232}Th and ^{40}K indicates that their distribution in soil is related to the mineral composition of parent materials.

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