ISSN: 2181-3337 | SCIENTISTS.UZ VOLUME 5 ISSUE 9 | SEPTEMBER 2026

ASSESSMENT OF RADIONUCLIDE PARAMETERS IN SOILS OF RECLAIMED AREAS BASED ON A MATHEMATICAL MODEL

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DOI: 10.67620/sai.59a0008

ABSTRACT

This article investigates the depth-dependent distribution of uranium and radium radionuclides in soils of reclaimed areas. The main objective of the study is to determine changes in the specific activities of ^{238}U and ^{226}Ra in soil layers and to evaluate differences in radionuclide distribution on the basis of the $^{226}Ra/^{238}U$ radioactive-equilibrium ratio. Data obtained at depths of 0.25, 0.50, 0.75, and 1.00 m were analyzed for each of the P1 and P2 sampling points. According to the results, the ^{238}U activity decreased from 1844.8 to 408 Bq/kg at P1 and from 3160 to 610 Bq/kg at P2. In contrast, the ^{226}Ra activity increased from 521 to 1400 Bq/kg at P1 and from 512 to 3240 Bq/kg at P2. The $^{226}Ra/^{238}U$ ratio changed from 0.28 to 3.43 at P1 and from 0.16 to 5.31 at P2. To interpret the observed vertical changes, the possible effects of soil-water interaction, vertical water infiltration, sorption-desorption processes, and geochemical conditions are discussed. The results may provide a basis for assessing the vertical distribution of radionuclides in reclaimed areas and for subsequent mathematical modeling of migration processes.

ACCEPTED

05.10.26

Keywords: *reclamation, soil, ^{238}U, ^{226}Ra, specific activity, radionuclides, depth distribution, $^{226}Ra/^{238}U$ ratio, water infiltration, radionuclide migration, sorption, desorption.*

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Introduction

The distribution and redistribution of radionuclides in environmental components within technogenically altered areas formed during uranium mining and processing are important scientific issues from the standpoint of environmental safety. In particular, studying the vertical distribution of uranium-series radionuclides, including ^{238}U and ^{226}Ra, in the soils of reclaimed areas makes it possible to evaluate their movement along the soil profile. The distribution of radionuclides in soil may be affected by their physicochemical properties, the mineralogical composition and moisture content of the soil, the degree of binding to particles, sorption and desorption processes, as well as vertical water infiltration. Therefore, it is important not to restrict the assessment to radionuclide contents in the surface layer alone, but also to analyze their specific activities at different depths. The ratio between ^{238}U and ^{226}Ra can be used as an additional indicator for analyzing the state of radioactive equilibrium between radionuclides and redistribution processes within the soil profile. Under conditions close to natural radioactive equilibrium, uranium and radium activities are expected to be similar; therefore, a pronounced depth-dependent change

in their ratio may indicate differences in radionuclide mobility or redistribution processes in the soil environment.

Foreign studies have also examined the vertical distribution of ^{238}U and ^{226}Ra in soils of uranium-mining areas. For example, Jiang et al. analyzed the contents of ^{238}U and ^{226}Ra in soil profiles to a depth of 0-1.0 m around a uranium-mining area and evaluated both their depth-dependent variation and the $^{226}Ra/^{238}U$ ratio.

In the present study, the specific activities of ^{238}U and ^{226}Ra at different depths in soils of a reclaimed area, together with their $^{226}Ra/^{238}U$ ratio, are analyzed. The main emphasis is placed on identifying the characteristics of vertical radionuclide distribution, evaluating the depth-dependent variation of their ratio, and discussing the observed changes in terms of water infiltration, sorption-desorption, and geochemical processes in soil.

Literature review

Issues related to the distribution and migration of radionuclides in soil and the assessment of their environmental risk in uranium-mining and processing areas have been investigated in a number of foreign studies. These studies have mainly focused on the distribution of ^{238}U and ^{226}Ra along soil profiles, the state of radioactive equilibrium between them, and radionuclide redistribution processes in the environment.

Jiang W., Gao B., Zhang H.Y. et al. investigated the distribution characteristics of ^{238}U and ^{226}Ra in soils surrounding a uranium-mining area. Soil samples were collected from the 0-0.2, 0.2-0.4, 0.4-0.6, 0.6-0.8, and 0.8-1.0 m layers. In addition to statistical analysis of vertical radionuclide distribution, the authors used the $^{226}Ra/^{238}U$ ratio to assess the relationship between radionuclides within the soil profile. The study identified changes in the contents of ^{238}U and ^{226}Ra with soil depth.

The $^{226}Ra/^{238}U$ ratio was also considered an important indicator in that study. According to the authors, under radioactive-equilibrium conditions this ratio may be approximately equal to one. The reported values were 0.82-1.11 in the 0.4-0.6 m layer and 0.60-1.15 in the 0.8-1.0 m layer. Overall, the depth-dependent variation of the $^{226}Ra/^{238}U$ ratio was used to characterize radionuclide redistribution within the soil profile.

The vertical distribution of radionuclides in uranium-bearing areas has also been investigated in other studies. In particular, the distribution of natural radionuclides and deviations from radioactive equilibrium in the uranium series have been examined in soil profiles from uranium-mineralized areas of Spain. Such studies show that the state of radionuclides in soil can be assessed not only from their total concentrations, but also from their mutual ratios and depth-dependent changes.

Water movement may also influence the redistribution of radionuclides among soil layers in uranium tailings and technogenically altered areas. Water infiltrating into soil may transport radionuclides vertically in dissolved form or alter their migration rate as a result of sorption onto mineral particles. From this perspective, sorption-desorption processes and water infiltration are regarded as important factors when studying radionuclide movement in the soil-water system.

At the same time, the literature emphasizes that the use of specific-activity values alone in assessing vertical radionuclide migration may not fully reveal all migration mechanisms. Radionuclide mobility can be affected by the mineralogical composition and moisture content of soil, pH, carbonate and sulfate compounds, iron and manganese oxides, organic matter, and sorption properties. Therefore, hydrological and geochemical data should also be taken into account when relating the depth distribution of ^{238}U and ^{226}Ra to water-driven migration.

The reviewed literature shows that existing studies have mainly examined radionuclide concentrations in soil, their vertical distribution, the $^{226}Ra/^{238}U$ ratio, and environmental risk. However, linking depth-dependent changes in ^{238}U and ^{226}Ra in reclaimed areas with vertical water infiltration, sorption-desorption processes, and possible radionuclide migration, and subsequently using these data for mathematical modeling, requires separate investigation. From this standpoint, it is appropriate to analyze

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the ^{238}U and ^{226}Ra data obtained at different depths at P_1 and P_2, determine their $^{226}Ra/^{238}U$ ratios, and evaluate the observed vertical changes in relation to possible migration processes in the soil-water environment.

Materials and methods

To assess the depth-dependent distribution of ^{238}U and ^{226}Ra in soils of the reclaimed area, data obtained at the P_1 and P_2 study points were used. For each point, uranium and radium activity values at different depths and their ratio were considered. The available experimental dataset includes depths of 0.25, 0.50, 0.75, and 1.00 m for P_1 and P_2. The main study parameters were the specific activities of ^{238}U and ^{226}Ra in soil. At P1, the ^{238}U values with depth were 1844.8, 1382.8, 948, and 408 Bq/kg, whereas the corresponding ^{226}Ra values were 521, 831, 1100, and 1400 Bq/kg. At P_2, the values were 3160, 2640, 1210, and 610 Bq/kg for ^{238}U and 512, 1340, 2360, and 3240 Bq/kg for ^{226}Ra.

Assessment of radionuclide distribution with depth

To determine changes in ^{238}U and ^{226}Ra along the soil profile, the specific-activity values at each depth were compared. In analyzing the vertical distribution, the following relative change can be used:

$$\Delta C = (C_z - C_0) / C_0 \times 100\% \quad (1)$$

where C0 is the specific activity of the radionuclide in the initial or upper layer, Bq/kg; Cz is the specific activity of the radionuclide at depth z, Bq/kg; and ΔC is the relative change in radionuclide activity, %.

This indicator makes it possible to quantitatively evaluate the degree to which radionuclide activity decreases or increases with increasing depth.

Calculation of the $^{226}Ra/^{238}U$ activity ratio

To further evaluate the characteristics of vertical radionuclide distribution, the ratio of the specific activities of ^{226}Ra and ^{238}U was calculated:

$$K_{Ra/U} = A(^{226}Ra) / A(^{238}U) \quad (2)$$

where $A(^{226}Ra)$ is the specific activity of ^{226}Ra, Bq/kg; $A(^{238}U)$ is the specific activity of ^{238}U, Bq/kg; and $K_{Ra/U}$ is the $^{226}Ra/^{238}U$ activity ratio.

This indicator was used to assess the relative distribution of the radionuclides. In the study data, the ratio was 0.28, 0.60, 1.16, and 3.43 at P1, and 0.16, 0.51, 1.95, and 5.31 at P2.

To evaluate the relative distribution of radionuclides, it is necessary to determine their balance between soil layers. The balance of radionuclides between soil layers is determined as follows:

$$dM_{i,j}/dt = Q_{i,j}C_{i,j-1} - Q_jC_{i,j} + A_jD_{i,j} - A_{j-1}D_{i,j-1} + \lambda_{i-1}M_{i-1,j} - \lambda_iM_{i,j} + S_{i,j} \quad (3)$$

where $M_{i,j}$ is the mass of the soil layer; i = 1, …, N is the layer number; $C_{i,j}$ is the substance concentration in the i,j-th layer; $D_{i,j-1}$ is the diffusion coefficient in the layer; A_j, A_{j-1}, and A_{j+1} are the specific activities of radionuclides in the j, j-1, and j+1 layers; λ_i and λ_{i-1} refer to the radionuclide decay parameters in the i and i-1 layers as stated in the source; and $S_{i,j}$ denotes the effective transfer coefficients between boundary layers.

Assessment of possible migration processes in the soil-water system

When interpreting changes in radionuclides along the soil profile, vertical water infiltration was considered as a possible migration factor. As water moves through soil pores, radionuclides may be transported in dissolved form or become bound to soil particles.

To characterize radionuclide partitioning between the soil and water phases, the distribution coefficient K_d is expressed as:

$$K_d = C_s / C_w \quad (4)$$

where Cs is the concentration of radionuclide sorbed in soil, and Cw is the radionuclide concentration in water.

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However, in the present study, Kd, soil bulk density, porosity, water filtration rate, and hydrochemical parameters were not measured experimentally. Therefore, water infiltration was considered only as a possible mechanism for explaining the observed vertical distribution of ^{238}U and ^{226}Ra.

Statistical analysis of the data

The obtained data were arranged according to the depth coordinate, and P1 and P2 were analyzed separately. For each point, the depth-dependent changes in ^{238}U, ^{226}Ra, and KRa/U were determined. The results were presented graphically, and the vertical radionuclide profiles were compared.

The general study algorithm was implemented in the following sequence: soil sampling → determination of ^{238}U, ^{226}Ra, and KRa/U values by depth → construction of vertical-distribution plots → consideration of migration mechanisms → discussion of the obtained results.

Based on the experimental data, the characteristics of vertical radionuclide distribution were identified, providing a methodological basis for the subsequent investigation of these processes using mathematical models related to water infiltration and radionuclide migration.

Results and discussion

In analyzing the study results, the specific activities of ^{238}U and ^{226}Ra measured at different depths at P1 and P2, together with the radioactive-equilibrium coefficient between ^{226}Ra and ^{238}U, were taken as the main indicators. According to the available data, differences were observed between P1 and P2 in the depth distribution of radionuclides. At P1, the specific activity of ^{238}U decreased consistently with increasing depth: 1844.8 Bq/kg at 0.25 m, 1382.8 Bq/kg at 0.50 m, 948 Bq/kg at 0.75 m, and 408 Bq/kg at 1.00 m. Thus, there was a substantial difference between the uppermost and deepest layer values. A similar vertical decrease was observed at P2: the specific activity of ^{238}U was 3160 Bq/kg at 0.25 m, 2640 Bq/kg at 0.50 m, 1210 Bq/kg at 0.75 m, and 610 Bq/kg at 1.00 m.

These results indicate a tendency for ^{238}U to decrease from the surface toward the deeper soil layers at both P_1 and P_2. At the same time, the ^{238}U values at P_2 were higher than those at P_1 at all analyzed depths.

In the case of the vertical distribution of ^{226}Ra, an opposite trend relative to ^{238}U was observed. At P1, the specific activity of ^{226}Ra increased from 521 Bq/kg at 0.25 m to 831 Bq/kg at 0.50 m, 1100 Bq/kg at 0.75 m, and 1400 Bq/kg at 1.00 m. At P2, this difference was even more pronounced: the specific activity of ^{226}Ra was 512 Bq/kg at 0.25 m, 1340 Bq/kg at 0.50 m, 2360 Bq/kg at 0.75 m, and 3240 Bq/kg at 1.00 m.

Thus, the analyzed data show that the activity of ^{238}U decreases with depth, whereas the activity of ^{226}Ra increases. This indicates that the two radionuclides are not distributed in the same manner across the soil layers.

The radioactive-equilibrium coefficient $K^{226}Ra/^{238}U$ changed as follows: at P1, the values at depths of 0.25, 0.50, 0.75, and 1.00 m were 0.28, 0.60, 1.16, and 3.43, respectively. At P_2, the corresponding values were 0.16, 0.51, 1.95, and 5.31.

This result demonstrates a marked change in the ratio between ^{226}Ra and ^{238}U with increasing depth. In particular, at a depth of 1.00 m, the value of 5.31 at P_2 was higher than the value of 3.43 at P_1.

The measured specific activities of ^{226}Ra and ^{238}U at P_1 and P_2 and the radioactive-equilibrium coefficient $K^{226}Ra/^{238}U$ are presented in Table 1.

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Table 1.

Specific activities of ^{238}U and ^{226}Ra and the radioactive-equilibrium coefficient $K^{226}Ra/^{238}U$ at P_1 and P_2

Depth, m	P1 radionuclide values	P1 radionuclide values	P1 radionuclide values	P2 radionuclide values	P2 radionuclide values	P2 radionuclide values
Depth, m	^{238}U, Bq/kg	^{226}Ra, Bq/kg	$K^{226}Ra/^{238}U$	^{238}U, Bq/kg	^{226}Ra, Bq/kg	$K^{226}Ra/^{238}U$
0.25	1844.8	521	0.28	3160	512	0.16
0.50	1382.8	831	0.60	2640	1340	0.51
0.75	948	1100	1.16	1210	2360	1.95
1.00	408	1400	3.43	610	3240	5.31

As can be seen from Table 1, at P_2 both the initial activity of ^{238}U and the activity of ^{226}Ra in the deeper layers are higher than at P_1.

Based on these results, the dependence of the specific activities of ^{238}U and ^{226}Ra on soil-layer depth at P_1 and P_2 is shown in Figures 1 and 2.

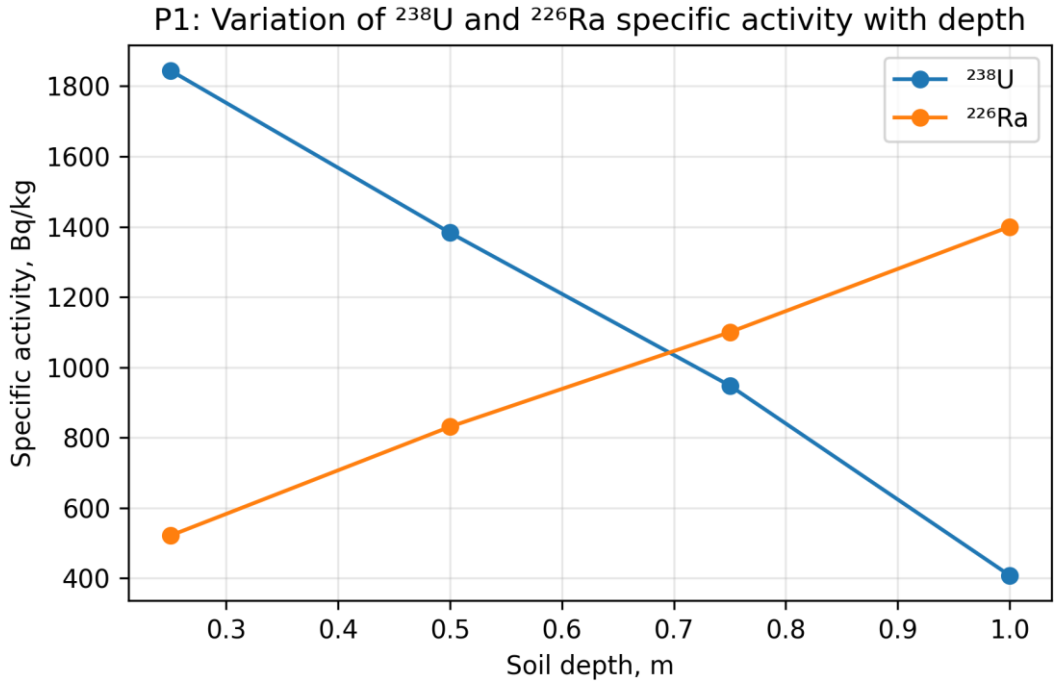


Figure 1. Dependence of the specific activities of ^{238}U and ^{226}Ra on soil-layer depth at P_1.

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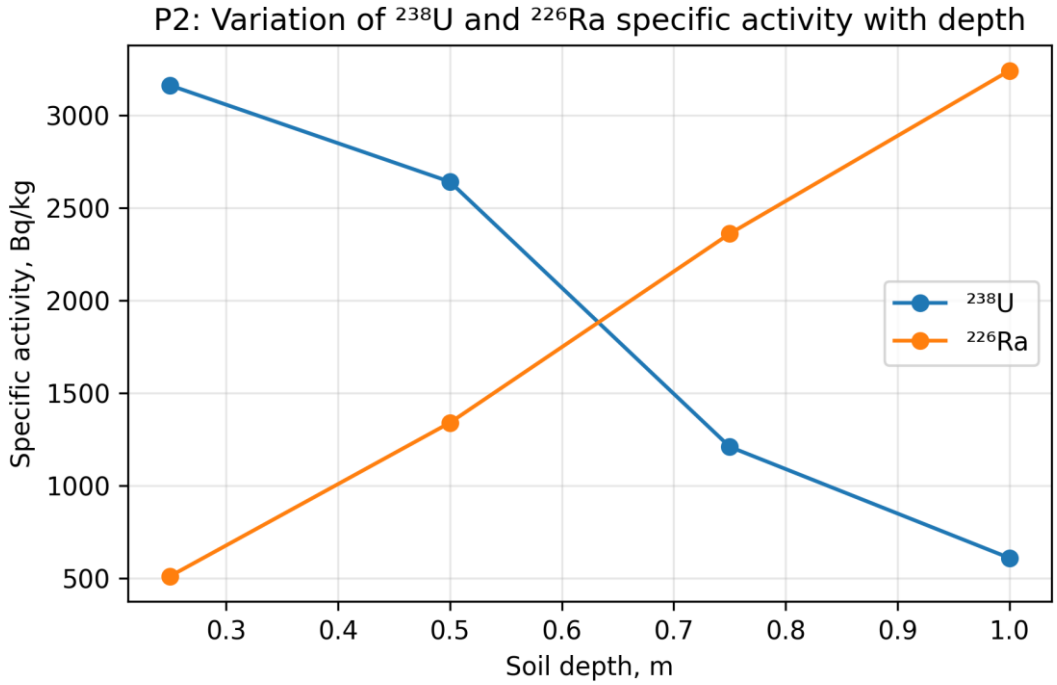


Figure 2. Dependence of the specific activities of ^{238}U and ^{226}Ra on soil-layer depth at P_2.

Based on the results of Table 1, a graph showing the variation of the radioactive-equilibrium coefficient $K^{226}Ra/^{238}U$ with soil-layer depth at P_1 and P_2 was constructed (Figure 3).

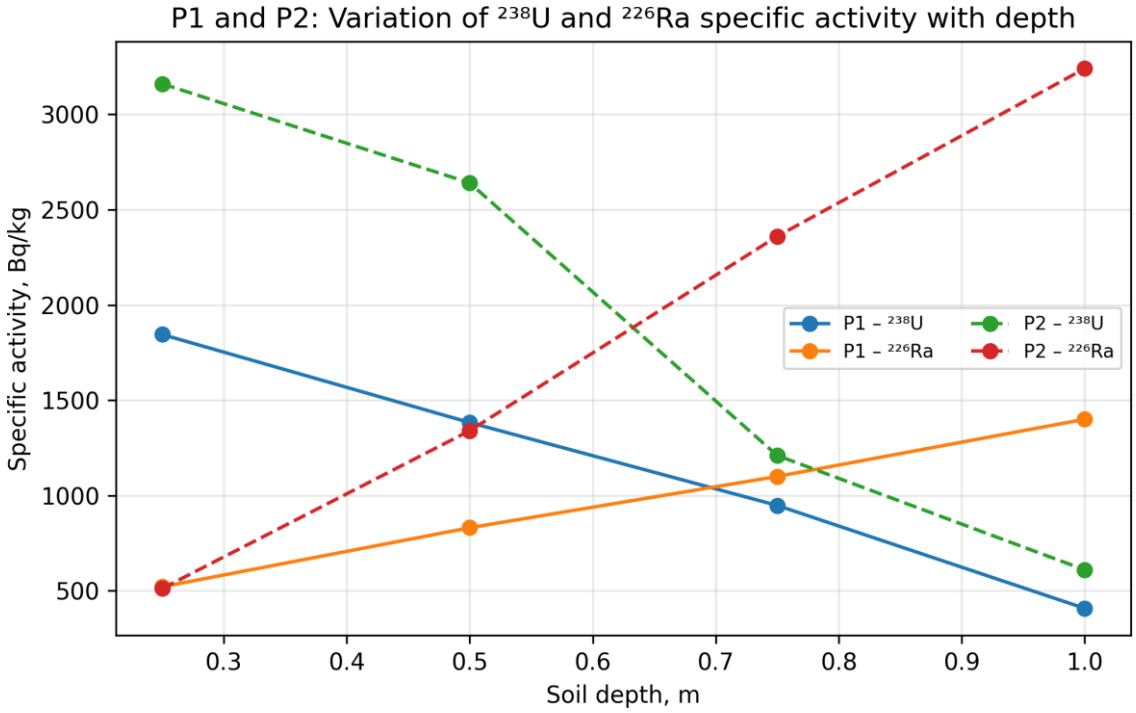


Figure 3. Graph of the depth-dependent variation of the radioactive-equilibrium coefficient $K_{^{226}Ra/^{238}U}$ between the ^{226}Ra and ^{238}U radionuclides at P_1 and P_2 based on the results of Table 1.

The obtained results show that the vertical distributions of ^{238}U and ^{226}Ra in the soil layers are not identical. The decrease in ^{238}U and the increase in ^{226}Ra with depth demonstrate an empirical difference in the way these radionuclides are distributed in the soil environment.

The results can be regarded as experimental evidence indicating the presence of vertical redistribution of radionuclides in the reclaimed area.

The study results showed that the vertical distributions of ^{238}U and ^{226}Ra in the soils of the reclaimed area do not have the same pattern. At both P1 and P2, the specific activity of ^{238}U decreased with increasing

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depth, while that of ^{226}Ra increased. This indicates that the radionuclides were redistributed differently among the soil layers.

At P_1, ^{238}U decreased from 1844.8 Bq/kg at 0.25 m to 408 Bq/kg at 1.00 m. At the same time, ^{226}Ra increased from 521 to 1400 Bq/kg. At P2, the trend was even more pronounced: ^{238}U decreased from 3160 to 610 Bq/kg, whereas ^{226}Ra increased from 512 to 3240 Bq/kg.

Conclusions

Based on the analysis of the study results, the following conclusions were drawn: clear vertical changes were observed in the distribution of ^{238}U and ^{226}Ra with soil depth at P1 and P2 in the reclaimed area; at P1, the specific activity of ^{238}U decreased from 1844.8 to 408 Bq/kg over the 0.25-1.00 m interval, while at P2 it decreased from 3160 to 610 Bq/kg; in contrast, the specific activity of ^{226}Ra increased from 521 to 1400 Bq/kg at P1 and from 512 to 3240 Bq/kg at P_2; with increasing depth, the $^{226}Ra/^{238}U$ activity ratio changed from 0.28 to 3.43 at P1 and from 0.16 to 5.31 at P_2. The obtained results indicate the presence of vertical redistribution of ^{238}U and ^{226}Ra in soil. The ^{238}U and ^{226}Ra data obtained in this study may be used as initial data for developing a mathematical model of their migration through vertical soil layers while taking radioactive-decay regularities into account.

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ISSN: 2181-3337 | SCIENTISTS.UZ