DISTRIBUTION OF ARTIFICIAL RADIONUCLIDES IN THE BALTIC SEASIDE ENVIRONMENT

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Investigation on ^{137}Cs and ^{239,240}Pu activity concentration was undertaken in a coastal zone of the Baltic Sea on the Lithuanian territory to study the vertical distribution of radionuclides (down to 30 cm). The Baltic seaside is one of the regions where the highest radionuclide concentrations after the Chernobyl NPP accident were detected. Moreover, this area is a significant recreational zone, therefore, peculiarities of radionuclide spreading in the environmental ecosystem are important from the radioecological point of view. The obtained results of vertical distribution of ^{137}Cs and ^{239,240}Pu in sand and forest soil suggest that the radionuclide downward migration depends on the structure of matrix and its chemical composition. Besides, the results of radionuclide distribution on the stripe between the Baltic Sea and the Curronian Lagoon indicate that the sea is a possible source of radioactive contaminants.

Keywords: plutonium, radiocesium, activity concentration, sand, forest soil, migration, organic matter

PACS: 28.60.+s, 82.80.-d, 89.40.Cc, 89.60.Ec

1. Introduction

When radionuclides are released into the environmental ecosystems, the knowledge concerning their transport in natural media, interactions with matrix, transformation processes, and biological uptake is needed. The understanding of these processes is important for the prediction of the further fate of radionuclides in the environment and for the assessment of long-term consequences to humans.

The major sources of man-made radionuclides in the Baltic Sea are both nuclear weapon test and Chernobyl accident fallout [1–7]. The Chernobyl accident significantly contributed to the radioactive contamination of the marine environment of the Baltic Sea. According to data published in Helsinki Commission (HELCOM) proceedings [8], 82% of ^{137}Cs and 13% of ^{90}Sr in Baltic Sea originated from Chernobyl accident, while the contribution of these radionuclides of about 14 and 81%, respectively, is from the nuclear weapons tests. The total amount of 30 TBq of ^{238}Pu and 61.5 TBq of ^{239,240}Pu in the Baltic Sea originated as a result of the atmospheric fallout derived after the accident in one of the units of Chernobyl NPP [9]. The deposition of Chernobyl origin plutonium in the Baltic Sea has been estimated to be about 10% of the total

^{239,240}Pu deposition density [1]. According to International Atomic Energy Agency (IAEA) data the total inventories of ^{137}Cs and ^{239,240}Pu, estimated in 2005, are 4.7 PBq and 16–18 TBq, respectively, for the whole Baltic Sea [10]. Other sources of anthropogenic radioactivity are discharges from operating nuclear installations around the Baltic Sea and reprocessing facilities in Sellafield and La Hague [11]. About 2 GBq of ^{239,240}Pu per year flows into the Baltic Sea with freshwater from 250 rivers [12]. The average ^{239,240}Pu activity concentration, estimated in 1997, in seawater and suspended matter was 3.5±0.5 μBq/l and 1.5 Bq/kg, respectively [4, 13]. As proposed by Holm [1], water in the Baltic Sea contains 0.6–6 μBq/l of ^{238}Pu and ^{239,240}Pu.

Though the activity concentration of ^{137}Cs in the Baltic Sea water is on decrease because of radioactive decay, sedimentation, and bioaccumulation processes, as well as the exchange of waters between the Baltic and North Sea, presently it remains most contaminated with radiocesium in comparison with any other part of the world ocean [2]. The mean activity concentration of ^{137}Cs in water was equal to 60 Bq/m^3 and the effective half-life in the Baltic Sea was calculated to be 14.1±1.8 years [10]. The cumulating and relatively

long residence time of contaminants in the Baltic Sea ecosystem is due to the fact that the Baltic Sea is a shallow, semi-closed sea with weak renewal of water mass. The average depth of the Baltic is 54 m and the renewal time of water is approximately 50 years [3].

The post-Chernobyl activity concentration values of ^{137}Cs at the Baltic coast (Curonian Spit) reached 380–440 Bq/kg, while pre-Chernobyl values varied from 4 to 8 Bq/kg [14, 15]. In 1996 the ^{239,240}Pu activity concentrations in the upper (0–5 cm) layer of beach sand varied in the range of 0.06±0.02 – 0.80±0.20 Bq/kg, meanwhile in the forest soil they ranged from 0.09±0.02 to 2.4±0.4 Bq/kg. The average ^{137}Cs activity concentrations in the beach sand and forest soil samples were determined to be 50±4 Bq/kg and 1190±45 Bq/kg, respectively [16].

The migration of radioactive contaminants in environmental media is a complicated process because of many factors. The distribution peculiarities of radionuclides in environmental compartments are strongly influenced by media type, form of deposition, sorption behaviour [17–20]. The vertical profiles of radionuclides in both the surf zone and dunes may be influenced by the seawater wash (flooding), rainfall, and weathering of the top layer of soil. Besides, the distribution pattern of radionuclides in the environment is predetermined by the features of the matrix. The low level of activity concentration of man-made radionuclides in beach sand, especially of plutonium, is a reflection of the low sorptive capacity of such accumulating material. According to [16] the low sorptive capacity is distinctive for sand or light loamy soils. The mineralogical composition of sand in the investigated near-shore zone is rather suited for adsorption of cesium. The analysis of the sand mineralogical composition has shown that it mainly consists of bazalts and granite debris with intrusion of clay minerals, carbonates, magnetites, iron oxides and hydroxides [21]. All these components of minerals are good absorbers of cesium. Cesium is particularly very strongly associated with silicates and clay [22]. As proposed by Abril [23], man-made radionuclides are generally associated within the surface layer of mineral particles only superficially. Another factor influencing the radionuclide migration in the terrestrial environment is organic matter [24]. Plutonium is known to form a number of complexes with organic and inorganic agents. The presence of these complexing agents in the environment can alter the oxidation state of plutonium, and thus can have multiple impacts on its mobility. The degradation of organic agents in the nature will change the environmental chemistry of plutonium resulting in inhomogeneous migration over time.

The Curonian Spit is a unique area due to its climatic conditions, landscape, and differences in the soil and vegetation types. Furthermore, the interaction between continental and marine air masses influences the accumulation and spreading of pollutants there. The aim of the present study is to compare distribution of activity concentration of ^{137}Cs and ^{239,240}Pu in different environmental matrices: in sand of the beach and pine forest soil.

2. Experiment

2.1. Sampling

Soil sampling was carried out in Juodkrantė on the Curonian Spit over a period of 2 years in 2003 and 2004. The sampling strategy was to compare the distribution pattern of contaminants in extremely different environment. The area contains two types of contrasting soil – beach sand and soil of pine forest. The sampling on the Baltic seaside was done according to the area topography. The samples were taken in the wash zone, in front of and on the top of the dune. In the forest, during sampling, the main attention was paid to the visual characterization of the soil composition. Figure 1 shows a map of the general area and outlines the positions of the three transect sites (*a*, *b*, and *c*). Sampling points *1–3* were located in the surf zone and dunes, points *4–6* were in the pine forest. The intervals between transects in the beach zone and dunes were of 30–50 m, and in the pine wood the distance between *4*, *5*, *6* points reached 200–300 m. The soil samples were collected with a corer with the inner cross-section of 12×12 cm^2 and the depth of 30 cm. The soil vertical profiles were sliced with a thickness of 5 cm. After sampling the soil samples were weighed and transported to the laboratory in plastic bags, dried in an oven at 100–110 °C for about 24 h and weighed again. Soil samples were then sieved through a 1-mm mesh, and remaining roots and vegetation were crushed and combined with the sieved soil.

2.2. Methods

The sand and forest soil samples collected at different distances from the sea waterline (surf zone, sand dune, and pine forest) and at different depths were analysed for ^{137}Cs by γ-spectrometry using a high-purity germanium (HPGe) detector (*CANBERRA*, resolution
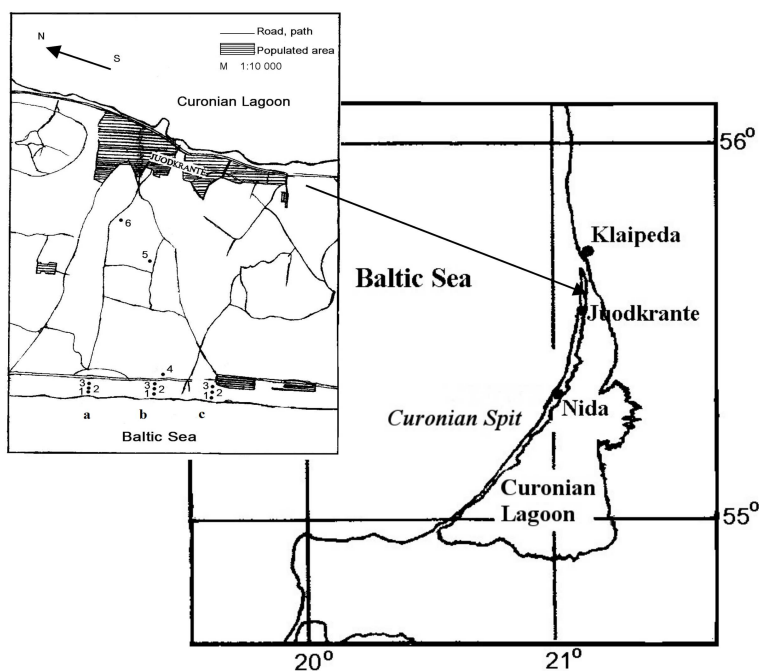
Fig. 1. Location of soil sampling points on the Curonian Spit.

2 keV, efficiency 15%). Each sample after drying and homogenization was transferred to a container with well-defined geometry and counted from 24 to 168 hours depending on the sample activity. ^{137}Cs activity was determined in terms of the peak at 661.6 keV. The background radiation spectrum was evaluated for 604800 seconds counting time. The maximum fractional measurement error did not exceed 12%. Experimental calibration of the HPGe detector was performed using a standard solid ^{152}Eu $+$ ^{137}Cs source of various densities with characteristic energies for estimation of the relationship between the channel number and gamma photon energy. The measurement accuracy was tested in the intercomparison exercises organized by Risø National Laboratory, Denmark (An Intercomparison on Radionuclides in Environmental Samples – Baltic–Danish Co-operation Project on Radiation Protection 2001–2003 and Intercomparison of Laboratory Analyses of Radionuclides in Environmental Samples – NKS-B Project LABINCO 2004–2005).

Plutonium analysis of samples followed the method of Chen et al. [25] involving ashing at 550 °C overnight and then 2-hour ashing at 700 °C in a muffle furnace, acid-leaching, anion-exchange, electrodeposition, and α-spectrometry. ^{242}Pu as internal tracer had been used. After electrodeposition on the stainless steel discs the samples were measured with the alpha-spectrometer OCTETE Plus (*ORTEC*) with the detector of 450 mm^2. The counting time was fixed to 198000 seconds depending on the sample activity. Alpha-counting effi-
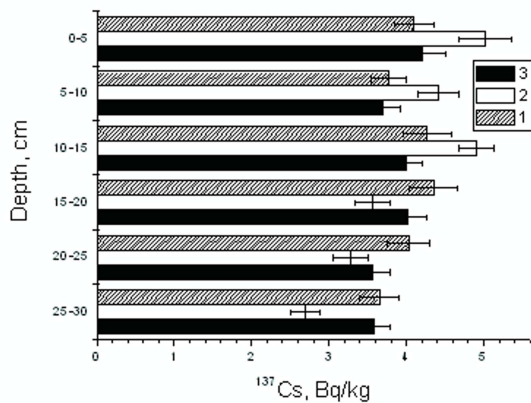
ciency and resolution were 25% and 19 keV, respectively. The detection limit for a counting time of 86400 seconds was about 10^{-3} Bq of ^{239,240}Pu.

The organic matter content of soil was estimated by loss on ignition at 550 °C in a muffle oven [25]. The pH$_{KCl}$ measurements were performed using a WTW pH-meter pH 315i with a measurement error of ± 0.01 pH unit [26].
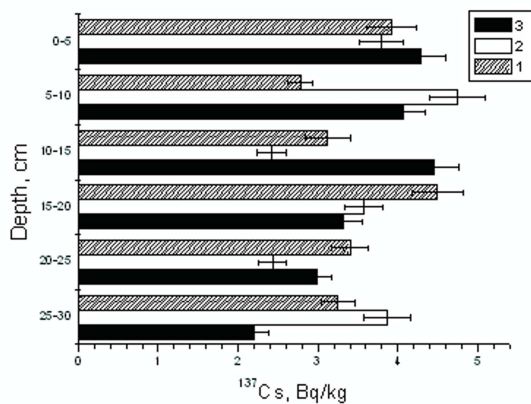
3. Results

The widest range of ^{137}Cs activity concentrations was observed in the upper 0–5 cm soil layer of the studied area. In the surf zone and dunes (points *1–3*, Fig. 2) they varied in the range of 2.5–5.0 Bq/kg, while in a pinery (points *4–6*, Fig. 3) they ranged from 58.4 to 339 Bq/kg. Activity concentration of ^{137}Cs in the 5–30 cm layer of the surf zone and the sand dune was in the range of 2.2–6.8 Bq/kg. Depth profiles *1–3* of ^{137}Cs activity concentration in the surf zone and dunes have shown rather homogeneous distribution of activity concentration (Fig. 2), the activity concentrations vary at different depths but the variations observed are not significant.
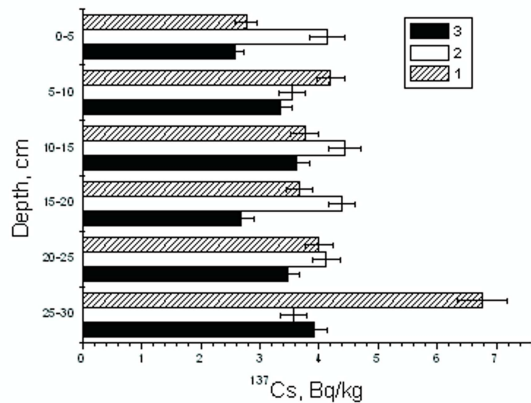
The maximum activity concentration of ^{137}Cs was obtained in the 0–5 cm layer of pine forest soil (profiles *5, 6*). The exponential drop of activity concentration is characteristic of vertical distribution of ^{137}Cs (Fig. 3). The lowest value of ^{137}Cs activity concentration (0.2–

(a)

(b)

(c)

Fig. 2. Depth distribution of ^{137}Cs activity concentration in sand in vertical profiles *1*, *2*, *3* of transects *a*, *b*, and *c*.

Fig. 3. Depth distribution of ^{137}Cs activity concentration in forest soil in vertical profiles *4*, *5*, *6* of transect *b*.

ranged from 0.002 to 0.021 Bq/kg in a depth profile of up to 30 cm. The maximum value of ^{239,240}Pu activity concentration of 0.021 Bq/kg was found on the top of the dune at the depth of 10–15 cm. The activity concentration of ^{239,240}Pu did not exceed 0.015 Bq/kg in the surf zone in front of the dune. The marked enlargement of ^{239,240}Pu activity concentration in the vertical profile of forest soil was determined. The ^{239,240}Pu activity concentration in the 0–30 cm forest soil profile ranged from 0.037 to 4.30 Bq/kg. The maximum activity concentration of this radionuclide was observed in the 5–10 cm soil layer of profile *6* (Fig. 5). The value of plutonium concentration at this sampling point reached up to 4.30 Bq/kg. Moreover, sufficiently high ^{239,240}Pu activity concentrations of 3.35 and 3.39 Bq/kg, respectively, were found both in 0–5 and 5–10 cm soil layers of profile *5*.

As shown in Figs. 3 and 5, the lowest activity concentrations for ^{137}Cs and ^{239,240}Pu were found in vertical profile *4* of forest soil. The amount of both radionuclides therein is by several orders of magnitude less than the amount obtained in vertical profiles *5* and *6*. The highest ^{137}Cs activity concentration reached 58.3 Bq/kg in the top layer (0–5 cm) and then sharply decreased with the depth with variation limits from 3.42 to 0.18 Bq/kg. The activity concentration of ^{239,240}Pu in soil samples of profile *4* was in the range of 0.08–0.01 Bq/kg with maximum activity in the top soil layer (0–5 cm). These results may be explained by the conditions of profile *4* location which are much more similar to the environment of the dune than to the forest. The vegetation cover here consists of thin grass and forest floor, the pines are growing at rare intervals. The amount of organic matter varied from 0.3 to 7.5%, while in the forest soil samples it was in

10.4 Bq/kg) was determined in the forest soil in the 25–30 cm layer.

The vertical distribution of activity concentration of ^{239,240}Pu in the surf zone and sand dunes was found to be non-uniform at different depths (Fig. 4). The observed variations of activity concentration fluctuated in a wide range. The activity concentration of ^{239,240}Pu
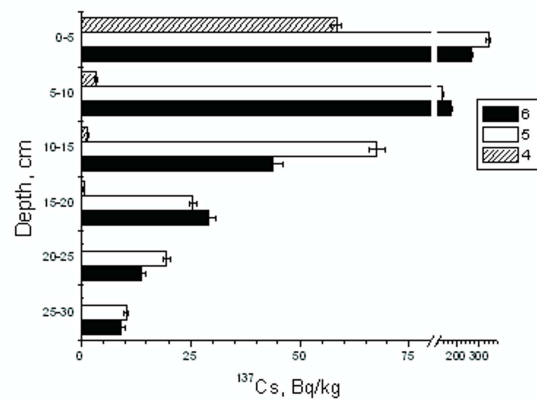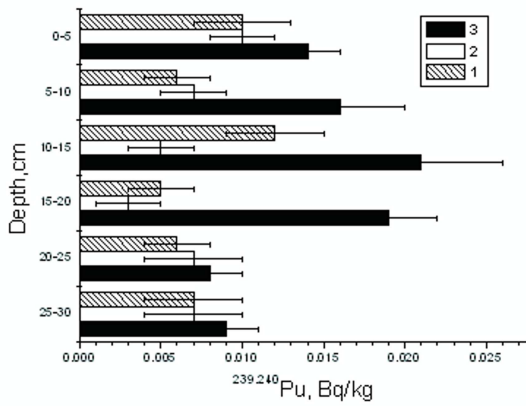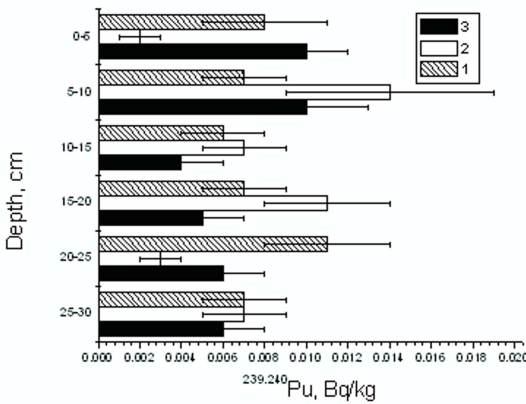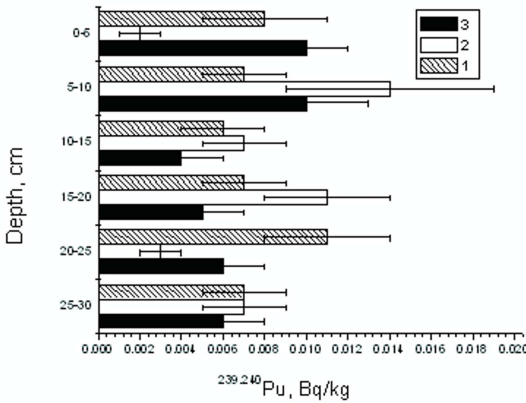
(a)

(b)

(c)

Fig. 4. Depth distribution of ^{239,240}Pu activity concentration in sand in vertical profiles *1*, *2*, *3* of transects *a*, *b*, and *c*.

Fig. 5. Depth distribution of ^{239,240}Pu activity concentration in forest soil in vertical profiles *4*, *5*, *6* of transect *b*.

for the 0–30 cm layer in the investigated area was 0.93 and 0.87, respectively. In the surf zone and sand dune samples the pH values varied from 5.59 to 6.70, and in the forest soil from 2.92 to 6.65. In the 0–30 cm sand layer of the investigated area, the correlation coefficient between ^{137}Cs and ^{239,240}Pu activities and pH was −0.47 and −0.35, respectively.

4. Discussion and conclusions

Current levels of contamination of the Baltic coastal zone with ^{137}Cs and ^{239,240}Pu and their depth profiles were identified. The maximum concentration of up to 339 Bq/kg of ^{137}Cs and up to 4.30 Bq/kg of ^{239,240}Pu was determined in the upper (0–5 cm) soil layer of the surf zone, the sand dunes, and pine forest. The activity concentrations of both ^{137}Cs and ^{239,240}Pu at the Baltic coast are close to the post-Chernobyl accident values [14]. The high amount of radionuclides in the upper soil layer in forest suggested that the vertical migration of radionuclides was very slow and the soil became the major reservoir of contaminants.

As the investigations have shown, the vertical distribution of both cesium and plutonium in the sand layer (0–30 cm) is moderately even. The sand of a narrow stripe of the coastal zone is being mixed because of the heavy swell or wind. On the other hand, as the sand is composed of mineral particles, there the tenuous geochemical processes do not strongly affect sorption of both cesium and plutonium, resulting in their negligible concentration in the full-length horizon. Thus, the vertical distribution of radionuclides has been caused by physical redistribution processes. According to [22], the radionuclide-bearing particles have been dispersed

the range of 9–91% with the highest values being attributed to the samples of 0–5 cm depth. The quantity of organic matter in sand samples did not exceed 1.5%. To evaluate the effect of this parameter on accumulation of radionuclides, a linear correlation analysis was performed. The correlation coefficient for ^{137}Cs and ^{239,240}Pu activities with the organic matter content (%)

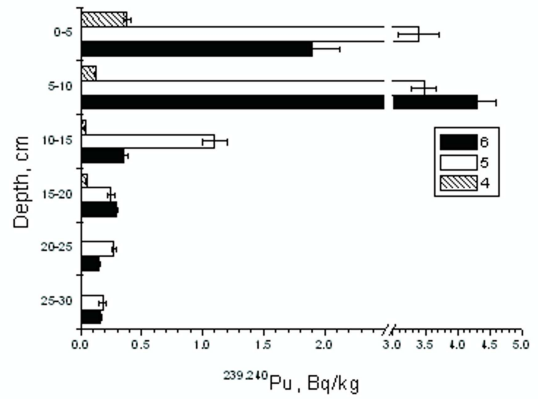horizontally and vertically because of wind and water in the coastal zone.

Sufficiently high plutonium concentrations obtained by us in the surf zone are comparable to measurements reported previously for the Baltic Sea water [1] and enable us to suppose that plutonium together with marine particles depending on their size are transported farther to the continent or deposited close to the sea. Furthermore, the mechanism of sea-to-land transfer, when radionuclides adhered to fine particulate material in the sea can be transported to the surface microlayer by air bubbles and then released with spray droplets to the surf zone, is well known [27].

In the forest the direct influence of sea on the vertical distribution of activity concentration of radionuclides is less considerable. Here the dispersion of radionuclides is caused by interaction between matrix and contaminants in forest soil. The clay content in the forest soil is very low, therefore the role of the organic component of the humic-rich soil in accumulation of ^{137}Cs is rather significant [28, 29]. Especially plutonium has strong affinity to humic substances. A very negligible amount of soil organic matter could affect the chemical nature of plutonium due to formation of stable organic complexes. The chemical transformation of plutonium could considerably influence its solubility and accumulation potentiality [22]. For example, in the soil layer, which contained 40–49% of organic carbon, the ^{239,240}Pu activity concentration up to 87% was observed. However, in the mineral soil, the amount of organic carbon varied from 0.9 to 2.8%, and plutonium activity concentration in organic fraction varied between 30 and 60% [27]. Therefore, the activity concentration of both radionuclides was significantly higher in the forest soil than in sand. The obtained results of vertical distribution of ^{137}Cs and ^{239,240}Pu in sand and forest soil suggest that the radionuclide downward migration depends on the structure of matrix and its chemical composition. Besides, the results of radionuclide distribution on the stripe between the Baltic Sea and the Curronian Lagoon indicate that the sea is a possible source of radioactive contaminants.

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DIRBTINIŲ RADIONUKLIDŲ PASISKIRSTYMAS BALTIJOS PAJŪRYJE

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Santrauka

Tirtas ^{239,240}Pu ir ^{137}Cs savitojo aktyvumo vertikalus pasiskirstymas Kuršių nerijos miško dirvožemio ir Baltijos pajūrio smėlio 30 cm paviršiaus sluoksnyje. Didžiausios ^{137}Cs ir ^{239,240}Pu savitojo aktyvumo vertės nustatytos miško dirvožemio 0–5 cm sluoksnyje, giliau jos eksponentiškai mažėjo. Pakrantės smėlio vertikaliame profilyje abiejų radionuklidų savitasis aktyvumas pasiskirstęs tolygiai. Tyrimų rezultatai parodė, kad radionuklidų vertikalią migraciją miško dirvožemyje lemia organinė medžiaga, kurios kiekis siekia iki 91 % viršutiniame 5 cm sluoksnyje. Pakrantės smėlyje radionuklidų savitojo aktyvumo vertikalųjį pasiskirstymą lemia mineralinė matricos sudėtis.