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^{137}Cs AND ^{239,240}Pu ACTIVITY CONCENTRATIONS DISTRIBUTION IN WATERLOGGED AND NON-BOGGY SOILS OF LITHUANIA

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Vertical distributions of ^{239,240}Pu and ^{137}Cs activity concentrations in the waterlogged and non-boggy soil cores sampled in the vicinity of Vilnius City and in the Varėna District were analyzed. The radionuclides appeared in the soils mainly as a result of the fallout after the nuclear weapon testing in atmosphere in 1960s and due to the Chernobyl NPP accident in 1986. The deposited radionuclides on the surface of waterlogged soils on the shores of lakes experienced processes of translocation and accumulation, therefore their total activity in the column differs from the activity in non-boggy soils. The mobility of plutonium is the highest in waterlogged organic-rich soils. Clay in the waterlogged soil significantly limits the migration of radiocesium and does not affect noticeably the mobility of plutonium. Compared with radiocesium, the mobility of plutonium in non-boggy soils is somewhat limited. So, in waterlogged and non-boggy soils, the radionuclide mobilizing factors act in the opposite directions for radiocesium and plutonium. A clear correlation between the radionuclide activity and soil organic matter content was not observed. The investigation showed that radionuclide mobility can be determined by studying radionuclide physicochemical forms in the soil, vertical profiles of radiocesium to plutonium ratios, soil composition as well as its oxidation regime.

Keywords: plutonium, radiocesium, soil organic matter, waterlogged soil, non-boggy soil

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1. Introduction

Soil is a complex heterogeneous system that is composed of mineral phases and organic matter, as well as of liquid and gaseous components. The contact interface of mineral phase and water in the soil is the reactive surface where sorption processes occur [1]. Precipitations of the isotopes of anthropogenic radionuclides ^{137}Cs and Pu in soils resulted from the global fallout of nuclear weapon tests (1952–1963) and after the Chernobyl Nuclear Power Plant accident (1986). ^{238}Pu/^{239,240}Pu, ^{240}Pu/^{239}Pu and ^{241}Pu/^{239}Pu activity ratios are widely used to identify the origin of plutonium contamination. Each plutonium contamination source shows an individual composition of plutonium isotopes. The ^{238}Pu/^{239,240}Pu ratios characteristic of the global fallout should currently be almost 0.028 [2]. The higher ^{238}Pu/^{239,240}Pu ratio of ~0.3 is characteristic of the releases of plutonium isotopes after the accident of Chernobyl NPP [3]. Besides, ^{239,240}Pu may be used to identify ^{137}Cs sources according to ^{239,240}Pu/^{137}Cs or ^{137}Cs/^{239,240}Pu activity ratios [4]. The plutonium to radiocesium ratio (~0.018) measured in 1963 is often used as typical of the global fallout [5]. At present, the radiocesium to plutonium ratio typical of the global fallout is equal to almost ~28 [6]. When technogenic radionuclides are deposited on the surface of the soil, soil particles greatly adsorb the radionuclides and form new compounds (radionuclides are caught not only by adsorption on reactive sites of the clay mineral of soil particles, or by ion exchange and precipitation as hydroxide or sulfide, but also by complex formation with organic compounds). In that case, the soil becomes a radionuclide reservoir in natural environment. Therefore, it is very important to analyze the behaviour of ^{137}Cs and Pu isotopes in nature by studying the soil as a system, which is composed of many compounds that influence the migration of radionuclides.

The natural phenomenon such as soil waterlogging due to floods or snowmelt strongly affects all components of the soil as well as influences soil-forming and radionuclide migration processes [7]. Study of the migration behaviour of radionuclides in waterlogged soils is important to gain a better insight into the mechanisms of natural self-cleaning in contaminated areas [8]. As waterlogging can create an anaerobic and reducing environment in the soil, factors that have the major influence on seasonally waterlogged (or wetland) soil include oxidation–reduction, dissolution–precipitation and adsorption–desorption processes [9].

The major diagenetic processes (the process by which sediment turns into the rock) in coastal sediments which influence the partitioning, solubility and mobility of radionuclides include degradation of organic matter with their consequent release into interstitial pore waters, bacterially mediated reductive dissolution of iron and manganese oxyhydroxides and sulphate reduction with subsequent production of soluble and insoluble sulphides. In the case of plutonium, with multiple oxidation states, redox transformations are of a particular significance. Clearly, the importance of these post-depositional processes in relation to transfer and/or remobilization will depend on the associated plutonium [10].

The primary factor in determining the mobility of plutonium in the groundwater environment is its oxidation state because it can simultaneously exist as Pu (III), Pu (IV), Pu (V) and Pu (VI) in a single solution [11]. This ability to exist in multiple oxidation states arises from similarities in reduction potentials and disproportionation. Reduction of Pu (V) or Pu (VI) to Pu (IV) results in lower solubility and increased favourability for mineral surfaces as summarized by the actinide trend which is based on the effective charge of each plutonium cation [11]. The actinide trend tells us that Pu (IV) is generally assumed to be the least mobile oxidation state of plutonium and Pu (V) the most mobile one. Each plutonium oxidation state has a different chemical behaviour, complicating prediction of the geochemical behaviour [12]. Originally insoluble PuO_2 (IV) is the most likely chemical form of Pu in deposited fallout particles. In the soil environment, complex formations occur by the interaction of fallout Pu with surfaces, surface coatings, colloidal materials and complexing ligands. Once Pu (IV) is stabilized by naturally occurring ligands such as humic and fulvic acids, Pu can remain in the soluble state [13]. It is known that dissolved organic carbon (DOC) in natural waters mostly originates from dissolved fulvic and humic acids. Under oxidizing conditions, the predominant forms of plutonium are Pu $(V)O_2^+$ and Pu $(VI)O_2^{2+}$, which form distinct inorganic/organic complexes when present as a contaminant in groundwater [14]. While PuO_2^+ (i.e. Pu (V)) does not form hydroxyl complexes up to pH > 8 and should be very mobile under most groundwater conditions, PuO_2^{2+} (i.e. Pu (VI)) forms strong hydroxyl complexes and sorbs strongly to aquifer surfaces [15]. Plutonium can exist in the groundwater when it is in a reduced form. Ferric iron (Fe^{3+}) is often present in contaminated sites, and its bioreduction to ferrous iron (Fe^{2+}) can lead to the reduction of Pu with the formation of a precipitate. On the other hand, Pu can be reduced directly by bacteria [15].

$^{137}Cs^+$ deposition in soil originated due to several ways: directly from atmosphere as global fallout, wash-off from vegetation, turnover from vegetation, re-deposition of eroded soil particles and deposition from water on floodplains and coastal regions. $^{137}Cs^+$ is rapidly and strongly adsorbed by soil particles, especially by clay minerals [16–17]. The highly selective sorption of trace amounts of $^{137}Cs^+$ occurs at the frayed edges of illitic type clay minerals [18–20]. In these highly selective sites only other cations with low hydration energies (notably K^+ and NH_4^+) can compete with $^{137}Cs^+$ [21]. $^{137}Cs^+$ interacts with all the charged surfaces in soil or sediments [18]. In soils, $^{137}Cs^+$ can be effectively adsorbed by clay minerals, particularly illite [19], but adsorption of $^{137}Cs^+$ on clay is diminished in the presence of organic matter [22]. Organic matter has a smaller affinity for $^{137}Cs^+$ in comparison to clay minerals, thus the extent of adsorption is limited due to its low selectivity for $^{137}Cs^+$ [22]. $^{137}Cs^+$ has a very small hydration energy, therefore the electrostatic attraction of ^{137}Cs ions by clay particles is large and they are preferentially sorbed [23]. The adsorption of ^{137}Cs mainly occurs through an anion exchange process and also depends on the content of organic matter. Generally, in the soil with a high organic matter content its adsorption is reversible and it is then more available for uptake by plants [24–25]. As the sorption of cations is known to be controlled by the ionic strength and pH of the system, it is important to understand the sorption behaviour of $^{137}Cs^+$ under different geochemical conditions [26]. The interactions between $^{137}Cs^+$ and organic matter are quite complex and there is evidence for $^{137}Cs^+$ immobility even in highly organic soils [5]. However, the adsorption of $^{137}Cs^+$ to organic matter is determined by its concentration in relation to other competing cations and the cation exchange capacity (CEC) of the specific organic substance [27–28]. The effect of pH on $^{137}Cs^+$ sorption includes the increase of variable negative charge, the modification of metal speciation, the displacement of the equilibrium of surface complexation reaction and the competition of H_3O^+ for negative sites [29]. The sorption of $^{137}Cs^+$ to iron oxides and organic matter is known to be negligible [30].

Plutonium isotopes and ^{137}Cs can be found on the Earth surface bound to soil components, penetrating down the soil profiles, or finally passing into sediments. The speed of movement of radionuclides in the environment depends on many factors such as the radionuclide chemical form, the chemical composition of the soil, physicochemical conditions and the amount of atmospheric precipitation, the presence of vegetation and its species, the activity of microorganisms and small animal's activities (earthworms), as well as human activities. Numerous works have been published on the vertical distribution of radionuclides in waterlogged and non-boggy soils [31–35]. In particular, the vertical transfer of various radionuclides in soil [36–37] and their binding to soil fractions with different physicochemical properties using sequential extraction [38] and transfer to plants [39] have been studied. The studies concerned, firstly, assessing the level of soil contamination with ^{137}Cs and plutonium isotopes, and secondly, calculating the rate of vertical migration of radioisotopes in different types of soils. It is generally assumed that radionuclides are distributed in soil according to an exponential profile [32]. The origin of the radionuclide is determined by studying soil profiles in accordance with ^{238}Pu/^{239,240}Pu, ^{241}Pu/^{239,240}Pu and ^{137}Cs/^{239,240}Pu and other ratios [4]. The analysis of the waterlogged soil profile shows two or more peaks of radionuclide activity concentrations [4, 33–34]. Vertical profiles of radionuclide activity concentrations in soil with several peaks have caused a lot of controversy among scientists, since the origin of these secondary peaks in the profile is unclear. Currently, there is little information about the behaviour of plutonium in contaminated soils or subsurface environments depending on natural processes. Understanding the environmental processes that can potentially affect the speciation and mobility of plutonium would enable better management of Pu-contaminated sites. Waterlogging is among many environmental processes that can affect Pu speciation. In particular, the reduction of metals under anaerobic conditions can potentially have a significant effect on the speciation of Pu in subsurface environments.

The aim of the present study was to investigate the vertical profiles of ^{238}Pu and ^{239,240}Pu activity concentrations in waterlogged and non-boggy soils, to compare the distributions of plutonium activity concentrations in the studied profiles with those of ^{137}Cs and, based on experimental results, to determine the origin of Pu and ^{137}Cs in the studied soils. Such comparative studies in seasonally waterlogged and non-boggy soils are limited.

2. Materials and methods

2.1. Soil sampling and sampling area

The waterlogged and non-boggy soil cores were taken in different locations of Lithuania from the shores of three lakes (Lake Juodis, Lake Paršelis and Lake Bedugnis). All sampling points were selected on the shores of lakes with undisturbed soils, which have not been processed for many years. In these lakes, the increased water level due to rain and melting snow repeats every year, thereby flooding the nearby coast. This flooded soil is considered to be waterlogged soil. The sampling sites of this type were located at a distance of 1–1.5 m from the water edge of the lake. The collection sites for samples of non-boggy soils were located at a distance of ~10 m from the water edge of a lake and were never flooded. Lake Juodis (N 54°46'54", E 25°26'54") is a small lake in the north eastern part of Vilnius, located about 0.5 km south of Lake Balžis in the Tapeliai Landscape Reserve. Lake Juodis belongs to the Neris River basin. The area of the lake is 9.1 ha. The lake is elongated in shape in the northeastern direction by 0.73 km. The maximum width of the lake is approximately 0.17 km. The shores are high, only the northern coast is low and swampy. The soil sample was taken in 2012. The non-boggy soil core was taken at the western shore of the lake. The waterlogged soil core was taken in the vicinity of the lake in an old canal, which was periodically flooded in spring.

Lake Bedugnis (N 54°11'12'', E 24°09'14'') is located in the Varėna District (the southern part of Lithuania). The lake has an oval shape, the length from the southwest to the northeast is 0.18 km and the width is up to 0.12 km. The coasts are high, completely overgrown with trees, only the northern shore is low and swampy. The lake is not running and it belongs to the Kempė stream basin. The soil sample was taken in 2018. Impurities of clay substances are characteristic of the region soils. According to the New Classification of Soils of Lithuania [40] soil in the Varėna District is described as arenosol. It consists essentially of more than 70% of sand (0.05–2.0 mm) and less than 15% of clay (<0.002 mm).

Lake Paršelis (N 54°05'0'', E 24°41'45'') is located in a hollow in the Varėna District. The lake has an oval shape: the length from north to south is 0.5 km and the width is up to 0.35 km. Like Lake Bedugnis, Lake Paršelis has also no bottom sources and is fed exclusively by atmospheric precipitations. Lake shores are with steep slopes and the coastal part is low and swampy. The lake belongs to the Ūla River basin. The soil sample was taken in 2015.

Samples were taken by stabbing a steel tube of 30 cm height (diameter 12.5 cm). In the laboratory, all soil cores were sliced into 1 cm-thick layers. The uppermost layers of the soil samples taken for the study consisted of live moss and needles.

2.2. Radiochemical analysis and measurements

After slicing, the samples were dried for 24 h at 105°C and then homogenized. For the Pu radiochemical analysis, the samples were burned in a muffle furnace at 500°C overnight and then at 700°C for 2 h to decompose the organic matter. The organic matter content was determined on loss of ignition. The samples were spiked with a 0.0125 Bq ^{242}Pu tracer as a yield monitor. Acid leaching with aqua regia and concentrated HNO_3 was employed. The leachate was filtered (Whatman 114, pore size 20–30 μm) and evaporated until wet residue.

The evaluation of Pu isotope activity concentration requires special procedures of radiochemical separation. Plutonium was pre-concentrated and purified by using ion exchange resin DOWEX 1×8 (NO_3^-), then the samples were processed and electrochemically deposited on stainless steel disks and measured by alpha spectrometry [41–42]. The plutonium activity concentration was measured with a semiconductor alpha spectrometer Octete Plus with the *ORTEC* large area (450 mm^2) Si detectors (BU-020-450-AS) with the resolution capability of 20 keV. The spectra were analyzed applying Alpha Vision and Maestro programs. The detection limit for the counting time of 86400 s was about 10^{-3} Bq for plutonium isotopes.

The soil samples were analysed for ^{137}Cs using a Canberra gamma-spectrometric system with a HPGe detector (model GC2520, 26.2% relative efficiency, resolution 1.76 keV/1.33 MeV) according to the gamma line at 661.62 keV of ^{137m}Ba (a daughter product of ^{137}Cs). Measurements of the soil samples were carried out in standard geometry ('dental' plastic containers) and at known efficiencies (~1.6 – ~1.8%), which depended on the packing parameters of the sample as well as on the sample weight according to the density of samples. The radionuclide mixtures (^{152}Eu + ^{137}Cs) with two different densities (1.0 and 1.45 kg/L) prepared by the Russian Scientific Research Institute of Physical-Technical and Radiometric Measurements (Moscow, Russia) were used for efficiency calibrations. The calibration quality was certified using IAEA standards. Measurement errors of the ^{137}Cs activity concentration in the samples (standard deviation) were evaluated using the GENIE software program. They were less than 5% (standard deviation) for the measuring time of about 86400 s for the active samples corresponding to the soil layers with the maximal radiocesium activity concentrations in the respective vertical profiles and were not higher than 20% for less active samples (usually, the samples of low activity, corresponding to the tails of the respective vertical profiles) when the above errors were achieved during long-term measurements up to 500000 s.

Clay minerals were separated using the sieves with pore sizes of 1, 0.25, 0.1 and 0.063 mm instead of the usual sedimentation method due to a too small quantity of the soil. The last (smallest) fraction was used for X-ray diffraction (XRD) measurements. The XRD measurements were performed using a X-ray diffractometer Smart Lab (*Rigaku*) with a 9 kW X-ray source with a rotating Cu anode. The results of the XRD measurements allowed us to identify clay impurity, however, the composition of soil mineral content could not be determined due to the deficiency of the samples.

The results of the XRD measurements allowed us to identify the presence of clay impurities (muscovite $KAl_3Si_3O_{10}(OH)_2$) in the studied samples. The studied samples also contained a large amount of quartz and other minerals such as albite, anorthite and orthoclase.

3. Results and discussion

3.1. Vertical profiles of ^{137}Cs and ^{239,240}Pu activity concentration in non-boggy and waterlogged soil cores

The vertical profiles of ^{137}Cs activity concentration as well as the organic matter content in the waterlogged and non-boggy soil cores were measured down to the 30 cm depth. The respective plutonium vertical profiles were measured down to the 30 cm depth in the soil cores near Lake Bedugnis and in the waterlogged soil core taken at the shore of Lake Juodis. In the non-boggy soil core taken near Lake Juodis, these measurements were carried out down to the 22 cm depth and in the both cores taken near Lake Paršelis down to the 15 cm depth. The activity concentrations of ^{239,240}Pu and ^{137}Cs in the waterlogged and non-boggy soil samples next to Lake Juodis, Lake Bedugnis and Lake Paršelis are presented in Figs. 1–3. According to the presented data (Figs. 1–3), the highest activity concentrations of ^{137}Cs and ^{239,240}Pu were in the surface layers down to the 5–8 cm depths.

The analysis of the vertical profiles of the radionuclide activity concentration in the non-boggy and waterlogged soil cores showed two more or less pronounced peaks of activity concentration. Two peaks of ^{239,240}Pu activity concentrations were determined in all profiles of the studied waterlogged and non-boggy soil profiles and three in the profile of the waterlogged soil of Lake Bedugnis (Figs. 1(a), 2(a) and 3(a)). All peaks of ^{239,240}Pu activity concentration are clearly visible, because these layers contain a relatively large amount of organic matter (see Fig. 4). Two peaks of ^{137}Cs activity concentrations were determined in the soil profiles of the waterlogged and non-boggy soil of Lake Juodis and Lake Bedugnis and one peak in the profiles of the waterlogged and non-boggy soil of Lake Paršelis

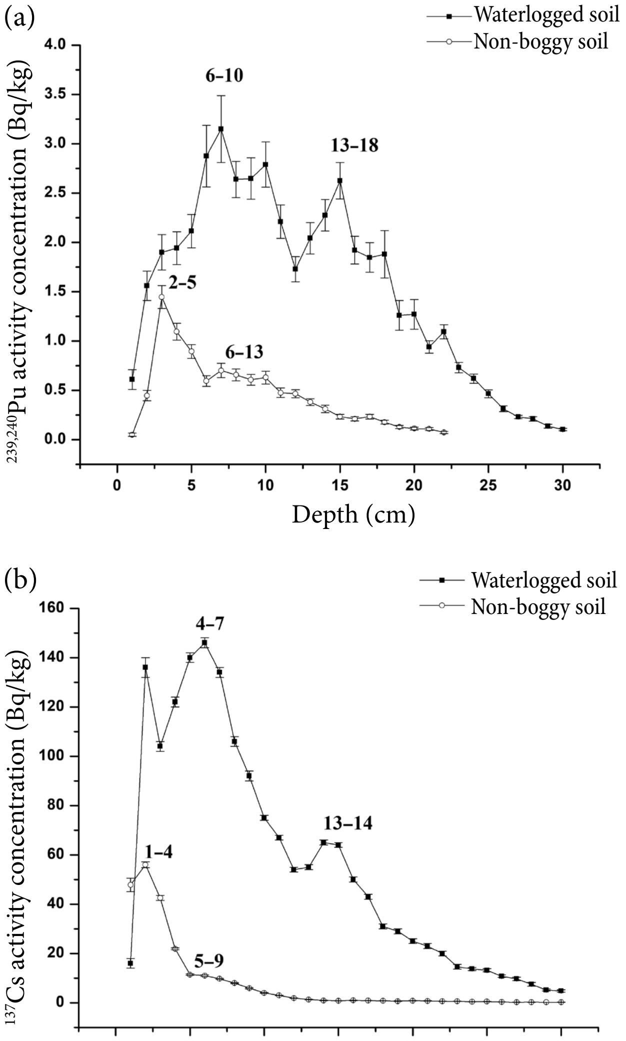

Fig. 1. Vertical profiles of the ^{239,240}Pu (a) and ^{137}Cs (b) activity concentrations in the waterlogged and non-boggy soil cores taken near Lake Juodis. ^{239,240}Pu activity concentrations were measured in the non-boggy soil core down to a depth of 22 cm.

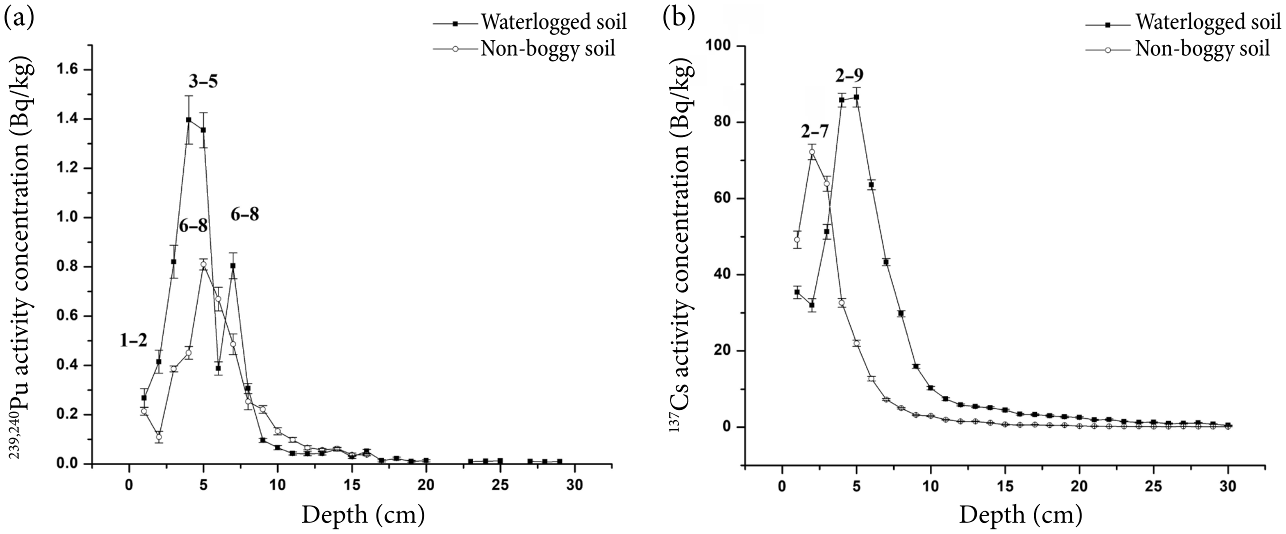

Fig. 2. Vertical profiles of the ^{239,240}Pu (a) and ^{137}Cs (b) activity concentrations in the studied waterlogged and non-boggy soil cores taken near Lake Paršelis. ^{239,240}Pu activity concentrations were measured in the soil cores down to a depth of 15 cm.

(Figs. 1(b), 2(b) and 3(b)). The secondary peaks of ^{137}Cs activity concentration are not clearly visible on the non-boggy soil profiles of Lake Juodis and Lake Bedugnis, because there is a very small amount of organic matter in these layers (see Fig. 4).

Different depths of ^{239,240}Pu and ^{137}Cs activity concentrations peaks in the respective vertical profiles of waterlogged and non-boggy soil cores may indicate different migration abilities of the radionuclides in those media. Thus, more enhanced migration of both radionuclides is evident in the waterlogged soil of an old canal in the vicinity of Lake Juodis (Fig. 1(a, b)). In the absence of adsorbing substances in the soil, radionuclides can migrate deep into the soil, reaching the groundwater level. The same feature can be seen for radiocesium and plutonium in the waterlogged soil at the shore of Lake Paršelis (Fig. 2(a, b)). In the waterlogged soil core taken near Lake Bedugnis (Fig. 3(b)), the radiocesium peak is located deeper than that in the non-boggy soil. This can also be considered as the evidence of the higher mobility of radiocesium in the waterlogged soil. However, this result is only apparent. The fact is that in waterlogged soils

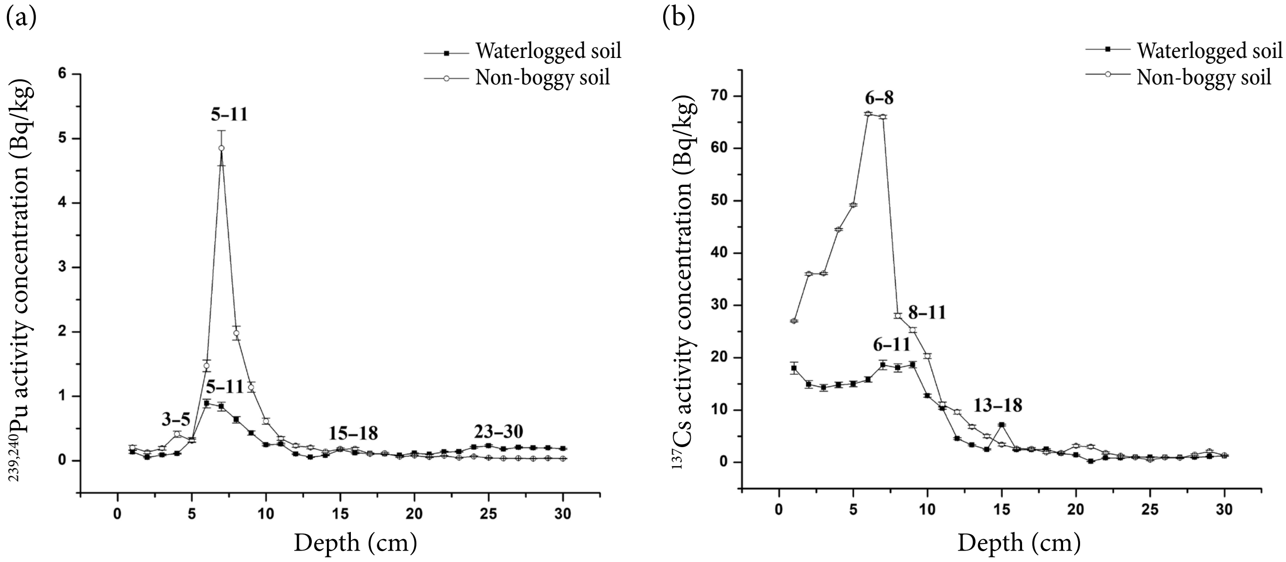

Fig. 3. Vertical profiles of the ^{239,240}Pu (a) and ^{137}Cs (b) activity concentrations in the studied waterlogged and non-boggy soil cores taken near Lake Bedugnis.

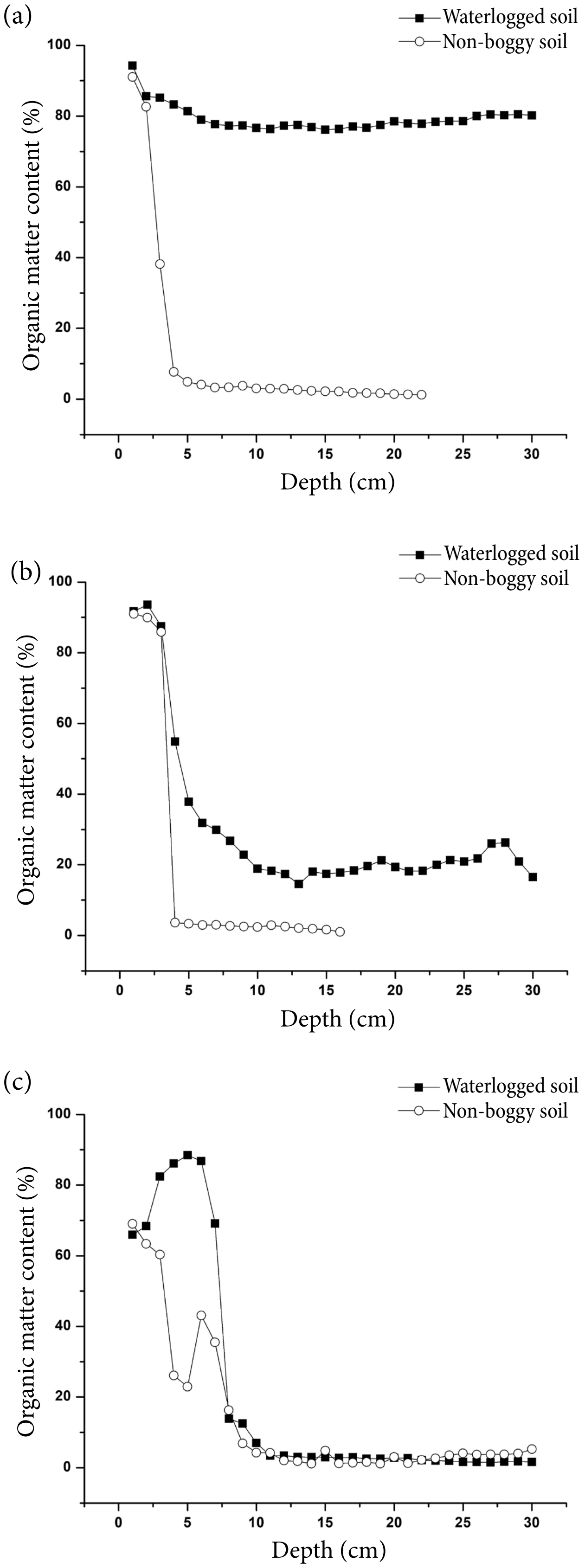

Fig. 4. Vertical profiles of the organic matter content (%) in the waterlogged and non-boggy soil cores taken near Lake Juodis (a), Lake Paršelis (b) and Lake Bedugnis (c).

during spring floods an additional layer of sediments may appear, contributing to the deepening of the peak of the radionuclide activity concentration. Also, it seems that in the waterlogged soil, radiocesium may be more mobile than plutonium, since its peak is located deeper. However, this may also be due to the radiocesium dominant uptake in these layers (6–10 cm) by impurities of clay materials, which are characteristic of these soils. Also, as can easily be seen from Figs. 1–3, the secondary peaks are most pronounced in the distributions of plutonium in the waterlogged soil. The appearance of the secondary peaks on the deep slope of the distribution also indicates the migration ability of radionuclides in the soil. This possibility arises during the decomposition of the organic substances and organic forms of the radionuclides due to the microbiological activity in soils when the soil was flooded with lake water. These processes are followed by the decomposition of other potentially unstable carbonate and oxide physicochemical forms of radionuclides and the release of radionuclides to the pore water. The black colour of the waterlogged soil samples indicates the possible presence of FeS compounds – typical compounds in the anaerobic soil regime. Comparing the mobility of radiocesium and plutonium in terms of possible sizes of the secondary peaks in Fig. 3 (soil core near Lake Bedugnis), we can conclude that radiocesium is more mobile in the non-boggy soil than plutonium.

For ^{239,240}Pu, taking into account additional deposits in the surface layer of waterlogged soil due to repeated floods, both peaks in the non-boggy and waterlogged soils near Lake Bedugnis coincide well (Fig. 3(a)). The vertical distribution of radionuclides showed an exponential decrease in the ^{239,240}Pu and ^{137}Cs activity concentration with depth (Figs. 1–3). These results agree with those of Koarashi et al. 2017 [43].

It is clearly visible (Figs. 1–3) that the ^{239,240}Pu and ^{137}Cs activity concentration in the waterlogged and non-boggy soil cores taken at the shores of Lake Juodis, Lake Paršelis and Lake Bedugnis noticeably differ. Thus, ^{137}Cs activity concentrations in the waterlogged soil core taken in the vicinity of Lake Juodis are much larger than those in the non-boggy soil. For Lake Paršelis, they are slightly larger than those in the non-boggy soil ones. And vice versa, in the non-boggy soil core taken near Lake Bedugnis, the activity concentrations of both radionuclides were noticeably larger than those in the waterlogged one. There is nothing unexpected in these results. These differences may be mainly due to different densities of the soil layer content. Also, the radioactive load of waterlogged soil depends on the conditions of deposition of the radioactive fallout on its surface. If the surface of such soil is covered with a layer of water for most time of the year, or, as it was in the case of the Chernobyl NPP accident – during the spring flood, then radionuclides fall accordingly to the water surface, which is usually covered by a film of surfactants. In this case, part of the radionuclides is removed from this surface due to the mechanism of surface self-cleaning [44]. Radionuclides that penetrate directly into the water are distributed depending on the stratification of the aquatic environment in significant water volumes. At the same time, their concentration in the water layer above the soil surface decreases, which leads to a decrease in the soil radioactive load. However, due to the water surface self-cleaning mechanism, in some sites of the lake shore, the so-called 'hot spots' may be created [44]. These are areas with a high concentration of radionuclides. They are usually located in the areas of waterlogged soil near the water edge. Their location on the coast is determined by the direction of dominant winds at the time of the arrival of radioactive air masses, since 'hot spots' are placed where radioactive foam was thrown onto the shore of the water body under the influence of the wind. Thus, on the coast of the same lake, in the areas of waterlogged soil, there may be zones with increased loads of radionuclides.

To compare the ^{137}Cs loads in soil cores from different lakes, the data for Lake Juodis and Lake Paršelis were corrected for the date when the loads of the soil cores from Lake Bedugnis were estimated (by the beginning of 2019). The maximum ^{137}Cs load in the core was estimated in the waterlogged soil sample taken in an old canal in the vicinity of Lake Juodis (2697 Bq/m^2). In the respective non-boggy soil core, the load was lower and amounted to 1078 Bq/m^2. In other cases, the radioactive loads were smallest for Lake Paršelis and for the cores of the waterlogged soil were somewhat lower than those for the non-boggy ones. Thus, the loads in the waterlogged and non-boggy soil cores taken near Lake Paršelis amounted to 1306 and 1585 Bq/m^2, respectively. For the soil cores taken near Lake Bedugnis, they amounted to 1638 and 1811 Bq/m^2, respectively. Plutonium (^{239,240}Pu) loads in the waterlogged soil cores were also less than those in the non-boggy ones. Thus, the loads in the waterlogged soil cores taken near Lakes Juodis, Lake Bedugnis and Lake Paršelis amounted to 89, 57 and 18 Bq/m^2, respectively. The load in the non-boggy soil core taken near Lake Bedugnis amounted to 72 Bq/m^2. The load in the non-boggy core taken near Lake Juodis was measured down to the 22-cm depth and amounted to 47 Bq/m^2. Also, in the respective core taken near Lake Paršelis, the load was measured down to the 15-cm depth and was almost 8 Bq/m^2.

3.2. Distribution of organic matter in waterlogged and non-boggy soil cores

The uppermost layers (1–5 cm) of the waterlogged soil and non-boggy soil cores were rich in organic matter. The soil surface layers in the waterlogged shores of lakes Juodis and Paršelis contained leaves, needles and some moss and their organic matter content was highest (94.2 and 93.6%, respectively) (Fig. 4). The uppermost layer of the waterlogged soil core near Lake Bedugnis contained only needles and some moss and its organic matter content was ~66.0%. The deeper layers of the non-boggy soil samples taken near lakes Juodis, Paršelis and Bedugnis were composed mainly of sand and their organic content was rather small.

It is clearly visible from Fig. 4 that the difference in organic matter content between the waterlogged and non-boggy soil cores is significant. The waterlogged soil core taken near Lake Juodis differed from the other samples having an elevated amount of organics in soil surface layers. The soil from Lake Juodis had an approximately uniform distribution of organic matter in depth where it varied in a quite narrow range of 85–76%. This core differed from others because it was sampled in a dry swamp of the old canal.

Suggesting that the studied radionuclides (^{137}Cs and ^{239,240}Pu) have an affinity to organic matter, the correlation relationship between these radionuclides and organic matter was assessed. However, no consistent pattern of correlations could be established in our case. A statistically significant (p-value <0.05) linear correlation for ^{239,240}Pu activity concentrations with organic matter was obtained (Table 1) for two waterlogged soil profiles of the cores taken near Lake Paršelis (r = 0.89) and Lake Bedugnis (r = 0.6) (Table 1). Small correlation coefficients (r = 0.02 for ^{137}Cs) were obtained for the waterlogged soil profile near Lake Juodis and for two non-boggy soil profiles of the cores taken near Lake Paršelis (r = –0.19 for ^{239,240}Pu) and Lake Bedugnis (r = 0.48 for ^{239,240}Pu). For ^{137}Cs, a significant correlation was obtained for the non-boggy soil core taken near Lake Juodis (r = 0.95), Lake Paršelis (r = 0.77) and Lake Bedugnis (r = 0.83) and two waterlogged ones taken at the shores of Lakes Paršelis (r = 0.88) and Lake Bedugnis (r = 0.7). The corresponding correlation for ^{137}Cs and ^{239,240}Pu in the waterlogged soil core taken near Lake Juodis was insignificant (Table 1). The studies conducted in contaminated soils in Fukushima showed that ^{137}Cs was retained in the top 5 cm of surface soils, with considerably reduced amounts below the 5 cm depth [43]. Soil organic matter also has the ability to retain ^{137}Cs [45]. Several studies have shown that ^{137}Cs is more easily available to biological systems in organic matter-rich soils [46].

In a vertical profile in the forest soil from southern Germany, more than 72% of the ^{239,240}Pu, originating from nuclear weapon tests, was concentrated in the litter/humus (decomposed organic matter in soils) layer and the uppermost soil layer. The sequential extraction revealed that 81% of that Pu was in an organically bound form [47]. Thus, the vertical distribution of Pu can be attributed to the affinity of plutonium for natural organic matter. In the Chernobyl forest, a large portion of plutonium was also found in an organic matrix [35]. It means that the greatest influence on Pu sorption is due to organic matter. The fallout of Pu isotopes investigated in the surface soil samples taken from north western Italy [48–50] showed that the content of organic matter is the main factor influencing the Pu activity concentration, while other soil parameters are of lesser significance. Americium and Pu have higher affinity for organic matter than Cs, which enhances their retention in organic-rich soil. Pu is also known to bind strongly to organic matter in soils [34]. Humic substances and their components (fulvic and humic acids, and some others) which cover various individual compounds, have a large number of functional groups that might bind some kind of trace elements and radionuclides. Therefore, humic substances have been recognized as decisive materials in affecting plutonium behaviour in the environment [35]. The complexation may influence the mobility of fallout radionuclides. Hence, investigating the association of fallout radionuclides with organic acids, it may be possible to interpret the mobility of fallout radionuclides in soils [37].

In order to check the validity of the correlation between radionuclides and the content of organic matter in the studied soils, the physicochemical forms of radionuclides were determined in the sixth layer of the waterlogged soil core sampled near Lake Bedugnis. This is a layer with a high organic matter content (86.8%) and sufficiently high activity concentrations of radionuclides (18.6 Bq/kg for ^{137}Cs and 0.89 Bq/kg for ^{239,240}Pu). So, it turned out that in this layer the fraction of plutonium associated with organic matter was 49.5% while 28.5% of the radionuclide was associated with the residual fraction. These results confirm a great potential for plutonium migration in this soil. As for radiocesium, the results of this analysis were unexpected – almost 99.3% of radiocesium was in its residual fraction. Apparently, this was caused by the presence of an admixture of clay materials in the soil sample. At the same time,

Table 1. Correlation coefficients (r) between ^{137}Cs and ^{239,240}Pu activity concentrations and the organic matter content in the non-boggy and waterlogged soil profiles for different soil core sampling sites.

Name of the sampling site	Soil type	r of ^{137}Cs	p-value	r of ^{239,240}Pu	p-value
Lake Juodis	Waterlogged	0.02	0.92	0.47	0.01
	Non-boggy	0.95	0.0001	0.93	0.0001
Lake Paršelis	Waterlogged	0.88	0.0001	0.89	0.0001
	Non-boggy	0.77	0.0005	–0.19	0.47
Lake Bedugnis	Waterlogged	0.7	0.0001	0.6	0.001
	Non-boggy	0.83	0.0001	0.48	0.01

the organic fraction of radiocesium was practically absent. The waterlogged soil of the sample was acidic (pH 4.28); therefore, the fraction associated with carbonates was also absent. Similar results in a waterlogged soil sample may also indicate the presence of anaerobic conditions in the soil where a stagnant pore water regime is dominant. These data indicate the limited migration possibilities of radiocesium in the presence of clay substances in the anaerobic waterlogged soil. It can also be concluded that the distribution of radiocesium by depth in the waterlogged soil near Lake Bedugnis is stationary. It follows from these data that the above-described correlations of radiocesium with the content of organic matter in the sample, as well as other similar results described in the literature, should be treated with caution. Objectively, data on such correlations can be obtained only when determining the physicochemical forms of radionuclides in the soil, as well as on the basis of knowledge about the composition of the soil and its oxidation regime.

3.3. Mean ratios (^{238}Pu/^{239,240}Pu and ^{137}Cs/^{239,240}Pu) of the radionuclide activity concentrations in the vertical profiles of the studied waterlogged and non-boggy soil cores

The radionuclide activity ratios in soil vary with the contamination sources and can be utilized for their identification. The mean concentration ratios of ^{238}Pu and ^{137}Cs with respect to ^{239,240}Pu in the waterlogged and non-boggy soils are shown in Table 2 and Fig. 5, respectively. To date, many approaches have been proposed for identifying dispersion sources of plutonium isotopes.

Thus, the results shown in Table 2 indicate that the ^{238}Pu/^{239,240}Pu ratios in all analyzed soils are on an average 0.04, which is consistent with the global fallout ratio 0.04 [4, 6]. The highly refractory oxides of plutonium formed at very high temperatures of weapon explosions remained mostly in the environment as solid particles for very long periods of time [12].

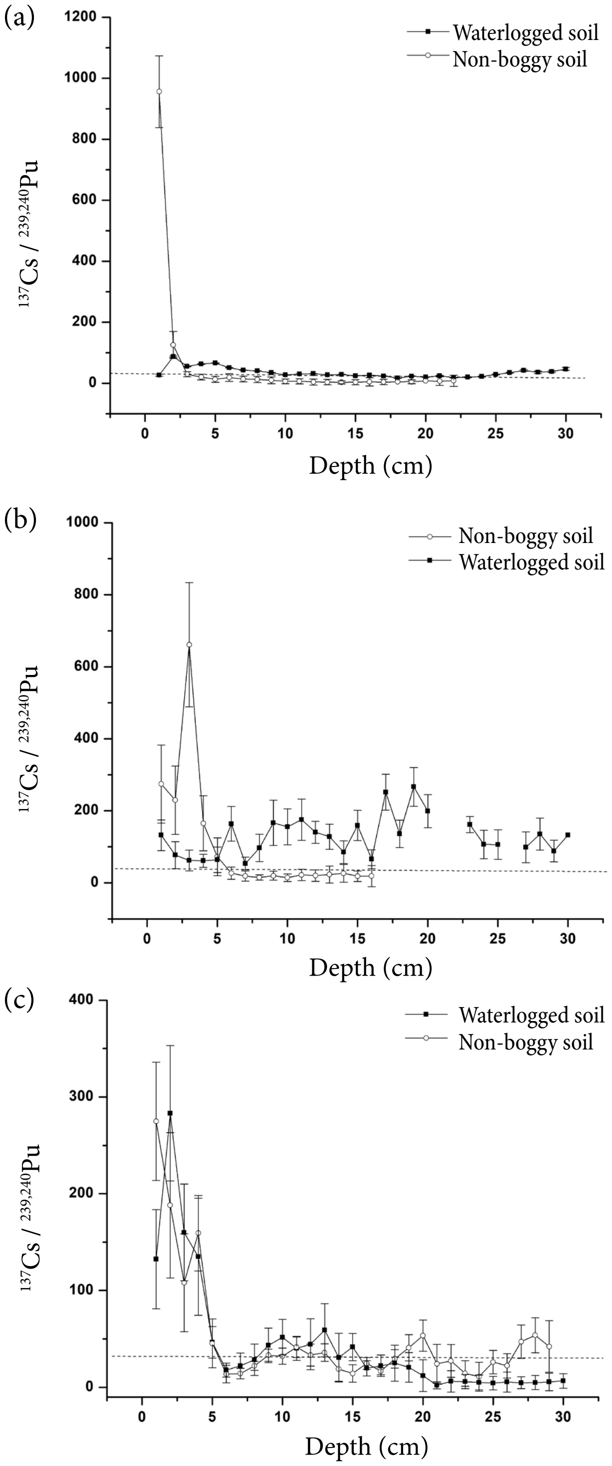

Fig. 5. Vertical profiles of the ^{137}Cs and ^{239,240}Pu activity concentration ratios in the waterlogged and non-boggy soil cores taken near Lake Juodis (a), Lake Paršelis (b) and Lake Bedugnis (c) (the dotted line indicates the ratio of global fallout ~28).

Table 2. ^{238}Pu/^{239,240}Pu activity concentration in the waterlogged and non-boggy soil cores.

	Lake Juodis		Lake Paršelis		Lake Bedugnis	
	Waterlogged	Non-boggy	Waterlogged	Non-boggy	Waterlogged	Non-boggy
^{238}Pu/^{239,240}Pu	0.04	0.03	0.03	0.05	0.06	0.03

It means that the appearance of radioactive air masses related to the Chernobyl accident over the studied area was not followed by new plutonium deposits. This result was very helpful in identifying the origin of radiocesium in vertical profiles of the waterlogged and non-boggy soils. The vertical profile of the radiocesium to plutonium activity concentration ratio in the non-boggy soil core sampled near Lake Juodis showed that mainly the surface layers down to the 3 cm depth were affected by the Chernobyl NPP deposits. For the waterlogged soil core, the presence of Chernobyl NPP radiocesium is traced throughout the entire depth of the vertical profile (Fig. 5). Since the current value of the ratio of activity concentration of radiocaesium to plutonium is estimated as 28, the ratios exceeding this value indicate the presence of Chernobyl NPP radiocesium. The presence of Chernobyl NPP radiocesium in deep soil layers in a sample taken near Lake Juodis indicates its increased migration. It is easy to see from Fig. 5 that enhanced radiocesium mobility is also characteristic of the non-boggy soils of the shores of Lake Bedugnis and the waterlogged soils of the shores of Lake Paršelis.

Thus, the vertical profile of the radiocesium to plutonium ratio in the waterlogged soil core sampled near Lake Bedugnis showed that the soil layers down to a depth of 15 cm are exposed to Chernobyl NPP radiocesium (Fig. 5). In the non-boggy soil core taken near Lake Paršelis, only the surface layers down to the 5 cm depth were affected by the Chernobyl NPP deposits.

Thus, the present data on plutonium and radiocesium vertical profiles in the soil core also imply that deeper-lying of radionuclide activities are related to the mobile fractions of the radionuclides in soils.

4. Conclusions

Investigations showed that the radioactive load of waterlogged soils on the shores of lakes may exceed or be lower than that of non-boggy soils. Its value depends mainly on the conditions of deposition of radioactive fallout on the surface of lake shores, such as the presence of a water layer on the surface of the soil, the direction of the prevailing winds at the time of deposition and the possibility of formation of the so-called ‘hot spots’ on the shore. The vertical profiles of ^{238}Pu/^{239,240}Pu activity ratios evidently showed that in the studied sampling sites near Lake Juodis, Lake Paršelis and Lake Bedugnis most plutonium deposits were due to global fallout. The vertical profiles of radionuclides in the studied soils showed that their distributions consisted of one to several pronounced peaks. The location of secondary peaks in accordance with their depth in soil and their inventory can be attributed to more or less high radionuclide mobility. So, radionuclides in waterlogged soil (swampy area) near Lake Juodis have the maximum mobility. According to the vertical profiles of ^{137}Cs/^{239,240}Pu activity concentration ratios in the studied soil cores, radiocesium of the Chernobyl NPP origin showed its elevated migration abilities in the waterlogged soils of the shores of Lakes Juodis and Lake Paršelis and the non-boggy soils near Lake Bedugnis. Conversely, the migration abilities of radiocesium in the waterlogged soils near Lake Bedugnis were significantly limited by the presence of clay impurities. This was facilitated by the formation of stagnant conditions in the soil pore water. Clay impurities in the waterlogged soil did not affect the mobility of plutonium. In the non-boggy soil near Lake Bedugnis, plutonium mobility is somewhat reduced. So it can be concluded that in the soils near Lake Bedugnis, the mobility capabilities of plutonium and radiocesium are of an opposite pattern.

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^{137}Cs ir ^{239,240}Pu AKTYVUMO KONCENTRACIJŲ PASISKIRSTYMAS SAUSAME IR UŽLIEJAMAME LIETUVOS DIRVOŽEMYJE

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Santrauka

Ištirtas vertikalusis ^{239,240}Pu ir ^{137}Cs aktyvumo koncentracijų pasiskirstymas sauso ir užliejamo dirvožemio kolonėlėse, paimtose netoli Vilniaus miesto ir Varėnos rajone. Tiriamieji radionuklidai yra technogeninės kilmės ir dirvožemyje atsirado dėl radioaktyviųjų iškritų po branduolinio ginklo bandymų atmosferoje šeštajame dešimtmetyje ir po Černobylio atominės elektrinės avarijos 1986 metais. Iškritę radionuklidai dalyvavo įvairiuose persiskirstymo ir kaupimosi procesuose, todėl erdvinis jų pradinio aktyvumo tankio (radioaktyviosios apkrovos) pasiskirstymas yra netolygus. Vadinasi, užliejamo dirvožemio radioaktyvioji apkrova gali būti tiek didesnė, tiek ir mažesnė nei sauso dirvožemio.

Didžiausias plutonio mobilumas nustatytas užliejamose, daug organinių medžiagų turinčiuose dirvožemiuose. Molio priemaišos užliejamame dirvožemyje stipriai apriboja radiocezio migraciją ir neturi pastebimos įtakos plutonio judrumui. Lyginant su radioceziu, plutonio mobilumas sausame dirvožemyje yra šiek tiek mažesnis. Taigi, sausame ir užliejamame dirvožemiuose radionuklidus mobilizuojantys veiksniai priešingai veikia radiocezį ir plutonį.

Mūsų atveju koreliacinė analizė nerodo vienareikšmės tiesinės priklausomybės tarp radionuklidų aktyvumo koncentracijos ir dirvožemio organinių medžiagų kiekio. Tačiau tyrimas demonstruoja, kad radionuklidų judrumą galima įvertinti tiriant radionuklidų fizines ir chemines formas dirvožemyje, vertikaliuosius radiocezio ir plutonio aktyvumo santykių profilius, dirvožemio sudėtį ir oksidacijos režimą.