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Investigation of ^{137}Cs and Pu isotopes accumulation in soil

SUMMARY OF DOCTORAL DISSERTATION

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^{137}Cs ir Pu izotopų akumuliacijos dirvožemyje tyrimas

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SANTRAUKA

1. INTRODUCTION

^{137}Cs and plutonium isotopes are anthropogenic radionuclides of great concern due to their prevalence in the environment after nuclear weapons tests and nuclear accidents at the Chernobyl and Fukushima NPPs. The processes of fixation of radionuclides in soil play an important role there, since they determine the further transfer and bioavailability of the deposited radionuclides. The property of soil to sorb and accumulate anthropogenic radionuclides has been confirmed by many scientific studies, which have indicated a significant retention of anthropogenic radionuclides mainly in the upper layers of soil [1 – 11]. The downward migration of anthropogenic radionuclides in soil depends on a number of factors, such as the initial physical and chemical form of the radionuclides [12] as well as on the climate and the ecosystem's characteristics. In particular, precipitation significantly controls the deposition and then the migration of radionuclides into deeper soil layers. Soil characteristics, such as pH, organic matter and clay content also significantly affect the migration of radionuclides [13]. The natural phenomenon such as soil waterlogging due to floods or snowmelt strongly affects all components of soil as well as influences soil formation and radionuclide migration processes [14]. Studying the migratory behaviour of radionuclides in waterlogged soils is important for a better understanding of the mechanisms of natural self-cleaning in contaminated areas [15]. Since waterlogging can create an anaerobic and reducing environment in soil, factors that have a major influence on seasonally or permanently waterlogged soil include oxidation–reduction, dissolution–precipitation and adsorption–desorption processes [16].

A gap in the available data on the dependence of sorption on various environmental factors (such as pH, temperature, organic matter content and competing ions) for many radionuclides and

especially for plutonium still exists. This is especially true for quantifying the solid-liquid distribution coefficient (K_d) and its dependency on the soil type and its characteristics [17], despite most of the models for the prediction of radionuclide transport in the environment being based on K_d concepts. The kinetics of plutonium sorption by soils is influenced mainly by soil properties, and their complex mechanisms are still not fully understood. In particular, reliable data on the migration ability of Pu, which can have different oxidation states under different conditions or types of soils, are scarce due to the complexity of the process and the relatively high costs of the experiments.

In most cases, the mechanism of sorption of radionuclides in soil is an ion-exchange or the reaction of surface complexation, depending on some physicochemical properties of the soil: soil type, pH, redox conditions, composition of organic compounds, amount of trace elements (Fe, Mn, Al, Ca) and their valence. A radionuclide sorbed in soil usually cannot exist in a permanently fixed form. The pH is a decisive factor for the sorption behaviour of plutonium due to its speciation or formation of various complexes, especially with organic molecules, or association with colloids [18]. However, when talking about ion exchange, pH is not the dominant factor in terms of sorption mechanisms. The pH is significant for surface complexation, which controls the sorption process. Ion exchange doesn't depend on the sorption capacity, but on the sorption strength and on water complexation. The sorption strength depends on factors such as ionic charge and ionic size, while water complexation depends on the nature of the complexing agent (ligands) as well as on the properties of cations [19].

In seasonally waterlogged soil, the alternating dry and wet conditions promote leaching and accumulation of metals in the soil profile [20].

Thus, among all other factors (pH, surface charge, soil particle size, organic matter and clay mineral content, etc.), the hydrological regime of the soil is also crucial for the migration of radionuclides in

the soil. Therefore, the influence of these main factors on the migration and accumulation of ^{137}Cs and $^{239,240}\text{Pu}$ radionuclides in non-boggy and periodically flooded natural soils was further investigated in my dissertation.

The aim of the doctoral dissertation is to establish the factors that determine the sorption/desorption and vertical transport of ^{137}Cs and Pu isotopes in soils of different types and different moisture regime (waterlogged soils due to periodical flooding and the moisture regime of non-boggy soils).

Research objectives:

1. To experimentally evaluate the sorption capacity for plutonium in different types of soil with different particle sizes, content of organic matter and of stable elements.
2. To apply in practice the sequential extraction method, estimating the mobility of Cs^+ and Pu isotopes in relatively non-boggy and waterlogged soils in a part of the exchangeable fraction.
3. To analyse impact of the factors determining the sorption/desorption of Cs^+ and Pu isotopes on the soil of different hydrological regimes.
4. To analyse the background level of $^{238,239,240}\text{Pu}$ and ^{137}Cs contamination in potentially important Lithuanian locations before the activation of the Belarusian Astravets NPP and to evaluate the radionuclide diffusion and accumulation patterns in the soil of various locations.

Scientific novelty and practical importance: the novelty of this work lies in the fact that it comprehensively studies the factors that determine the dependence of the values of the Pu distribution coefficient K_d on soil parameters (content of organic matter, mineral composition, particle size, pH, filtration rate, content of micro- and

macroelements) in different types of natural Lithuanian soils. Record correlations were observed between the K_d value of the plutonium distribution coefficient and the amount of organic matter in the soil.

For the first time, the factors determining the migration of ^{137}Cs and plutonium isotopes in non-boggy and waterlogged soils are described. After the practical application of the sequential extraction method, it was found that the increased mobility of $^{137}\text{Cs}^+$ ions in non-boggy soil is determined by the positive surface charge of soil particles, low content of organic matter and the acidic pH of the soil. In waterlogged soil, $^{137}\text{Cs}^+$ ions are immobile due to the negative surface charge of soil particles. In this case, plutonium ions in non-boggy soil have low mobility due to the formation of compounds with Fe and Mn (hydr)oxides, and in the waterlogged soil, Pu is mobile due to the formation of soluble compounds with fulvic and humic acids, that are soluble in water in the conditions of the studied soil.

Statements for defence:

1. The migration of Pu in the soil is determined by the size of soil particles and the amount of organic matter therein. The order of plutonium sorption capacity in different soil types was determined as follows: peat > clayey sand > clayey loam > silty sand > sand.
2. Cs^+ is more mobile in non-boggy soils than in waterlogged soils due to the predominant positive surface charge of the soil particles.
3. Pu isotopes are more mobile under waterlogged soil conditions due to the formation of soluble compounds with organic matter in the soil.
4. The bottom sediments of the Curonian Lagoon and on the coast of Nida as well as floodplain soil near Krokų Lanka and near the village of Plaušvariai are potential zones for increased long-term accumulation of radionuclides (Cs and Pu) in Lithuania due to potential contamination from nuclear power plants located in Lithuania's neighbours.

2. CONTENT OF THE DISSERTATION

The introduction presents the relevance of the study, defines the purpose and objectives of the research, outlines the defended statements, and discusses the scientific novelty as well as practical significance of the doctoral dissertation.

The literature review contains general information on the properties of ^{137}Cs and $^{239,240}\text{Pu}$, their behaviour in soil and the possibilities of their migration in soil. The vertical transport of radionuclides in the soil is described. The components of the soil that accumulate radionuclides, as well as the surface charges of soil particles, are described.

The methodology and object of research section describes the reagents and solutions that were used, as well as the equipment and methodology of experimental studies (sample processing, chemical separation and purification, ion exchange chromatography, sequential extraction and flow-through-column method). The soil is considered as the main object of study. Sampling points are described.

Samples of bottom sediments (Neris River, Nemunas River and the Curonian Lagoon) and floodplain soil (Nemunas River) were collected to identify potential radionuclide accumulation zones (Table 1).

Table 1 Description of the sampling sites located in the Neris River, the Nemunas River, the Curonian Lagoon and in the floodplain of the Nemunas River.

Sampling site	Coordinates	Locality
The Neris River	54°50.336',25°44.471'	At the Vilnius city
	54°42.535',25°18.467'	Near Buivydžiai village
The Middle Nemunas River	54°4.896',24°3.367'	Near Liškiava village
	54°47.867',24°12.150'	Near Barevičiai vilage
	54°43.983',24°7.133'	Near Darsūniškis village
The Lower Nemunas River	55°12.000',21°33.441'	Near Šilininkai village
	55°17.720',21°23.122'	Near the Rusnė Island
	55°21.857',21°19.825'	The central part of the Lake Kroku Lanka (lake in the Nemunas Delta)
	55°18.967',21°18.580'	The bay on the Pakalnė River (branch of the Nemunas river in a delta)
	55°19.169',21°17.600'	The Lower Pakalnė River
The Curonian Lagoon (territory of Lithuania)	55°21.422',21°14.970'	Kniaupo bay near Mingė village (South-East part of the lagoon)
	55°18.655',21°13.688'	The South-East part of the lagoon (Nemunas River avandelta)
	55°16.861',21°00.836'	The West part of the Curonian lagoon near Nida town
Floodplain of the Nemunas River	55°05.341',21°53.803'	Near Panemunė village
	55°06.074',21°50.800'	Near Plaušvariai village
	55°07.448',21°45.904'	Near the Lake Sennemunė
	55°12.334',21°35.255'	Near the River Veižas
	55°11.713',21°31.929'	Near the Lake Ilgasis
	55°17.137',21°24.582'	Near the Leitė River
	55°17.951',21°23.264'	Near the bridge to the Rusnė Island
	55°21.764',21°21.259'	Near the Lake Kroku Lanka

For experiments on plutonium sorption and determination of K_d , samples of five different types of soils were collected in different locations in Lithuania: in four sites in the Varėna region: BLZ (N54°11'05", E24°10'32" - sand, BDG (N54°11'12", E24°09'14") - silty sand, IGB (N54°11'12", E24°10'24") - peat, MLS (N54°12'17", E24°07'09") - clayey sand and one sample was taken in Vilnius: VLN (N54°34'54" E25°10'56") - clayey loam.

The waterlogged and non-boggy soil cores were taken in different locations in Lithuania from the shores of three lakes (Lake Juodis, Lake Paršelis, and Lake Bedugnis). All sampling points were selected on the banks of lakes with undisturbed soils that have not been cultivated for many years. In these lakes, the increased water level due to repeated rains and snowmelt every year leads to the flooding of nearby shores. This flooded soil is considered to be waterlogged soil. Lake Juodis (N54°46'54", E25°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. A soil sample from that location was taken in 2012. A core of non-boggy soil was taken from the western shore of the lake. A core of waterlogged soil was taken in the vicinity of the lake in an old channel, which was periodically flooded in spring.

Lake Bedugnis (N54°11'12", E24°09'14") is located in the Varėna region (the southern part of Lithuania). The lake is stagnant and belongs to the Kempė river basin. A soil sample was taken in 2018.

Lake Paršelis (N54°05'0", E24°41'45") is located in a depression in the Varėna region. A soil sample was taken in 2015.

Soil samples were taken by stabbing a steel tube 30 cm in height and 12.5 cm in diameter. 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.

Experimental technique:

The concentrations of micro- and macroelements (Cu, Mn, Co, Na, Fe, Ca, Mg, Pb, K) in the soil samples were determined using an AAnalyst 800 PerkinElmer atomic absorption spectrometer (AAS). Soil samples were dried (24 h, 105°C) and then mineralized at a temperature of 400°C. 1 g of the soil was weighed and digested with a mixture of concentrated HCl and concentrated HNO₃ (3:1) for 1 h. Subsequently, it was evaporated to dry salt. Then, 10 ml of concentrated HCl was poured onto it and again evaporated to dry salt. The obtained resulting salt was dissolved in distilled water and the solution was filtered. The prepared samples were measured using AAS.

The soil samples were analysed for ¹³⁷Cs using a CANBERRA gamma-spectrometric system with the HPGe detector (model GC2520, 26.2 % relative efficiency, resolution of 1.76 keV/1.33 MeV) according to the gamma line at 661.62 keV of ¹³⁷Ba (a daughter product of ¹³⁷Cs). Measurements were carried out in standard geometry and calibrated at known efficiencies according to densities of samples. Measurement errors of radiocesium activity concentrations in samples were evaluated by the GENIE software tool. They were less than 5 % (standard deviation) for active samples and not larger than 20 % for less active layers of soil cores. When measuring very weak radiocesium activity of the samples of its physicochemical fractions, an error of the order of 20 % was achieved during long-term measurements of up to 2,000,000 seconds.

The organic matter content was determined by the loss on ignition method. Soil samples were burnt in a muffle furnace at 500°C overnight, and the next day the samples were burnt at 700°C for 2 h to decompose organic matter.

The determination of the zero charge point of the studied soils by the salt addition method [21] was carried out by adding equal amounts of waterlogged and non-boggy soils to a set of solutions with the same ionic strength at different pH values. In a series of

50 ml flasks, 0.2 g of soil was added to 40.0 ml of 0.1 mol/l KNO₃ solution. The pH was adjusted with 0.1 mol/l HNO₃ and 0.1 mol/l NaOH as needed to obtain a suitable pH range of 2, 4, 6, 8, 10, and 12 (± 0.1 pH units). The pH values of the supernatant in each flask were designated as pH_i (initial). The samples were shaken for 24 h using a shaker (GFL 3006) at 150 rpm. After settling, the pH of the supernatant in each flask was measured and referred to as pH_f (final). The pH_{PZC} was obtained from a plot of ΔpH ($=\text{pH}_f - \text{pH}_i$) versus pH_i.

Evaluation of the Pu isotope activity concentrations in soil samples requires special radiochemical separation procedures. Plutonium was pre-concentrated and purified using DOWEX 1×8 (NO₃⁻) ion exchange resin, then the samples were processed and electrochemically deposited on stainless steel disks and measured by alpha spectrometry [22]. The specific activity of plutonium was measured with an Octet Plus semiconductor alpha spectrometer equipped with large area (450 mm²) ORTEC Si detectors (BU-020-450-AS) with a resolution of 20 keV. The spectra were analysed using the Alpha Vision and Maestro software [22].

The value of the distribution coefficient K_d (l/kg) was calculated using the formula [23]:

$$K_d = \frac{(C_i - C_e)}{C_e} \cdot \frac{W_1}{W_s} \quad (1)$$

where C_i is the initial radionuclide concentration of the radionuclide in the solution (Bq/l), C_e is the equilibrium concentration of the radionuclide in the liquid phase of the filtrate (eluate) (Bq/l), W₁ is the volume of the solution (l) and W_s is the weight of dry soil (kg).

The sequential extraction of ^{239,240}Pu isotopes, ¹³⁷Cs and stable elements (Ca, Mn, Fe, Al) from soil samples was performed according to Tessier et al., (1979) [24] with minor modifications by Kimber et al., (2015) [25] (Table 2). Typically, a soil sample is subjected to a series of chemical treatments designed to affect a

unique biochemical phase (compound) of the sample. At each step, the sample and reagent are usually shaken or agitated (at a specified temperature) in some way for a specified period of time. After this reaction, the solid and aqueous phases are separated by filtration. Residual sediment is reserved for the next reaction step in the sequence and the fluid phase is analysed for the analyte of interest.

For the analysis, we used 20 g of samples from each of the non-boggy and waterlogged soil layers at a depth of 6 - 8 cm, where the activity concentrations of radionuclides are maximums. A ^{242}Pu tracer (0.12 Bq/sample) used to monitor the chemical yield. After separation of each fraction, samples were washed with 100 ml of distilled water.

Table 2 Fractions, reagents and experimental conditions (temperature and extraction duration).

Sequential extraction fraction	Extractors	Temperature (°C)	Time (h)
Exchangeable	200 ml 1 mol/l $\text{CH}_3\text{COONH}_4$ pH 7 shake	Room	1
Bound to carbonates	200 ml 1 mol/l CH_3COONa pH 5 shake	Room	2
Bound to oxides (Fe/Mn)	200 ml 0.04 mol/l $\text{NH}_2\text{OH}\cdot\text{HCl}$ 25 % v/v CH_3COOH pH 2 heat	96	6
Bound to organic matter	2× (60 ml 0.02 M HNO_3 with 100 ml 30 % H_2O_2 pH 2) 100ml 3.2 mol/l $\text{CH}_3\text{COONH}_4$ v/v 20 % HNO_3 heat	85	2
		85	0.5
Residual fraction	80 ml (3 HCl:1 HNO_3) heat	100	1

After the filtration step, the filters and soil residues again placed in the reaction vessel for the next extraction. After a reaction period, the solid and aqueous phases were separated by filtration, while 5 ml of this filtrate was taken to determine the stable elements. The rest of the filtrate was evaporated and the dry residue was measured on a gamma spectrometer to determine the ^{137}Cs content of the filtrate.

3. RESULTS AND DISCUSSION

3.1. Investigation of potential accumulation sites for $^{239,240}\text{Pu}$ and ^{137}Cs

In the context of the construction of the Belarusian NPP, this study was focused on the search for and selection of potential accumulation (barrier) zones for radionuclides in the Nemunas River basin. Heavy metals and anthropogenic radionuclides comprise an important part of pollutants released into the environment from NPPs. For this reason, not only radionuclide activity and concentrations of Fe, Al, Mn (the compounds which play one of the main roles in radionuclide accumulation and distribution processes) were measured in this study, but also other elements potentially hazardous to the environment (As, Ni, Cr, Cu, Pb). All the determined parameters of the studied soils can be found in [26].

3.1.1. Distribution of ^{137}Cs and Pu concentrations in potential radionuclide accumulation zones

The activity of ^{137}Cs in bottom sediments in the 0 - 5 cm layer of sediments of the Neris River did not exceed 18 Bq/kg, that of the Lower Nemunas River varied from 1 Bq/kg in sediments near Rusnė Island and the Lower Pakalnė River to 12.6 Bq/kg in the central part of the Lake Krokų Lanka, while those collected in the Middle Nemunas in 1995 - 1996 and recalculated in the year 2016 were significantly higher. The ^{137}Cs activity in the Middle Nemunas were

as follows: 33.7 Bq/kg (Near Liškiava village), 47.0 Bq/kg (Near Barevičiai village) and 44.3 Bq/kg (Near Darsūniškis village). ^{137}Cs activity variations in the sediments of the Curonian Lagoon were as follows: 1.6 Bq/kg (The South-East part of the lagoon), 4.1 Bq/kg (Kniaupo bay near Mingė village), and 3.4 Bq/kg (The western part of the Curonian lagoon near the town of Nida).

The $^{239,240}\text{Pu}$ concentration in the sediments of the studied sampling sites varied between 0.016 and 0.34 Bq/kg (see [26] Table 2). The ^{238}Pu activity concentrations in most samples were not determined because they were lower than the detection limit. The level of $^{239,240}\text{Pu}$ activity in the bottom sediment samples in 1995 - 1996 differed insignificantly compared to those in the sediments collected between 2011 and 2012, with the exception of samples from the central part of the Lake Krokų Lanka, the western part of the Curonian lagoon near the town of Nida and near Darsūniškis village, where $^{239,240}\text{Pu}$ activity reached 0.34, 0.21 and 0.18 Bq/kg, (see [26], Table 2) respectively.

The sampling sites from the western part of the Curonian lagoon near the town of Nida, near Liškiava village, near Barevičiai village and near Darsūniškis village tended to accumulate ^{137}Cs and $^{239,240}\text{Pu}$ at higher rates than other sites. The range of ^{137}Cs activity in the sediments of these sampling sites was from 33.4 to 47.0 Bq/kg. The highest values of the $^{239,240}\text{Pu}$ activity were characteristic for the central part of the Lake Krokų Lanka, the western part of the Curonian lagoon near the town of Nida, and near Darsūniškis village, they were 0.34, 0.21 and 0.18 Bq/kg, respectively.

Activity of ^{137}Cs in the tested soils ranged from 5.3 Bq/kg (near the Leitė river) to 32.9 Bq/kg (near the Lake Krokų Lanka) (see [26], Table 3). The activity measured in Near the Lake Krokų Lanka was significantly higher ($p < 0.05$) compared to the activity determined in the other samples. The lowest values of the $^{239,240}\text{Pu}$ activity in soil were determined near Panemunė village and near the Lake Sennemunė (0.086 and 0.064 Bq/kg respectively). The top layer (0 - 5 cm) of near the Lake Krokų Lanka, the soil sample, contained

significantly high $^{239,240}\text{Pu}$ activity (0.55 Bq/kg) (see [26], Table 3). The ^{238}Pu activity in all tested soil samples was lower than the detection limit.

The ^{137}Cs and $^{239,240}\text{Pu}$ activity and their relationship with some parameters in bottom sediments and in floodplain soil can be seen in [26].

Data on ^{137}Cs and $^{239,240}\text{Pu}$ activity in sediments and flooded soil have indicated the following: direct relations of these radionuclides to the total organic carbon; the tight correlation of Fe, Al and Mn with the activity of both radionuclides; and interconnection of both radionuclides. Despite the different chemical and physical properties of ^{137}Cs and plutonium, tendencies of radionuclide accumulation in bottom sediments and flooded soil, depending on grain size composition and total organic carbon content, were of rather similar character. Therefore, ^{137}Cs at the initial investigation stage can be used as the alternative tracer element for plutonium in the bottom sediments and top layer of the flooded soil. It is relevant in order to avoid labour- and time-consuming plutonium radiochemical analysis in the prediction of sites of the plutonium isotope dispersion in those compartments of the ecosystem as well as in searching for possible accumulation zones of this radionuclide.

3.2. Determination of the distribution coefficient K_d in different types of soils

Physical and chemical parameters of the BLZ, BDG, IGB, MLS and VLN soil samples are presents in Tables 3 - 4.

Fractions of particles ≤ 0.25 mm and $0.25 \div 0.5$ mm in size were separated in soils, and each derivative was filled in four columns (40 columns in total). In every case an empty plastic column 30 mm in diameter and 115 mm in height with a permeable bottom was filled with an equal volume of soil up to 80 mm. Depending on the density of the soil, its mass ranged from 0.018 to 0.091 kg. Column natural gravity infiltration tests were performed under atmospheric

conditions to estimate K_d using 4 different tracer solutions with initial pH = 1.6, 4, 6 and 9. The tracer solutions of ^{236}Pu (IV) were prepared from a stock solution of ^{236}Pu in 4 mol/l HNO_3 by mixing it with distilled water. A solution with a suitable pH was passed through a column filled with soil, pre-moistened with 50 ml of distilled water. Then, 50 ml of a previously prepared ^{236}Pu solution was passed through the column, and finally, the column was washed with 10 ml of distilled water. The contact time of the solution and the solid, regardless of particle size, ranged from about 0.5 h for columns filled with sand (BLZ), clayey sand (MLS) and clayey loam (VLN), to 40 h for columns filled with silty sand and peat. The eluates were collected and their pH was measured (Table 5). A ^{242}Pu tracer (0.12 Bq/sample) was used to control the chemical yield both in the soil and the eluate of each column.

Table 3 The content of organic matter, pH and concentrations of stable elements in the studied soils.

Code of the sample and type of soil	Soil pH	Organic matter, %	Concentration of the element, mg/g								
			Cu	Mn	Co	Na	Fe	Ca	Mg	Pb	K
BLZ, sand	9.4	1.7	0.003	0.06	0.006	0.04	0.840	8.50	0.25	0.005	0.26
BDG, silty sand	6.0	1.3	0.006	1.08	0.008	0.04	2.70	0.000	0.25	0.002	0.23
IGB, peat	6.3	66.8	0.001	0.007	0.005	0.03	8.50	5.90	0.38	0.02	0.65
MLS, clayey sand	6.7	11.8	0.046	0.10	0.012	0.10	6.50	4.88	0.25	0.01	0.90
VLN, clayey loam	5.8	3.7	0.003	0.23	0.007	0.05	7.50	0.002	0.99	0.01	0.60

Table 4 Mineralogical composition (%) of the soil samples.

Type of minerals	Soil samples				
	BLZ	BDG	IGB	MLS	VLN
NaAlSi ₃ O ₈ Albite	3.5	6.0	16.0	-	11.0
(Na,K)(Si ₃ Al)O ₈ Anorthoclase	3,5	-	-	-	-
SiO ₂ Quartz	86,0	88.0	12.0	85.0	86.0
CaCO ₃ Calcite	7,0	-	-	-	-
(Mg,Al,Fe) ₆ (Si,Al) ₄ O ₁₀ (OH) ₈ Clinocllore	-	-	-	1.1	-
(K,H ₃ O)Al ₂ Si ₃ AlO ₁₀ (OH) ₂ Illite	-	-	-	3.5	-
KAlSi ₃ O ₈ Microcline	-	5.0	-	7.0	-
(K,Na)(Al,Mg,Fe) ₂ (Si _{3,1} Al _{0,9})O ₁₀ (OH) ₂ Muskovite	-	2.0	-	-	2.9
Ca(Al ₂ Si ₂ O ₈) Anorthite	-	-	-	3.0	-

3.2.1. Sorption dependence on pH of tracer solution

The initial soil pH ranged from 5.8 to 6.7, with the exception of BLZ soil, which had an initial pH of 9.4. The initial pH of the tracer solution percolating through the soil column changed, and the eluate in the cases of IGB, MLS and VLN became almost neutral or remained alkaline in the case of BLZ (Table 5). Only in the case of the BDG soil, the eluate remained strongly acidic (pH = 3.0) when the tracer was added with the initial pH = 1.6. Both soil particle size fractions showed the same pH buffering capacity. The sorption experiment in most cases showed a non-linear dependence of K_d on the initial pH of the tracer. The K_d values did not significantly correlate with the pH, probably due to the high buffering capacity of the soils. The highest values of K_d were usually obtained using a Pu tracer solution with pH = 4 and 6. The buffering capacity of the soil indicates its ability to withstand changes in the pH. The buffering capacity is determined by the number of surface areas that can hold H^+ ions. Based on the obtained data on the influence of pH on the sorption of the tracer, the mechanism of Pu sorption by soils acting in our column experiments can be attributed to the ion-exchange mechanism with the surface complexation type for BDG, IGB, MLS and VLN soil samples (pH was buffered in the range from 3 to 7). Sorption of Pu in a soil sample from BLZ sand, whose pH changed from neutral to alkaline (pH from 7 to 9) after contact with the influx at a pH from the strong acid (pH 1.6) to a strong alkaline (pH 9), can be attributed to the complex type surface reactions due to the strong hydrolysis of Pu [19].

Table 5 The obtained plutonium distribution coefficient K_d for the studied soil particles with a size of ≤ 0.25 mm and $0.25 \div 0.5$ mm at various initial pH values of the tracer.

Code of the sample and type of soil		pH and K_d , l/kg							
		≤ 0.25 mm				$0.25 \div 0.5$ mm			
Applied tracer solution pH		1.6	4	6	9	1.6	4	6	9
BLZ pH 9,4	Eluate pH	7	8	9	9	7	8	9	9
	Sand K_d	291±58	718±57	397±87	82.6±7	4.6±0.2	16.9±1.6	9.3±0.9	14.4±1.1
BDG pH 6.0	Eluate pH	3	5	6	7	3	5	6	7
	Silty sand K_d	54.6±11	208±29	149±16	57.6±3	3.9±0.2	6.4±0.7	7.2±1	10.9±1.3
IGB pH 6.3	Eluate pH	5	6	6	7	5	6	6	7
	Peat K_d	4958±991	6597±589	6200±1240	2878±607	46.6±2.3	49.1±4.9	1994±199	478±67
MLS pH 6.7	Eluate pH	6	7	6	7	6	7	6	7
	Clayey sand K_d	2039±407	3240±712	1607±187	666±28	13.2±0.7	29.4±5.8	481±67	56.2±4.5
VLN pH 5.8	Eluate pH	5	6	6	7	5	6	6	7
	Clayey loam K_d	1155±231	810	739±27	172±18	8.5±0.8	12.7±2.3	45.2±5.4	31.0±2.5

3.2.2. Influence of soil particle size and liquid-solid contact time

Our results confirm the general trend towards a fast plutonium sorption and distribution coefficient for a finer fraction of soil particles ≤ 0.25 mm (Table 5), which indicates an increase in the adsorption capacity of plutonium due to a larger total surface area. The lack of correlation (mean $r = -0.43$) between flow rate and estimated K_d values for Pu for various soil types means that contact time was not a critical factor for sorption compared to chemical characteristics.

Our studies of plutonium sorption were carried out under dynamic flow conditions, when the relatively short sorption time was determined by the liquid flow rate due to natural infiltration, therefore, equilibrium was not reached. However, the results show that the rate of plutonium sorption on different soil types was high (sorption of up to 99 %), which could be facilitated by fast surface-mediated processes. We believe that the behaviour of Pu in the flow-through-column experiment is a true reflection of that in the field.

3.2.3. Influence of soil organic matter

The obtained results suggest that, depending on soil type and pH, the K_d values for plutonium can vary by up to three orders of magnitude. Such variability is caused by the physical and chemical properties of soils, as well as different contact times between the solid and liquid phases. A significantly higher K_d value was obtained for the IGB sample, which contained about 67 % organic matter. This was due to the fact that plutonium has a strong affinity for humic substances, and even a very small amount of soil organic matter can affect the chemical nature and geochemical behaviour of plutonium. Correlation analysis showed a very significant positive highly reliable linear dependence (mean $r = 0.98$, $p < 0.02$) between the K_d

values of Pu and the content of organic matter in different soil types, regressing separately for soil fractions $0.25 \div 0.5$ and ≤ 0.25 mm and tracer pH values. For a more uniform distribution of soil organic matter and K_d values for a whole range of concentrations, the logarithms of the values were analysed (Fig. 1). A very high average correlation $r = 0.96$ (ranged $r = 0.89 \div 0.998$) was also observed on the logarithmic scale.

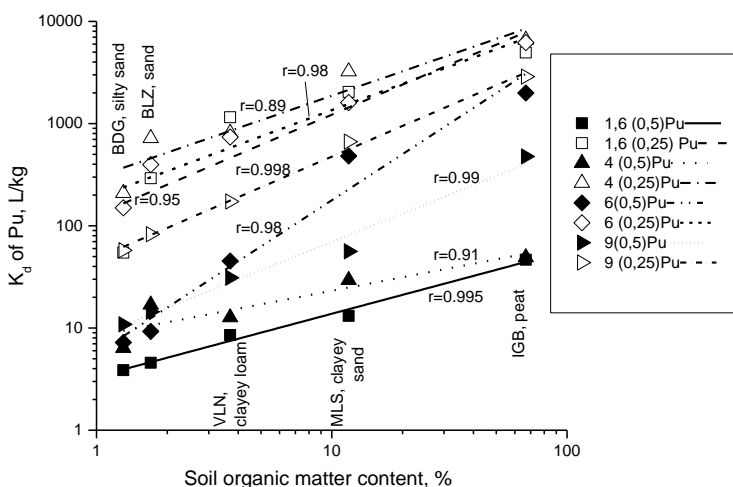


Fig. 1. The dependence of the K_d values for Pu on the content of organic matter in the soil.

3.3. Distribution of ^{137}Cs and $^{239,240}\text{Pu}$ activity concentrations in the studied waterlogged and non-boggy soil cores

Analysis of the vertical profiles of the radionuclide activity concentrations in the non-boggy and waterlogged soil cores showed two more or less pronounced peaks of these concentrations. Although the secondary peaks of ^{137}Cs activity concentration are not clearly visible on the respective profiles of the studied non-boggy

soil cores sampled near Lakes Juodis, Bedugnīs and Paršelis (Figs. 3 - 4).

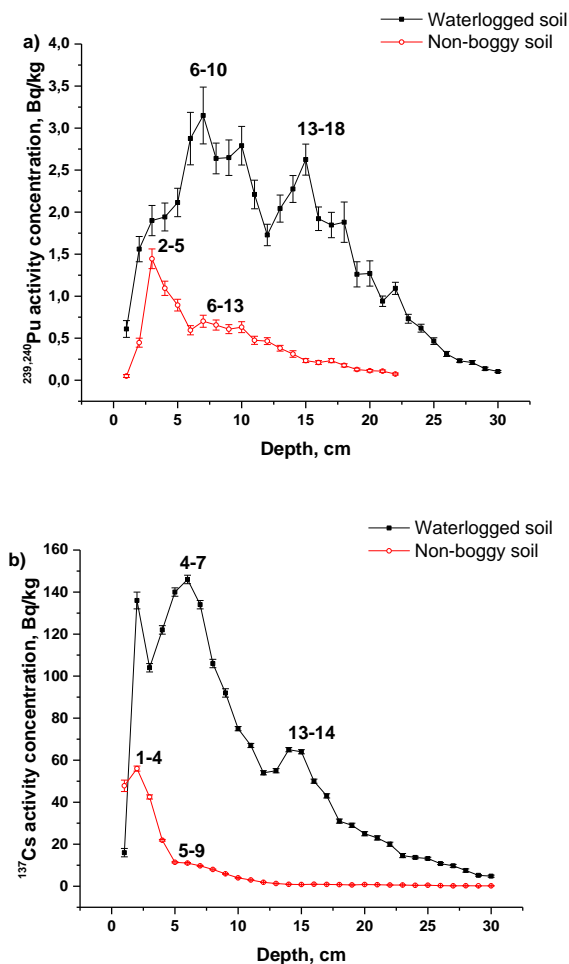


Fig. 2. Vertical profiles of $^{239,240}\text{Pu}$ (a) and ^{137}Cs (b) activity concentrations in the waterlogged and non-boggy soil cores sampled near Lake Juodis. $^{239,240}\text{Pu}$ activity concentrations were measured in a non-boggy soil core down to a depth of 22 cm.

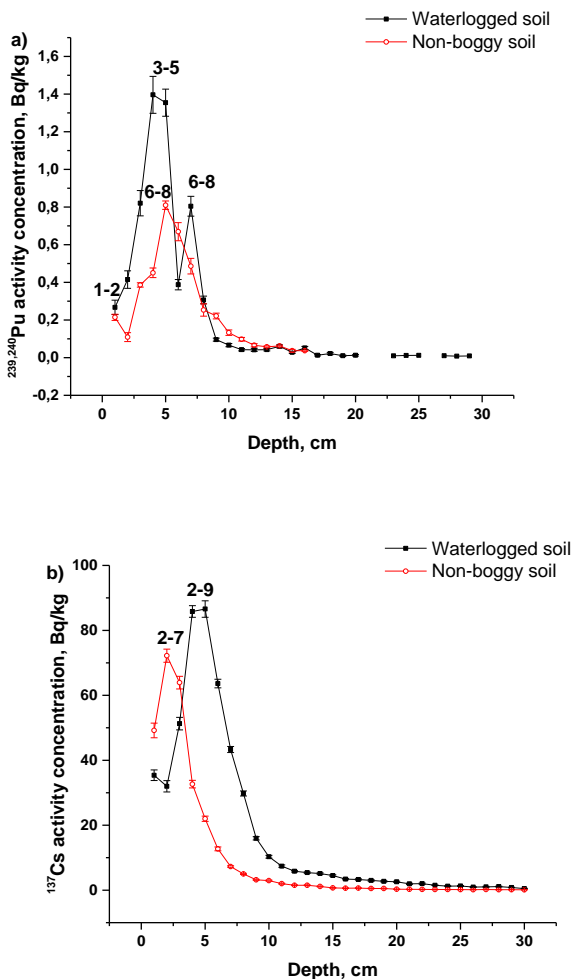


Fig. 3. Vertical profiles of $^{239,240}\text{Pu}$ (a) and ^{137}Cs (b) activity concentrations in the waterlogged and non-boggy soil cores sampled near Lake Paršelis. $^{239,240}\text{Pu}$ activity concentrations were measured in the soil cores down to a depth of 15 cm.

The different depths of $^{239,240}\text{Pu}$ and ^{137}Cs activity concentration peaks on the respective vertical profiles of the waterlogged and non-

boggy soil cores may indicate different migration abilities of radionuclides in these media. Thus, more intense migration of both radionuclides is obvious in the waterlogged soil of the old channel in the vicinity of Lake Juodis (Fig. 2 (a, b)).

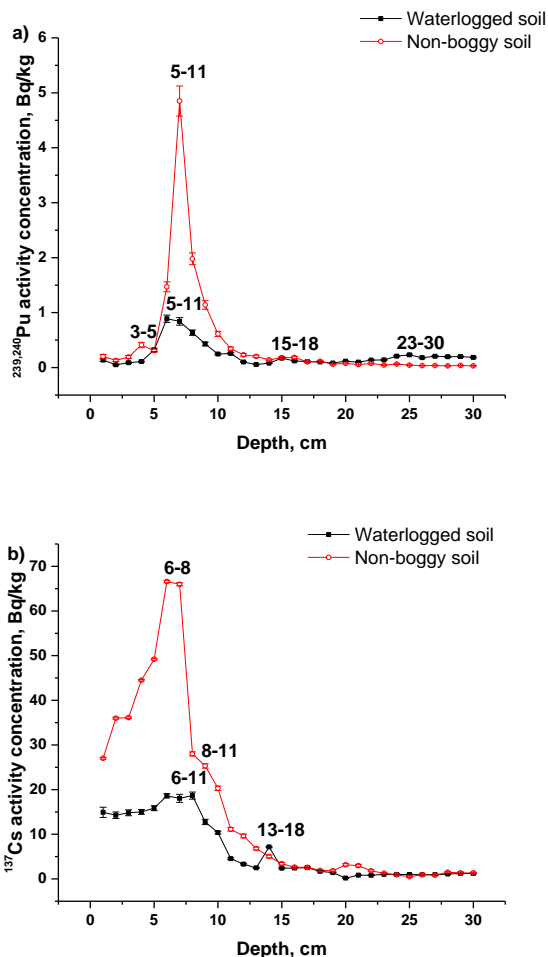


Fig. 4. Vertical profiles of $^{239,240}\text{Pu}$ (a) and ^{137}Cs (b) activity concentrations in the waterlogged and non-boggy soil cores sampled near Lake Bedugnīs.

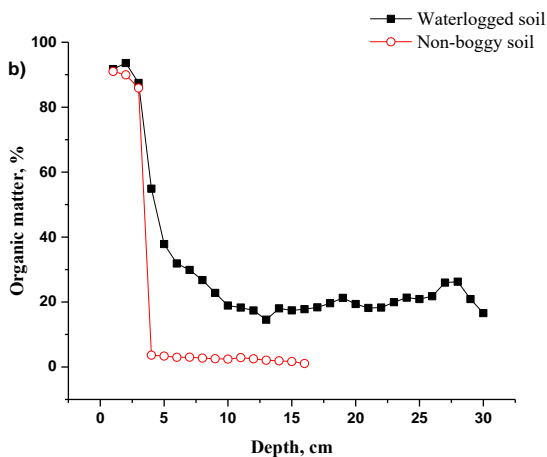
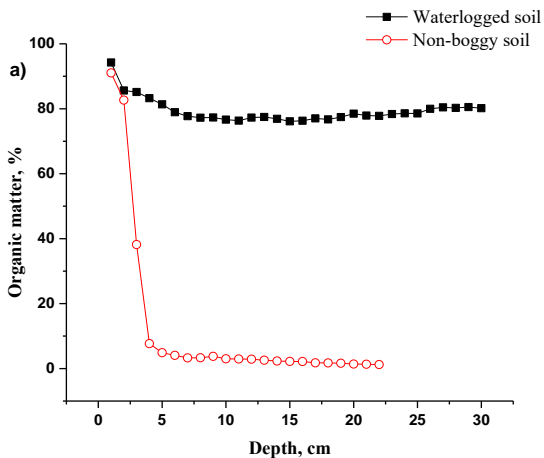
In the absence of adsorbing substances in the soil, radionuclides can migrate deep into the soil, reaching the water table. The same feature can be seen for radiocesium and plutonium in waterlogged soil on the shores of Lake Paršelis (Fig. 3 (a, b)). In the waterlogged soil core taken near Lake Bedugnis (Fig. 4 (b)), the radiocesium peak is located deeper than that in non-boggy soil. It can also be considered as evidence of the higher mobility of radiocesium in waterlogged soil. However, as it will be shown below, this conclusion in this case turns out to be only apparent. The appearance of secondary peaks on a deep slope of the vertical profiles may also indicate an increased migration ability of radionuclides in the soil due to their unstable physicochemical forms. This possibility occurs in the process of decomposition of organic substances and organic forms of radionuclides due to microbiological activity in the soil, when the soil was flooded with lake water. These processes are accompanied by the decomposition of the other potentially unstable carbonate and oxide physicochemical forms of radionuclides and the release of radionuclides into pore waters. If we compare the mobility of radiocesium and plutonium in terms of possible sizes of secondary peaks in Fig. 4 (soil core near Lake Bedugnis), we can conclude that radiocesium is more mobile in non-boggy soil than plutonium.

The results of XRD measurements made it possible to identify the clay admixture (muscovite $\text{KAl}_3\text{Si}_3\text{O}_{10}(\text{OH})_2$) in the studied samples. The studied samples also contained large amounts of quartz and other minerals, such as albite, anorthite and orthoclase

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

The uppermost layers (1 - 5 cm) of the waterlogged and non-boggy soil cores were rich in organic matter (Fig. 5). The deeper layers of the non-boggy soil samples taken near Lakes Juodis, Paršelis and Bedugnis consisted mainly of sand and their organic matter content was rather low.

Assuming that the studied radionuclides (^{137}Cs and $^{239,240}\text{Pu}$) have an affinity for organic matter, an estimate was made of the correlation between these radionuclides and organic matter. However, in our case, it was not possible to establish a consistent pattern of correlations. A statistically significant ($p\text{-value} < 0.05$) linear correlation of $^{239,240}\text{Pu}$ activity concentrations with organic matter



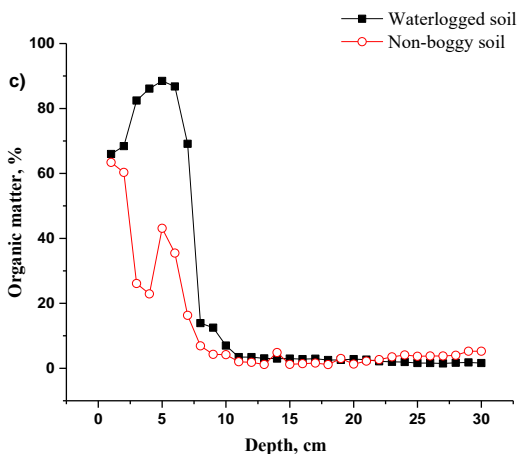


Fig. 5. Vertical profiles of the organic matter content (%) in the waterlogged and non-boggy soil cores taken near Lake Juodis (a), Lake Paršelis (b) and Lake Bedugnis (c).

was obtained for two waterlogged soil cores taken near Lake Paršelis ($r = 0.89$) and Lake Bedugnis ($r = 0.6$). The absence of correlation for ^{137}Cs ($r = 0.02$) was recorded in a waterlogged soil core taken near Lake Juodis and for $^{239,240}\text{Pu}$ in a non-boggy soil core taken near Lake Paršelis ($r = -0.19$). The correlation for plutonium was also insignificant for the waterlogged soil core taken near Lake Juodis. For the profile of the non-boggy soil core sampled near Lake Bedugnis, the correlation coefficient for $^{239,240}\text{Pu}$ was only 0.48. For ^{137}Cs , a significant correlation was obtained for the non-boggy soil cores sampled near Lake Juodis ($r = 0.95$), Lake Paršelis ($r = 0.77$) and Lake Bedugnis ($r = 0.83$), and two waterlogged soil samples taken on the shores of Lake Paršelis ($r = 0.88$) and Lake Bedugnis ($r = 0.7$). We concluded that a good correlation was obtained when the depth of the radionuclide activity concentration peak coincided well with the peak of soil organic matter, although this phenomenon remains likely to have been coincidental.

3.4. Investigation of the distribution of ^{137}Cs and $^{239,240}\text{Pu}$ activity concentrations in geochemical forms in the studied cores of non-boggy and waterlogged soils

3.4.1. Parameters of the studied samples of non-boggy and waterlogged soils (6th - 8th layers)

Data on the parameters of the studied samples of non-boggy and waterlogged soils (6th - 8th layers) are presented in Tables 6 - 7.

Table 6 Activity concentrations of radionuclides and properties of the 6th - 8th layers of the non-boggy soil core.

Depth	6-th layer	7-th layer	8-th layer
pH (H ₂ O)	3.91	4.21	4.43
Organic matter quantity, %	35.5	16.3	6.9
$^{239,240}\text{Pu}$, Bq/kg	1.47	4.85	1.98
^{137}Cs , Bq/kg	66.6	66.0	28.0

Table 7 Activity concentrations of radionuclides and properties of the 6th - 8th layers of the waterlogged soil core.

Depth	6-th layer	7-th layer	8-th layer
pH (H ₂ O)	4.28	4.19	3.94
Organic matter quantity, %	86.8	69.1	13.9
$^{239,240}\text{Pu}$, Bq/kg	0.89	0.84	0.64
^{137}Cs , Bq/kg	18.6	18.0	18.7

3.4.2. The point of zero charge (pH_{PZC}).

Experimental curves (ΔpH versus pH_i - pH of the initial solution) obtained by the salt addition method for assessing pH_{PZC} of non-boggy (a) and waterlogged soils (b) are shown in Fig. 6.

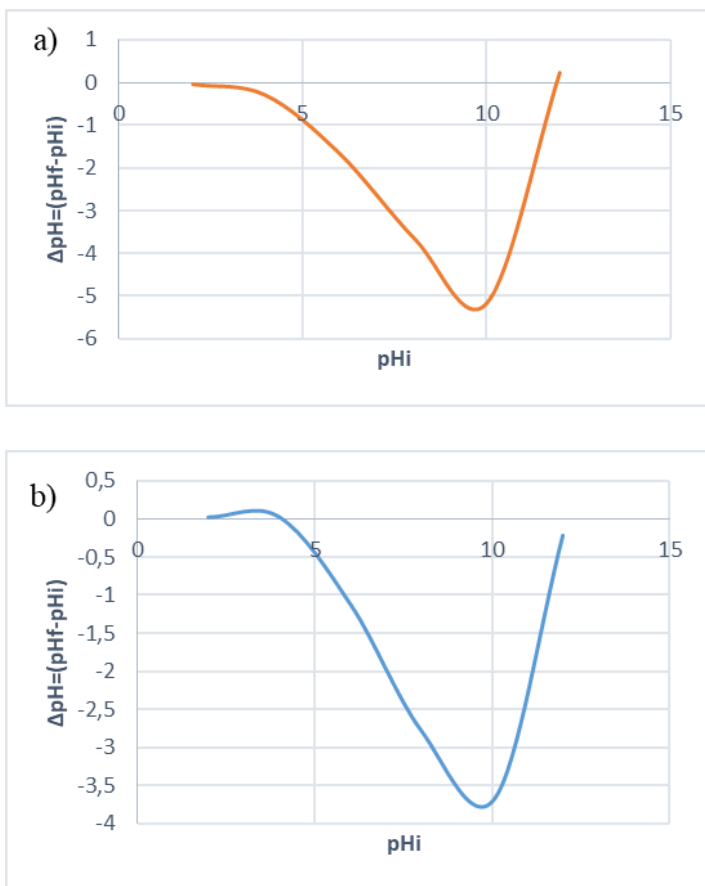


Fig. 6. The plots of ΔpH versus pH_i for assessing the point of zero charge of non-boggy (a) and waterlogged soils (b).

The pH_{PZC} of the non-boggy soil was 11.8 (Fig.6 (a)) and the pH_{PZC} of the waterlogged soil was 4.01 (Fig. 6 (b)). Thus, soil particles of the non-boggy soil sample have a positive surface charge and a negative charge is characteristic for the waterlogged soil sample.

3.4.3. Distribution of ^{137}Cs activity concentrations in physicochemical fractions in the studied layers of the waterlogged and non-boggy soil cores

Table 8 presents data on ^{137}Cs activity concentrations (fractions, %) in physicochemical forms (extracts) of the studied layers 6th - 8th cm of non-boggy and waterlogged soil cores.

Table 8 Distribution of ^{137}Cs activity concentrations in physicochemical forms (fractions, %) in layers 6 - 8 of the waterlogged and non-boggy soil cores (BDL - below detection limit) (1 - exchangeable; 2 - bound to carbonates; 3 - bound to oxides; 4 - bound to organic substances; 5 - residue).

	Layers of waterlogged soil			Layers of non-boggy soil		
	6-th	7-th	8-th	6-th	7-th	8-th
1	0.4±0.1	0.4±0.1	0.4±0.1	12.2±1.6	10.1±1.6	10.7±1.5
2	BDL	BDL	BDL	3.0±0.1	5.2±1.3	4.6±0.8
3	0.3±0.1	0.3±0.1	0.4±0.1	7.1±0.01	5.1±2.2	3.6±1.0
4	BDL	0.1±0.01	0.7±0.1	16.4±1.7	8.3±1.4	11.2±1.7
5	99.3±4.3	99.2±4.0	98.5±4.2	61.3±2.1	71.3±0.4	69.9±4.2

The largest activity concentrations of ^{137}Cs were determined in the residual fractions of the physicochemical forms of layers 6 - 8 of the waterlogged soil core. This distribution of ^{137}Cs activity concentration (Table 8) in the waterlogged soil can be explained by the negative surface charge of the sorbing soil components, in this case, clay minerals. Thus, the sorption of $^{137}\text{Cs}^+$ ions on clay particles of waterlogged soil is irreversible.

In non-boggy soil, the largest activity concentration of ^{137}Cs was also found in the residual fractions of the physicochemical forms of the layers 6 - 8. However, the activity concentrations of ^{137}Cs in these layers of non-boggy soil in the residual fraction were lower than those in the waterlogged soil. Relatively large activity concentrations of ^{137}Cs in the exchangeable and bound to organic matter fractions

were found in the layers 6 - 8 of this soil, which indicates an increase in the soluble ^{137}Cs concentration and an increase in its mobility in this soil. In non-boggy soil, the determining factor for sorption of radiocesium is the positive surface charge of the soil particles.

3.4.4. Distribution of $^{239,240}\text{Pu}$ activity concentrations in physicochemical fractions in the studied layers of the waterlogged and non-boggy soil cores

Data on the $^{239,240}\text{Pu}$ activity concentrations (fractions, %) in physicochemical forms (extracts) in the studied layers 6 to 8 (6th - 8th cm) of the non-boggy and waterlogged soil cores are presented in Table 9.

Table 9 Distribution of $^{239,240}\text{Pu}$ activity concentration in the geochemical forms (fractions, %) in the layers 6 - 8 of the waterlogged and non-boggy soil cores (1 - exchangeable; 2 - bound to carbonates; 3 - bound to oxides; 4 - bound to organic matter; 5 - residual fraction).

	Layers of waterlogged soil core			Layers of non-boggy soil core		
	6-th	7-th	8-th	6-th	7-th	8-th
1	9.6±2.2	13.9±3.7	10.3±2.3	3.2±0.7	4.8±0.1	5.1±1.1
2	0.3±0.09	1.0±0.3	0.3±0.7	1.0±0.1	2.7±0.7	1.4±0.3
3	12.1±2.4	15.9±4.1	13.4±3.0	54.5±4.5	49.1±3.3	45.1±6.7
4	49.5±3.7	53.4±5.5	52.2±11.8	0.7±0.2	1.9±0.5	9.4±0.9
5	28.5±4.8	15.8±4.5	23.8±3.0	40.6±7.6	41.5±5.8	39.0±6.3

The largest activity concentrations of $^{239,240}\text{Pu}$ were determined in the fraction associated with organic matter in layers 6 - 8 of the waterlogged soil core. A relatively small part of $^{239,240}\text{Pu}$ activity concentration was found in the residual and Fe/Mn oxide fractions of the studied layers, while the exchangeable fraction was relatively large. The large exchangeable fraction of the activity concentration of plutonium indicates an increase in the radionuclide solubility

potential in waterlogged soil and its high mobility under flood conditions. This distribution of Pu ions is due to the fact that plutonium tends to form complexes with fulvic and humic acids, which are soluble at the pH of waterlogged soil. This is confirmed by the relatively large Pu exchangeable fraction, which was determined in the studied layers of waterlogged soil.

On the contrary, in the layers 6 - 8 of the non-boggy soil core, the largest activity concentration of $^{239,240}\text{Pu}$ was found in the oxide fraction. A smaller part of the $^{239,240}\text{Pu}$ activity concentration was recorded in the residual fraction, but it was much higher than that in waterlogged soil. A small part of the $^{239,240}\text{Pu}$ activity concentration was found in the exchangeable and organic matter fractions. The determinant factor of plutonium sorption in this soil is the formation of complex compounds with Fe and Mn (hydr)oxides.

The study of the physicochemical forms of some stable elements was carried out in order to determine the elements with the distribution of the respective forms, similar to plutonium. However, analogues of stable elements for this radionuclide have not been found. The concentrations of Ca and Mn in the organic matter fraction were low in layers 6 - 8 of waterlogged soil at relatively large activity concentrations of plutonium in these layers. In the non-boggy soil core, the pattern was opposite.

Summarizing the data of all three layers of the non-boggy soil core, it can be stated that the trend in the distribution of radionuclides in the physicochemical forms of these layers is similar. $^{239,240}\text{Pu}$ is mainly concentrated in the oxide fraction, less in the residual fraction and even less in the fraction associated with organic matter. The largest part of ^{137}Cs is concentrated in the residual fraction, less in the organic matter, and even less in oxide and carbonate fractions. In contrast to plutonium, rather large exchangeable fractions of ^{137}Cs (9.6 - 12.2 %) indicate its high mobility in non-boggy soil.

In waterlogged soil, we see a completely different distribution of radionuclides and stable elements in physicochemical forms than in non-boggy soil. Here, most of $^{239,240}\text{Pu}$ is associated with the

organic matter fraction and concentrated in the residue. Also, its significant exchangeable fractions (9.6 - 13.9 %) confirm high mobility of the radionuclide. Summarizing the data of the layers 6 - 8 of waterlogged soil, we can say that more soluble fractions appear with increasing depth. At the same time, ~99 % of ^{137}Cs is concentrated in the residual fraction. Insignificant amounts of Ca, Fe and Al associated with carbonates are characteristic of acidic soils. A decrease in the fractions of Ca bound to oxides in the studied layers of waterlogged soil in comparison with those in non-boggy ones may indicate the presence of anaerobic conditions.

CONCLUSIONS

1. The distribution coefficient K_d for plutonium in soil is a variable parameter whose values vary within three orders of magnitude, depending on certain properties of the soil, specifically particle size and the amount of organic material ($r = 0,98$). The order of plutonium sorption capacity in different soil types was determined as follows: peat > (6597 l/kg) > clayey sand (3240 l/kg) > clayey loam (1155 l/kg) > sand (718 l/kg) > silty sand (208 l/kg).

2. The mobility of Cs^+ in waterlogged soil is limited because the highest concentration thereof (99 %) has been established in the immobile residual fraction due to the negative surface charge of soil particles and strong Cs^+ sorption in clay minerals. Meanwhile, Cs^+ is more mobile in non-boggy soil due to the positive surface charge of the soil particles and the acidic pH of the soil, where the percentage of its exchangeable fraction has been established to be 10 - 12 %, while the percentage of Cs^+ concentration in the residual fraction is 61 - 71 %.

3. Pu has lower mobility in non-boggy soil because the largest percentage of its concentration (45 - 54 %) has been established in the immobile oxide fraction due to dominant sorption of Pu by

materials containing Fe and Mn (hydr)oxides, while a lesser percentage thereof (23 - 40 %) has been detected in the residual fraction. Pu ions in waterlogged soil have notably high migration potential due to the formation of compounds that are soluble with soil organic matter, as the majority of its concentration has been established in potentially mobile fractions: in the organic material fraction (49 - 53 %), as well as a significant percentage in the exchange fraction (12 - 16 %).

4. An uneven background distribution of $^{238,239,240}\text{Pu}$ and ^{137}Cs in potentially important Lithuanian locations that has already been evaluated before the activation of certain nuclear facilities by Lithuania's neighbours, has allowed us to identify the zones of long-term radionuclide accumulation: the bottom sediments of the Curonian Lagoon and on the coast of Nida as well as floodplain soil near Krokų Lanka and near the village of Plaušvariai. The measurements performed may serve as a baseline for the assessment of future environmental contamination caused by said nuclear facilities and are important for the characterisation of the sorption properties of soil with regard to the barrier zones of radioactive waste sites.

THE LIST OF AUTOR ORIGINAL PUBLICATIONS

Articles in journals:

1. D. Marčiulionienė, B. Lukšienė, D. Montvydienė, O. Jefanova, J. Mažeika, R. Taraškevičius, R. Stakėnienė, R. Petrošius, E. Maceika, N. Tarasiuk, Z. Žukauskaitė, L. Kazakevičiūtė, M. Volkova. 2017. ^{137}Cs and plutonium isotopes accumulation/retention in bottom sediments and soil in Lithuania: A case study of the activity concentration of anthropogenic radionuclides and their provenance before the start of operation of the Belarusian Nuclear Power Plant (NPP). J. Environ. Radioact. **178-179** 253-264. DOI:[10.1016/j.jenvrad.2017.07.024](https://doi.org/10.1016/j.jenvrad.2017.07.024).

2. L. Kazakevičiūtė-Jakučiūnienė, R. Druteikienė, E. Maceika, B. Lukšienė, R. Juškėnas, V. Pakštas, Z. Žukauskaitė, R. Gvozdaitė, N. Tarasiuk. 2021. Impact of soil organic matter on Pu migration in five Lithuanian surface soils. J. Environ. Radioact. **237** 106702. DOI:<https://doi.org/10.1016/j.jenvrad.2021.106702>.

3. L. Kazakevičiūtė-Jakučiūnienė, N. Tarasiuk, E. Maceika, R. Druteikienė, M. Konstantinova, Z. Žukauskaitė, R. Gvozdaitė, Š. Buivydas. 2021. ^{137}Cs and $^{239,240}\text{Pu}$ activity concentrations distribution in waterlogged and non-boggy soils of Lithuania. Lithuanian Journal of Physics.

Thesis of conferences:

1. L. Nedzveckienė, B. Lukšienė, S. Tautkus. Plutonio K_d verčių palyginamasis tyrimas skirtingo tipo dirvožemiuose” Doktorantų ir jaunųjų mokslininkų konferencijoje 2015 m. spalio mėn. 22-23 d., Vilnius.

2. L. Nedzveckienė, B. Lukšienė, N. Tarasiuk, Z. Žukauskaitė: Comparative research of vertical migration of ^{137}Cs in the soil of flooded and upland banks of lakes. Chemistry and Chemical Technology 2016 balandžio 28-29 d, Vilnius

3. L. Nedzveckienė, B. Lukšienė, Š. Buivydas, R. Gvozdaitė „Vertical distribution of $^{239,240}\text{Pu}$ and ^{238}Pu analytes in the soil. 59th International Conference for Students of Physics and Natural Sciences Openreadings 2016. kovo mėn. 15-18 d., Vilnius.

4. L. Nedzveckienė, B. Lukšienė „Determination of plutonium sorption capacity in the different type of soil. Fourth international conference on radiation and application in various fields of research 2016 gegužės 23-27 d., Niš, Serbija.

5. L. Nedzveckienė, B. Lukšienė, N. Tarasiuk, Doktorantų ir jaunųjų mokslininkų konferencijoje su pranešimu ^{137}Cs ir Pu izotopų savitojo

aktyvumo koncentracijų pasiskirstymas ežero kranto dirvožemio profiliuose, 2016 spalio mėn. 26 d., Vilnius.

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ANOTACIJA

^{137}Cs ir plutonio izotopai aptinkami paviršiniame dirvožemio sluoksnyje, prisijungę prie dirvožemio komponentų. Radionuklidų migracijos greitis dirvožemyje priklauso nuo daugelio veiksnių, tokių kaip cheminė radionuklidų forma, geocheminė dirvožemio sudėtis, fizikinės ir cheminės sąlygos, atmosferos kritulių kiekis, augmenija ir jos rūšys, mikroorganizmų aktyvumas, gyvūnų veikla, taip pat žmogaus veikla. Siekiant užtikrinti deramą radiacinę saugą bei kurti efektyvias užterštų teritorijų atstatomąsias priemones būtina kaupti ir analizuoti ilgalaikių radionuklidų koncentracijų matavimų duomenis, apibūdinančius buvusią bei esamą taršą dirvožemyje bei jos evoliuciją.

Šioje daktaro disertacijoje apibendrintų mokslinių tyrimų tikslas - nustatyti veiksnius lemiančius ^{137}Cs ir Pu izotopų sorbciją/desorbciją ir vertikaliąją pernašą skirtingų tipų bei skirtingo drėgmės režimo (santykinai sauso ar periodiškai užliejamo dirvožemio drėgmės režimo) dirvožemiuose.

Įvertinta plutonio sorbcinė geba skirtingo tipo dirvožemiuose, esant skirtingiems dalelių dydžiams, organinių medžiagų bei stabilių elementų kiekiams. Išanalizuoti Pu ir ^{137}Cs sorbciją/desorbciją lemiantys veiksniai sausame ir užliejamame dirvožemiuose. Praktiškai pritaikytas nuosekliosios ekstrakcijos metodas vertinant ^{137}Cs ir Pu migracijos galimybes sausame ir užliejamame dirvožemiuose.

NOTES

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