Lithuanian Journal of Physics, Vol. **56**, No. 2, pp. 111–123 (2016)

INVESTIGATION OF ^{137}Cs AND PLUTONIUM ISOTOPE SORPTION–DESORPTION IN BIO- AND SYNTHETIC MATERIALS

B. Lukšienė, Z. Žukauskaitė, N. Tarasiuk, E. Maceika, V. Filistovič, Š. Buivydas, L. Nedzveckienė, and D. Jasinevičienė

Center for Physical Sciences and Technology, Savanorių 231, LT-02300 Vilnius, Lithuania
E-mail: bena.luksiene@ftmc.lt; zita.zukauskaite@ftmc.lt

Received 21 May 2015; revised 5 March 2016; accepted 21 June 2016

Investigations on the pre-concentration of radionuclides (^{137}Cs and plutonium isotopes) from fresh water on solid matrices are presented in this study. A particular focus was given to an innovative physico-chemical removal process such as adsorption of radionuclides from an aqueous medium on new type adsorbents, environmental-friendly materials. Sorption of the tested radionuclides from the lake water solution by environmental assays and synthetic sorbents was compared. Lake water was analyzed for main anions, micro- and macroelements, using ion and atomic absorption chromatography methods, respectively. Batch type and dynamic flow column laboratory experiments were performed. The sorption–desorption capacity of radionuclides by the tested sorbents was estimated based on the results of α- and γ-spectrometric measurements. According to the removal efficiency results, moss can be considered as the best sorbent for plutonium of the tested environmental-friendly sorbents, whereas the moss sorption capacity exceeded even that of the tested synthetic ones. The highest ^{137}Cs removal efficiency from the lake water solution was obtained for granular activated carbon. Application of the dynamic flow method carried out *in situ* confirmed the reversibility of sorbed background ^{137}Cs activity in the moss observed during the batch type experiments, and the method of the moss bed column turned to be unfit for the pre-concentration of ^{137}Cs from the lake water.

Application of a single moss bed column for the ^{137}Cs and plutonium isotope pre-concentration for the first time carried out *in situ* allowed us to determine the activity concentration of ^{239,240}Pu and ^{238}Pu in the lake water – 4.87±0.98 and 0.67±0.21 mBq/m^3, respectively.

The reversible ^{137}Cs and different plutonium sorption in the moss apparently indicates different binding properties of these radionuclides to the moss, therefore further investigations on this issue are foreseen.

Keywords: radionuclide, sorbents, batch experiments, dynamic flow method

PACS: 29.87.+g, 28.60.+s, 29.30.Kv

1. Introduction

Nuclear and non-nuclear industrialization has caused world-wide spreading of radionuclides, heavy metals, organics, etc. in the environment. Nowadays, artificial radionuclides are found side by side with naturally occurring radionuclides in the environmental samples [1–12].

Cesium and plutonium isotopes generally exist in the environment as a result of releases during nuclear weapons testing or accidental events at nuclear facilities. ^{137}Cs is one of the main contributors to the long-term radioactive dose to the population [13]. The majority of plutonium isotopes are α-particles emitting, radiotoxic, some of them are extraordinary long-lived (^{239}Pu: $T_{1/2} = 2.4\times10^4$ y.; ^{240}Pu: $T_{1/2} = 6.6\times10^3$ y.), and all of them need to be radiochemically isolated from environmental samples for the assessment of their activity concentration.

Removal of pollutants from industrial wastewater and assessment of artificial radionuclide activity concentrations in natural water bodies are of great concern. The conventional processes for removing heavy metals from wastewater include many processes such as chemical precipitation, flotation, adsorption, ion exchange, and electrochemical deposition. Over the past decade, adsorption has become the method of choice for the purification of water and wastewater. Basically, adsorption is a mass transfer process by which a substance is transferred from the liquid phase to the surface of a solid, and becomes bound by physical and/or chemical interactions [14]. The main principle of sorbents applied for the adsorption of contaminants from aquatic media is their capability to participate in the sorption–regeneration cycles. The biosorbent of such a type – a rice straw bed column – was used to remove heavy metals (Cd and Pb) from single and binary solutions. The biosorbent is promising because of its feature to be reused for several sorption–regeneration cycles and the possibility of employing this biosorbent for the remediation of the actual electroplating wastewater containing Cd(II) and Pb(II) ions [15].

Various low-cost adsorbents, derived from agricultural waste, industrial by-products, natural materials, or modified biopolymers, have been recently developed and applied for the removal of heavy metals from metal-contaminated wastewater [16].

Biosorption is a physicochemical process that occurs naturally in certain biomass which allows it to passively concentrate and bind contaminants onto its cellular structure [17]. Biosorbents are cheaper, more effective alternatives for the removal of metallic elements, especially heavy metals from an aqueous solution [18]. When choosing biomass, the main factor to be taken into account is its availability and cheapness [19]. According to the factors mentioned above, biomass can appear from industrial wastes and organisms which can be found easily in nature or specially prepared for biosorption studies [20]. A wide range of materials have been evaluated for the adsorption of heavy metals, including microbial and seaweed biomass (living or dead), cellular products, activated carbons, sewage sludges, clays and clay-like minerals, hybrid materials and ordered mesoporous silica [20]. Among these, living or dead lignocellulosic biomass consisting of natural polymeric materials is a promising alternative to commercial adsorbents for the purification of metal-bearing effluents [20].

Biosorption processes are still at the stage of a laboratory-scale study in spite of the unquestionable progress [21]. In this connection, we tried to apply the biosorbent-moss bed column for evaluation of heavy-to-measure radionuclide (the case of plutonium isotopes) activity concentration in the lake water. Data on distribution of radionuclides, especially of plutonium isotopes, in the freshwater column are scarce enough and, according to the available literature, only lake water in Greenland and Finland was analyzed for plutonium isotopes [22, 23].

In addition, there are no data on the activity concentration of plutonium isotopes in the water of Lithuanian lakes. Low ^{137}Cs and ^{239,240}Pu activity concentrations in lake water require that large volumes of water should be analyzed. The lake water of 20 L volume should be taken for ^{137}Cs analysis [24], and if ^{134}Cs is present, to determine the ^{134}Cs/^{137}Cs ratio the lake water volume of 900 L should be used [22]. The concentrations of ^{239,240}Pu and ^{241}Am in lake water samples of 400 L volume in Finland were investigated in 2007 [23]. In 2007, the average activity concentrations of ^{239,240}Pu and ^{137}Cs in the unfiltered water of Lake Päijänne, Finland, were 4.9±0.9 mBq/m^3 and 19.3±1.4 Bq/m^3, while the average total activities of sediment profiles were 45±15 Bq/m^2 and 100±15 kBq/m^2, respectively [23]. Changes in the ^{137}Cs activity concentration in the filtered water of Lake Juodis, Lithuania, were in the range of 1.4±0.2 to 3.3±0.5 Bq/m^3 within different seasons of the 2004–2006 period [25]. The ^{137}Cs activity concentration in the water of Lake Lago Maggiore, Italy, was rather constant at less than 1 mBq/L in 2004 [26].

In this connection, removal of radioactive pollutants and the recovery of important radionuclides from Lithuanian aqueous systems require a special attention. Therefore, the main aim of the current work was to apply some environmental-friendly biomaterials to the pre-concentration of ^{137}Cs and plutonium isotopes from aqueous media. Considering biomaterials as promising alternatives to commercialized sorbents, the investigation of sorption–desorption processes of ^{137}Cs and ^{236}Pu to/from the tested sorbents as well as sorption of ^{137}Cs, ^{239,240}Pu and ^{238}Pu from the radionuclide-bearing fresh water onto the moss bed column *in situ* was carried out.

2. Materials and methods

All experiments were conducted using the cesium isotope ^{137}Cs and plutonium isotope ^{236}Pu under ambient conditions at pH 6_{H_2O}. The test-solutions of ^{137}Cs (I) with the concentration of 1 Bq/L and ^{236}Pu (IV) with the concentration of 0.6 Bq/L were prepared in the lake water from commercially available ^{137}Cs and ^{236}Pu standard solutions (the Czech Metrology Institute Inspectorate for Ionizing Radiation). The pH value of solutions was adjusted to the pH value of 6 (such as that of natural lake water) with 6 mol/L NaOH.

2.1. Determination of physicochemical parameters of tested assays

The mineralization of the surface water of Lake Akmena was determined. The lake water was analyzed for the anions SO_4^{2-}, NO_3, Cl$^-$, F$^-$, PO_4^{3-} and the cation NH_4^+ by means of the ion chromatography – Dionex 2010i with a conductivity detector, and the Ion Pac AS4A-SC column was used for anion analyses.

Concentrations of micro- and macroelements (Cu, Mn, Co, Na, Fe, Ca, Mg, Pb, K) in the lake water were determined by using an atomic absorption spectrometer with a flame atomizer (AAnalyst 800, *PerkinElmer*, USA).

The activity concentration of ^{137}Cs in the surface water of Lake Akmena was determined by evaporating 20 L of unfiltered water and measuring the dry residue γ-spectrometrically. The initial activity (as a background activity) concentration of ^{137}Cs, ^{239,240}Pu and ^{238}Pu in moss and lichen assays was estimated.

2.2. Laboratory-scale sorption experiments

Environmental assays such as moss (*Ptilium crista-castrensis*), lichen (*Cladonia rangiferina*), mycorrhizal fungi chanterelles, pine needles and raw clay were used for ^{137}Cs and ^{236}Pu pre-concentration from the lake water. As it is known, moss, lichen and pine needles possess a specific ability to bind heavy metals and radionuclides. Chanterelles, among the main carbohydrates containing chitin in their composition [27, were selected for radionuclide sorption from an aqueous solution. Chitin occurs in nature as ordered crystalline microfibrils forming structural components in the cell walls of fungi and yeast [28]. Chitin-based materials can be used for the treatment of industrial pollutants and adsorb actinides [29]. Clay, filter material AFA-RMP-20 and granular activated carbon are generally recognized sorbents.

All these materials were dried at 70 °C until constant mass. Biomaterials were chopped into small pieces, the dried clay sample was used as it is and all sorbents were put into glass columns (sorbent bed height 11.5 cm) up to the 100 mL volume. The mass of separate materials used in laboratory experiments (moss 9.07 g, lichen 14.54 g, mycorrhizal fungi chanterelles 20.84 g, pine needles 15.28 g, clay 132.33 g, filter material AFA-RMP-20 12.93 g, granular activated carbon 29.01 g) was different in the column, but their volume was 100 mL. A principal scheme of experimental procedures is presented in Fig. 1.

Two sets of the column packing were used. In one set, up to 100 mL of the material was packed into a separate column. The packing of columns of the second set depended on the obtained sorption capability of sorbents in the first set of experiments. In the first column, five sorbents of equal slices of up to 100 mL were placed from the lowest sorption capability sorbent on the column bottom to the highest sorption capability one at the top. In the second column, sorbents were placed in the opposite direction – from the highest sorption capability sorbent at the bottom to the lowest sorption capability sorbent at the top.

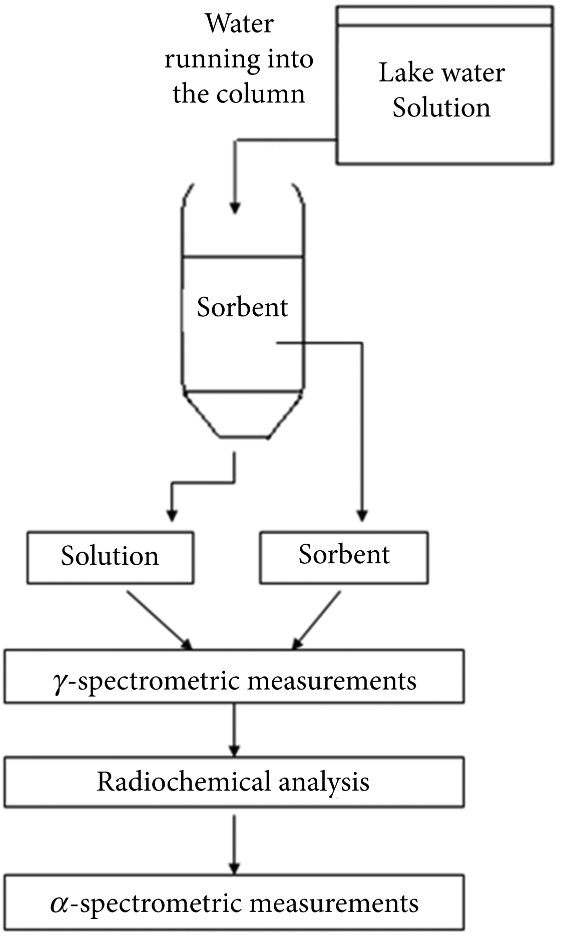

Fig. 1. A principal scheme of experimental procedures.

The test-solution (volumetric solid : solution ratio 1:5) was poured onto the top of the column and the flow rate was regulated to 3 mL/min. The solution, passing through sorbents, was collected, evaporated and prepared for the α- or γ-spectrometric measurement. When the sorption process was finished, the sorbents were taken out dividing them into two equal segments: upper and bottom. Both sorbent segments were dried, the effluent from the columns was evaporated and all aliquots were prepared for the α- or γ-spectrometric measurement. According to the results of spectrometric measurements we evaluated the activity of ^{236}Pu and ^{137}Cs accumulated by the sorbent.

2.3. Laboratory-scale desorption experiments

One set of desorption experiments was carried out applying a static batch method. For batch experiments 9.07 g of moss and 13.89 g of lichen with known activities of ^{137}Cs were placed in dishes and 450 mL of the unfiltered lake water was added. Dishes were agitated at a constant speed of 100 rpm for 20 h. After the suspensions were shaken for 20 h, the solid and liquid phases were separated by filtration. The filtrate residue after the evaporation and the solid phases (moss and lichen) were prepared for measurements. The measured ^{137}Cs activity was compared with the initial ^{137}Cs activity in moss and lichen.

2.4. *Application of a dynamic flow column method in situ*

To test the ^{137}Cs and plutonium isotope possibility to be pre-concentrated by an environmental-friendly assay, the moss bed columns were employed on the shore of Lake Akmena *in situ*. Two columns (moss bed height 14 cm) were installed and packed with 90±5 g of moss which contained 38.7±1.5 Bq/kg of ^{137}Cs and 0.0057±0.0009 and 0.0014±0.00041 Bq/kg of ^{239,240}Pu and ^{238}Pu, respectively. The lake water of 300 L volume was loaded through one column within 100 min (flow rate 0.050 L/s) and through the second column within 200 min (flow rate 0.025 L/s). Samples of moss were prepared in the laboratory for the γ- and α-spectrometric measurement. Actually, 20 L of lake water was taken for the ^{137}Cs activity concentration evaluation followed by evaporation and the γ-spectrometric measurement.

2.5. *Radiochemical method and measurement of plutonium isotope analytes*

The determination of plutonium analytes requires the application of the sample destruction method. The organic matter, which complicates the chemical Pu separation, was decomposed by heating at 550 °C overnight and after that to 700 °C for 2 h. The organic matter content was determined on the loss of ignition. The samples were spiked with 0.0125 Bq ^{242}Pu as a yield monitor. The ^{236}Pu analyte in the model experiments was separated by applying a simplified radiochemical method, only the anion-exchange resin column method. Plutonium isotope purification and separation in the environmental samples were based on the use of the combined two-column method – ion exchange and extraction chromatography.

The measurement of plutonium analytes, emitting α particles, is possible when thin-layered sources are used. The electrochemical method was applied to obtain thin layer plutonium sources for the alpha spectrometric measurement. Plutonium was electrodeposited for 1 h under direct 0.6 A/cm^2 current onto a one-side polished stainless steel disc-electrode in a sulphate medium [7, 8].

The plutonium isotopic composition and its amount in the sample were determined by the use of a semiconductor alpha spectrometer OctetePlus with ORTEC large square (450 mm^2) Si detectors (BU-020-450-AS) with the resolution capability of 20 keV. The spectra were treated and analyzed applying AlphaVision and Maestro programs. The detection limit for the counting time of 86400 s is about 10^{-3} Bq for plutonium isotopes [8, 9].

2.6. *^{137}Cs sample treatment and measurement*

The solid samples were air-dried at room temperature and then, if needed, were burned in a muffle furnace up to 400 °C temperature overnight. The aqueous samples were evaporated thoroughly and residual material was prepared for the γ-spectrometric measurement. The solid and aqueous samples were analyzed for ^{137}Cs using a CANBERRA γ-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 were carried out in standard geometry and at known efficiencies according to the densities of samples. The measurement time needed for obtaining the acceptable low counting errors, 5–15%, varied from 24 to 72 h. Measurement errors of the radiocesium activity concentrations in the samples were evaluated by the GENIE software program [9].

2.7. *Physical parameters*

The removal efficiency R of ^{137}Cs and plutonium isotopes by the treated sorbents was calculated using the following equation [30]:

$$R(\%) = \frac{C_0 - C_e}{C_0} \times 100. \tag{1}$$

Here C_0 (Bq/L) and C_e (Bq/L) are the initial and final radionuclide activity in the solution, respectively.

The removal efficiency for desorption can be calculated according to the activity concentration in the solid biomass:

$$R(\%) = 100 - \frac{C_e}{C_0} \times 100. \tag{2}$$

The sorption/biosorption capacity q_e was calculated based on the amount of the radionuclide adsorbed per unit dry weight of the treated sorbent mass [30]:

$$q_e(\text{Bq/kg}) = (C_0 - C_e) \times \frac{V}{m}, \tag{3}$$

where C_0 and C_e are the initial and equilibrium aqueous volumetric activity (Bq/L), V (L) is the solution volume, and m (kg) is the dry mass of the sorbent.

The calculated radionuclide distribution (partition) coefficient (K_d) values were presented as follows [31]:

$$K_d(\mathrm{mL/g}) = \frac{C_0 - C_e}{C_e} \times \frac{V}{m}. \quad (4)$$

Here C_0 and C_e are the initial and equilibrium aqueous volumetric activity (Bq/L), respectively, V is the solution volume (L), and m is the sorbent dry mass (kg).

3. Results and discussion

3.1. Physical and chemical parameters of treated systems

Concentrations of the main anions (SO_4^{2-}, NO_3^-, Cl^-, F^-, PO_4^{3-}) and the cation NH_4^+ in the lake water were evaluated (Table 1). The basic anion in the lake water according to the concentration values appeared to be SO_4^{2-}, whereas the PO_4^{3-} concentration was lower than 0.2 mg/L (PO_4^{3-} detection limit (d.l.) 0.2 mg/L). The value of NH_4^+ concentration less than 0.03 mg/L was determined in Lake Lago Maggiore [26].

Table 1. Anion and cation concentrations in the studied lake water.

Ion concentration, mg/L					
SO_4^{2-}	NO_3^-	Cl^-	F^-	PO_4^{3-}	NH_4^+
14.23	0.42	4.76	0.38	d.l.	0.05

The NH_4^+ concentration value was 0.05 mg/L (close to the value of 0.03 mg/L in Lake Lago Maggiore [26]) and such a concentration level is characteristic of the surface water layer due to the rather high oxygen concentration. Under such oxidizing conditions, plutonium in the lake water could be as Pu(IV), Pu(V), and Pu(VI). Based on the equilibrium reactions, ligands, such as chloride and nitrate, form weak complexes with plutonium, whereas fluoride, sulfate, phosphate, citrate, and oxalate form stronger complexes. Among the strongest complexes of plutonium the hydroxy-carbonate mixed ligand complexes can be mentioned (e. g. $Pu(OH)_2(CO_3)_2^{2-}$). Pu(IV), mainly as hydrolysis products and humic complexes, and Pu(V), mainly as PuO_2^+, are generally the dominant oxidation states in most natural waters [32]. In our opinion, adsorption of plutonium onto particulates in the lake water is quite probable due to the low levels of dissolved plutonium (about 10^{-15} M) in the environment. Consequently, the particulates with adsorbed plutonium may be the principal regulator of the mobility of this radionuclide in the lake water system.

The data of Table 2 show that the concentration values of microelements Cu, Mn, Co, Pb were lower than the detection limit (detection limits (d.l.), μg/L: Cu 1.5, Mn 1.5, Co 9, Pb 15). The highest concentration of macroelements was determined for Ca, 16.2 mg/L. The macroelement potassium (the competing ion of ^{137}Cs) showed a rather low concentration, it equalled 0.90 mg/L. In Italian Lake Lago Maggiore within the water column it was nearly constant and equalled 1.5 mg/L [26].

Na^+, K^+, Ca^{2+}, and Mg^{2+} as typical components characteristic of natural waters have a competing effect on the sorption/removal of cesium radionuclides from aquatic media [33]. It can be assumed that dissolved potassium and ammonium would compete with cesium for adsorption sites, thereby decrease K_d values.

It has been found that the Na^+ and Ca^{2+} cations do not affect the Cs^+ sorption in a wide range of their concentrations (up to 250 mg/L), and the effect of K^+ is insignificant to the lower concentration limit (50 mg/L); at higher concentrations of potassium ions, they have a competing effect on the selective removal of cesium from aquatic media. At the K^+ concentration of 50 mg/L, the Cs^+ sorption value decreases by a factor of almost 1.5, and at the concentration of 250 mg/L the K^+ ions practically suppress the sorption, which additionally confirms the ion-exchange mechanism of the Cs^+ sorption, taking into account the close radii of the hydrated potassium and cesium ions [33]. Considering low concentration values of K, Na, and Ca in the tested lake water of this study (Table 2), it could be hardly probable that these cations affected the ^{137}Cs sorption/desorption processes. However, it could be taken into account that large hydrated cations, such as Na^+, Ca^{2+}, and Mg^{2+}, tend to pry the layers of sorbents further apart, whereas small hydrated cations, such as K^+, have the opposite effect [33].

Table 2. Concentrations of micro- and macroelements in lake water.

Heavy metal and macroelement concentration, mg/L								
Cu	Mn	Co	Pb	Na	Fe	Ca	Mg	K
<d.l.	<d.l.	<d.l.	<d.l.	2.06	0.04	16.2	3.4	0.90

The mineralization of the surface water of Lake Akmena during the sampling period (autumn) was 158.5 g/m^3, while in Lake Juodis [25] the maximum values reached ~174 g/m^3 in winter. The activity concentration of ^{137}Cs in the surface water of Lake Akmena was determined to be 0.98±0.24 Bq/m^3.

The initial activity (as a background activity) concentration of ^{137}Cs, ^{239,240}Pu and ^{238}Pu in the moss assay was 38.7±1.5, 0.0057±0.0009, and 0.0014±0.00041 Bq/kg, respectively. The initial concentration of ^{137}Cs, ^{239,240}Pu and ^{238}Pu in the lichen assay was 9.71±0.59, 0.0157±0.0018, and 0.0032±0.0008 Bq/kg, respectively.

3.2. ^{137}Cs and ^{236}Pu in laboratory-scale column experiments

3.2.1. ^{236}Pu and ^{137}Cs removal from lake water by different sorbents

The efficiency of ^{236}Pu removal from the lake water by different sorbents under the same experimental conditions was calculated applying Eq. (1). It is seen (Fig. 2) that raw clay exhibited the highest removal efficiency, it came up to ~90%. Moss removed ~50% of the initial plutonium activity in the solution. Removal of Pu by assays of lichen, chanterelles and pine needles was in the range of 30–40%. The granular activated carbon and filter material AFA-RMP-20 showed a very high removal efficiency with the percentage of removed ^{236}Pu in the range of approximately 60–70% (Fig. 2). According to the experimental results, the tested sorbents, referring to the removal efficiency, can be ranked in the following sequence: raw clay > granular activated carbon > filter material > moss > lichen > mycorrhizal fungi chanterelles > pine needles. The characteristic feature for clay and moss samples was observed during the process of sorption: sorbents became swollen and their volume in the column significantly increased. The flowing water with ^{236}Pu penetrated between clay particles, formed pore water, and the flow rate was very slow. In contrast, the moss volume increased due to the penetration of flowing water inside the moss. The amount of water consumed for the pore water formation in clay and the amount of water soaked in the moss were evaluated by measuring the water volume passed through the column.

Although the efficiency of ^{236}Pu removal by clay is very high, clogging of the column shortly causes limitations in the application of the clay column as a natural sorbent. Consequently, in studying the radionuclide removal from the aqueous medium under the tested conditions, the moss bed column appeared to be the best sorbent compared to the other environmental-friendly sorbents (chanterelles, lichens, pine needles).

Taking into account that the same solution volume, spiked with ^{236}Pu, was used for each kind of sorbents, the sorption/biosorption capacity (Eq. (3)) was determined to be significantly dependent on the sorbent dry mass. As it can be seen, moss of all

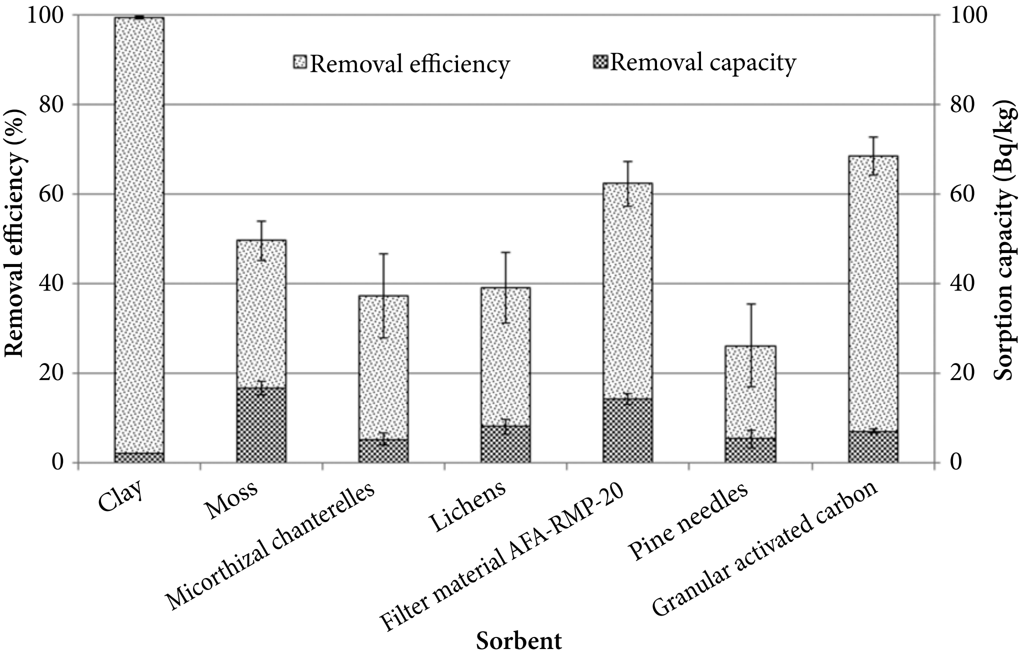

Fig. 2. Removal efficiency of ^{236}Pu and sorption capacity of environmental-friendly and synthetic sorbents from lake water spiked with ^{236}Pu (pH 6), using the dynamic flow column method.

tested sorbents showed the highest sorption capacity and the sorption capacities of the rest sorbents were decreasing approximately according to their mass (Fig. 2).

As regards ^{236}Pu, its removal efficiency values allow us to state (rejecting defective clay) that moss can be considered as the best sorbent of the tested environmental-friendly sorbents (Fig. 2), whereas the sorption capacity of moss is the largest even compared to that of all tested sorbents (Fig. 2).

Results of the investigation of ^{236}Pu sorption by various sorbents revealed that the raw clay column clogged very rapidly, and thus complicated an implementation of the experiment. Therefore, ^{137}Cs sorption experiments were carried out with four selected sorbents, which showed the higest removal efficiency and sorption capacity for ^{236}Pu, i. e. moss, lichens, granular activated carbon and filter material AFA-RMP-20. By analyzing the results of γ-spectrometric measurements and calculations in Eq. (1), we determined that granular activated carbon had the highest removal efficiency, about 50%, while the removal efficiency of the filter material AFA-RMP-20 reached ~10% (Fig. 3). Among sorbents, environmental-friendly materials, the lichen bed column showed a higher removal efficiency for ^{137}Cs compared to that of the moss bed column (Fig. 3).

Environmental-friendly sorbents, lichens and moss showed a significantly larger sorption capacity to ^{137}Cs, compared to those of the filter material and granular activated carbon (Fig. 3).

3.2.2. Mixed type sorbent columns for ^{236}Pu removal from lake water spiked with ^{236}Pu

The mixed type sorbent bed was applied to remove Pu from the spiked lake water solution with the purpose to achieve a higher accumulation efficiency of this radionuclide by means of the mixed sorbent bed and the higher removal efficiency at the same time.

The set of experiments was aimed at investigating whether the efficiency of ^{236}Pu removal from the lake water solution depends or not on the sorbent location in the column. For this purpose, five sorbents of equal slices were prepacked up to 100 mL in two separate columns in a different way. Column A at the bottom contained three sorbents of a lower sorption capacity for ^{236}Pu (Fig. 2), and above those sorbents, at the top of the column, two sorbents of a higher sorption capacity (moss and filter material) were placed. Slices of sorbents in the other column, column B, were placed in the order opposite to that in column A. The sorbents of a higher sorption capacity (moss and filter material) were placed at the column bottom and three sorbents of a lower sorption for plutonium (in Fig. 2 lichens, pine needles, granular activated carbon) were placed at the top of the column.

The lake water, spiked with ^{236}Pu, was leaked through the columns under definite experimental conditions. A radiochemical analysis was performed for the obtained filtrates and sorbents from two columns. It is necessary to point out that the sorbent beds before the analysis were divided into two segments/layers.

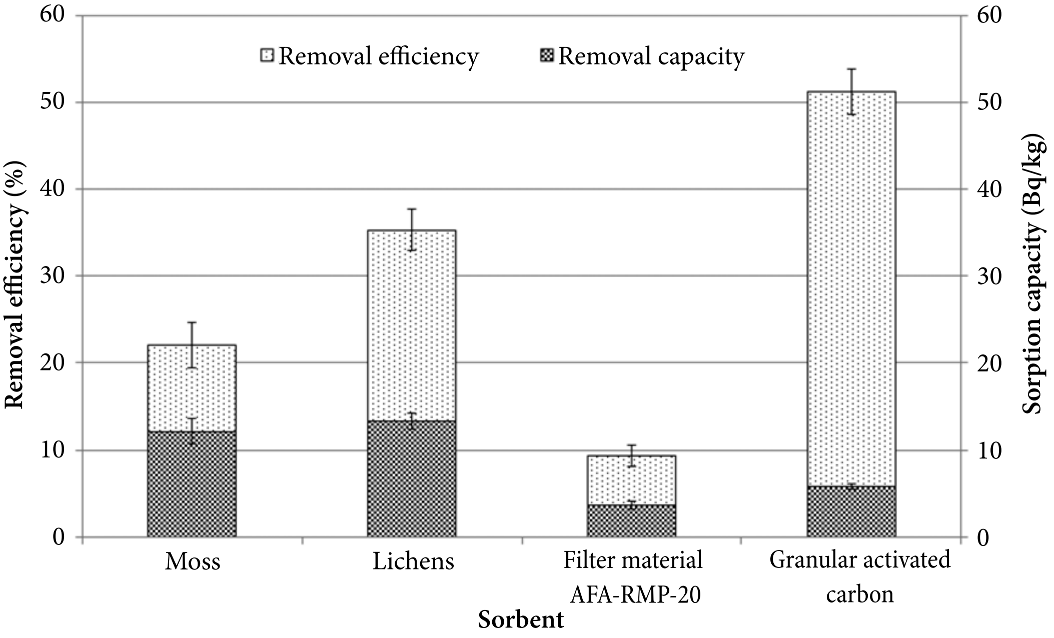

Fig. 3. Removal efficiency of ^{137}Cs and sorption capacity of environmental-friendly and synthetic sorbents from lake water spiked with ^{137}Cs (pH 6), using the dynamic flow column method.

The first layer of the sorbent bed of column A consisted of two slices (moss and filter material), whereas the second layer included three slices (lichens, pine needles, granular activated carbon). The layers of the mixed sorbent bed of column B were packed in the reverse order: lichens, pine needles and granular activated carbon in the first layer, and moss and filter material in the second one.

The filtration of the prepared lake water solution, spiked with ^{236}Pu, through column A started with sorbents of a higher sorption capacity, while through column B it started from sorbents with a lower sorption capacity for plutonium. The calculations of the removal efficiency (Eq. (1)) showed that within the error limits the sorbents of a higher capability (moss and filter material), being at the top of the column (A) or at the bottom (B) compared to the sorbent layer of lichens, pine needles and granular activated carbon, sorbed ^{236}Pu better, and the removal efficiency was significantly higher (Fig. 4).

The sorbent layer of lichens, pine needles and granular activated carbon were found to be worse sorbents of plutonium from the unfiltered lake water at pH of 6 compared to those of moss and filter material. The experimental results proved that the sorbent feature to have a higher or lower sorption capacity and the removal efficiency were independent of the order of sorbent packing in the column (Fig. 4).

3.2.3. ^{137}Cs desorption experiments from moss and lichen

Laboratory-scale batch type desorption experiments were performed to evaluate the ^{137}Cs background activity desorption from moss and lichen. The moss mass for the analysis was 9.07 g with the background ^{137}Cs activity concentration of 38.6±1.5 Bq/kg. The lichen mass was 13.89 g with the background ^{137}Cs activity concentration of 9.7±0.59 Bq/kg. The volume of the solution applied in the batch experiments was 450 mL. Since the initial ^{137}Cs activity concentration of moss and lichen samples was known, the radionuclide desorption/removal efficiency was evaluated applying Eq. (2).

The volume of distilled water decreased after its agitation with moss and lichen for 20 h. According to the calculations, 119 mL of distilled water was soaked in the moss and 68 mL of water penetrated into the lichen. The percentage of the ^{137}Cs desorption/removal efficiency from moss and lichen probably depends on the volume of water, which filled sorbents and removed ^{137}Cs (Table 3). To understand

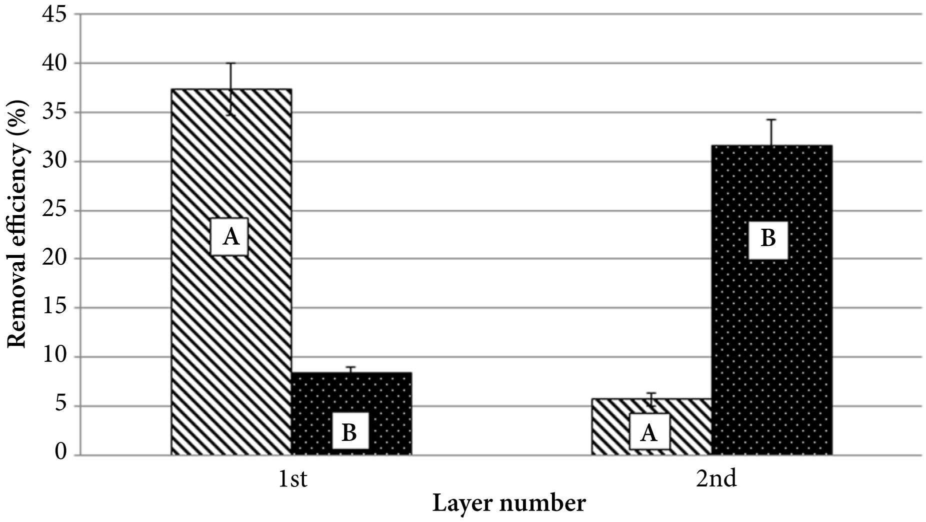

Fig. 4. Removal efficiency of ^{236}Pu by mixed sorbents: the upper (1st layer) and bottom (2nd layer) segments of sorbents prepacked in columns A and B in a different way. Filtration takes place beginning with sorbents of the highest sorption capability and ending with the lowest capability sorbents (A), and vice versa (B).

Table 3. Data of the batch type desorption of ^{137}Cs in distilled water at 100 rpm agitation for 20 h.

Sorbent	Solid phase mass, g	Solution volume after experiment, L	^{137}Cs activity after 20 h batch experiment		Removal efficiency, %
			In solid phase, Bq/kg	In solution, Bq/L	
Moss	9.07±0.05	0.331	24.26±1.32	0.39±0.027	37.1
Lichen	13.89±0.05	0.382	7.85±0.58	0.68±0.013	19.3

sorption/desorption processes better more extensive experiments are foreseen in the nearest future.

3.2.4. Assessment of the distribution (partition) coefficient K_d in tested sorbents

A generalizing approach on the use of the batch technique and flow-through method for K_d value assessment is presented in [34]: "Batch tests are commonly used to measure K_d values. The primary advantage of the batch method is that such experiments can be completed rapidly for a wide variety of elements and chemical environments. The primary disadvantage of the batch technique for measuring K_d is that it does not necessarily reproduce the chemical reaction conditions that take place in the real environment. Flow-through column experiments are intended to provide a more realistic simulation of dynamic field conditions and it is the second most common method of determining K_d values. The basic experiment is completed by passing a liquid spiked with the contaminant of interest through a sorbent column. The column experiment combines the chemical effects of sorption and the hydrologic effects of groundwater flow through a porous medium to provide an estimate of retarded movement of the contaminant of interest".

Particularly higher values of the distribution coefficients of most of the studied radionuclides between solid and liquid phases are obtained by the static method. The dynamic column method (open system) seems to be more reliable and its results can be used for characterization of radionuclide transport parameters. Values of distribution coefficients K_d, stat (static batch method), and K_d, dyn (dynamic column method) of ^{137}Cs for the same sorbents were obtained to be significantly different. For example, the following differences in the distribution coefficient (cm^3/g) values were determined: K_d, stat 610; 410; 30 while K_d, dyn 170; 85; 10 [35].

Based on the distribution coefficient values, two types of organoclay (>2.8×10^3 mL/g; >1.23×10^3 mL/g), illite (>1.17×10^4 mL/g), chabazite (>4.0×10^4 mL/g) and surfactant modified chabazite (>6.59×10^3 mL/g) were determined to be the most effective sorbents for ^{137}Cs [36]. The Pu(IV) K_d values from 10000 to 40000 mL/g have been reported for smectite-rich sediments in the pH range of 5–12 [37].

In this study K_d values for environmental-friendly sorbents and synthetic ones were calculated applying Eq. (4). Experimental K_d values were obtained using the dynamic flow column method or batch tests. They are presented in Table 4.

Table 4. Distribution (partition) coefficient K_d values for tested sorbents.

Radionuclide	Sorbent and method used	K_d, L/kg
^{236}Pu	Clay (dynamic flow method)	516.9
	Moss (dynamic flow method)	55.8
	Mycorrhizal fungi chanterelles (dynamic flow method)	14.2
	Lichens (dynamic flow method)	22.5
	Filter material AFA-RMP-20 (dynamic flow method)	63.8
	Pine needles (dynamic flow method)	12.4
	Granular activated carbon (dynamic flow method)	37.6
^{137}Cs	Moss (dynamic flow method)	15.5
	Lichens (dynamic flow method)	20.6
	Filter material AFA-RMP-20 (dynamic flow method)	4.0
	Granular activated carbon (dynamic flow method)	11.8
	Moss (batch tests)	28.8
	Lichen (batch tests)	7.7

3.3. Application of the dynamic moss bed column method in situ

The laboratory-scale experiments have shown that moss is the best sorbent among the tested environmental-friendly sorbents. In order to apply the moss bed column for ^{137}Cs and plutonium isotope pre-concentration from the lake water, a two column equipment was arranged on the shore of Lake Akmena. One column was able to leak water with a flow rate of 0.025 L/s and the other with a rate of 0.05 L/s. The lake water of 300 L volume leaked through each column. It needs to be stated that moss, used as a sorbent, has its own background activity. ^{137}Cs, ^{239,240}Pu and ^{238}Pu activity concentrations at zero time indicate radionuclide background activity concentrations in the moss bed (Fig. 5).

Moss taken out from the columns was at first prepared for the γ-spectrometric measurement, and then a plutonium radiochemical analysis was carried out. The results obtained after the moss bed analysis are presented in Fig. 5.

It is obviously seen that the background activity concentration of ^{137}Cs decreased with time, whereas those of ^{239,240}Pu and ^{238}Pu increased. According to the results (Fig. 5), during 100 min the flowing lake water of 300 L volume removed 35.6% of the background

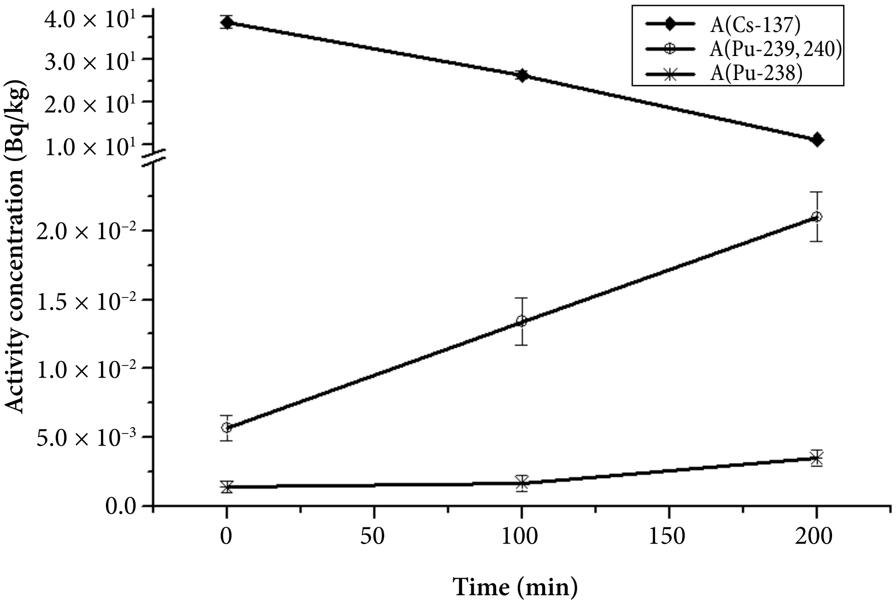

Fig. 5. Time-dependent changes in ^{137}Cs, ^{239,240}Pu and ^{238}Pu activity concentration in the moss bed of 14 cm cm height in the column during dynamic flow method experiments *in situ*. ^{137}Cs, ^{239,240}Pu and ^{238}Pu activity concentrations at zero time indicate radionuclide background activity concentrations in the moss bed.

^{137}Cs activity of moss while during 200 min 71.2% of the background ^{137}Cs activity was removed, meaning that cesium was desorbed at a much faster rate than sorbed to the moss bed.

The results of the measured ^{239,240}Pu and ^{238}Pu activity showed that the single moss bed in the column had sorbed these radionuclides from the lake water. As the lake water flow rate increased from 0.025 to 0.05 L/s, the exhaustion time reduced from 200 to 100 min. Considering the limited contact time of the radionuclide with the biosorbent bed, and the limitation in the radionuclide diffusion from the liquid to the solid phase at a higher flow rate, the dynamic uptake/sorption capacity of the moss bed decreased. The ^{239,240}Pu activity sorbed during 100 min was 0.0134±0.0017 Bq/kg and during 200 min it came up to 0.021±0.0018 Bq/kg, while the ^{238}Pu activity sorbed was 0.0017±0.00057 Bq/kg and 0.0035±0.00057 Bq/kg, respectively. According to the α-spectrometric measurements, if equilibrium conditions were achieved and all plutonium was sorbed from 300 L of the lake water during 200 min, the calculated activity concentration of ^{239,240}Pu in the unfiltered lake water was 4.87±0.98 mBq/m^3 and that of ^{238}Pu was 0.67±0.21 mBq/m^3. The activity concentration of ^{239,240}Pu in the unfiltered surface lake water determined in this study is quite consistent with that in lake water in Finland [23]; the average activity concentration of ^{239,240}Pu in the unfiltered water of Lake Päijänne, Finland, was 4.9±0.9 mBq/m^3 in 2007.

4. Conclusions

The present study has shown that according to the removal efficiency results, moss can be considered as the best sorbent for plutonium of the tested environmental-friendly sorbents, whereas the moss sorption capacity exceeds that of the environmental-friendly sorbents and the tested synthetic ones. The highest efficiency of ^{137}Cs removal from the lake water solution was obtained for granular activated carbon, and lichen showed higher sorption capacity parameters for ^{137}Cs compared to those of moss and synthetic sorbents. The experimental results proved that the sorbent feature to have higher or lower sorption capacity and the removal efficiency are independent of the sorbent placing in the column when different kinds of sorbents are sliced. The removed ^{137}Cs background activity from moss and lichen during the batch type experiments indicates the possibility to extract background radionuclides and other contaminants and probably to increase their sorption capability.

Application of the dynamic flow method carried out *in situ* confirmed the reversibility of the sorbed background ^{137}Cs activity in the moss observed during the batch type experiments. The flowing lake water of 300 L volume through the moss bed column desorbed 35.6 and 71.2% of ^{137}Cs background activity, respectively, within 100 and 200 min. Hence, a single moss bed column is not a promising sorbent for ^{137}Cs pre-concentration from fresh water bodies. However, application of the same single moss bed column for the first time *in situ* allowed us to determine the activity concentration of ^{239,240}Pu 4.87±0.98 and ^{238}Pu 0.67±0.21 mBq/m^3 in the lake water.

To better understand the sorption phenomenon and to apply a potentially useful technology in many areas, a series of experiments are planned to be implemented in the future.

Acknowledgements

This research was funded by a Grant (No. MIP-041/2012) from the Research Council of Lithuania. We are grateful to Prof. S. Tautkus from the Department of Analytical and Environmental Chemistry of Vilnius University for the measurement of macro- and microelements in the environmental assays tested in our study, and R. Gvozdaitė, Junior Scientific Researcher from the State Research Institute Center for Physical Sciences and Technology, for α-spectrometric measurements. We would like to thank two reviewers, whose comments helped us to improve the manuscript.

References

[1] D. Marčiulionienė, D. Kiponas, and D. Hansen, Accumulation of technogenic radionuclides in the environment of Ignalina NPP, Ekologija **1**, 52–59 (2001) [in Lithuanian].

[2] J. Mažeika, *Radionuclides in Geoenvironment of Lithuania* (Institute of Geology, Vilnius, 2002).

[3] V. Mironov, V. Kudrjashov, F. Yiou, and G.M. Raisbeck, Use of ^{129}I and ^{137}Cs in soils for the estimation of ^{131}I deposition in Belarus as a result of the Chernobyl accident, J. Environ. Radioact. **59**, 293–307 (2002), http://dx.doi.org/10.1016/S0265-931X(01)00080-7

[4] V. Remeikis, R. Gvozdaitė, R. Druteikienė, A. Plukis, N. Tarasiuk, and N. Špirkauskaitė, Plutonium and americium in sediments of Lithuanian lakes, Nukleonika **50**(2), 61–66 (2005).

[5] D. Butkus and M. Konstantinova, Studies of long-term vertical migration of ^{137}Cs in soil, Environ. Chem. Phys. **25**, 75–80 (2003).

[6] P. Carbol, D. Solatie, N. Erdmann, T. Nylèn, and M. Betti, Deposition and distribution of Chernobyl fallout fission products and actinides in a Russian soil profile, J. Environ. Radioact. **68**(1), 27–46 (2003), http://dx.doi.org/10.1016/S0265-931X(03)00027-4

[7] B. Lukšienė, R. Druteikienė, R. Gvozdaitė, and A. Gudelis, Comparative analysis of Pu-239, Cs-137, Pb-210 and K-40 spatial distributions in the top soil layer at the Baltic coast, J. Environ. Radioact. **87**(3), 305–314 (2006), http://dx.doi.org/doi:10.1016/j.jenvrad.2005.12.005

[8] R. Druteikienė, R. Morkūnienė, and B. Lukšienė, Distribution of artificial radionuclides in the Baltic seaside environment, Lith. J. Phys. **51**(1), 75–81 (2011), http://dx.doi.org/10.3952/lithjphys.51107

[9] B. Lukšienė, E. Maceika, N. Tarasiuk, E. Koviazina, V. Filistovič, Š. Buivydas, and A. Puzas, On peculiarities of vertical distribution of ^{239,240}Pu, ^{238}Pu and ^{137}Cs activity concentrations and their ratios in lake sediments and soils, J. Radioanal. Nucl. Chem. **300**, 277–286 (2014), http://dx.doi.org/10.1007/s10967-014-3026-0

[10] L. Popov, G. Mihailova, and I. Naidenov, Determination of activity ratios of $^{238,239+240,241}$Pu, ^{241}Am, ^{134,137}Cs, and ^{90}Sr in Bulgarian soils, J. Radioanal. Nucl. Chem. **285**, 223–237 (2010), http://dx.doi.org/10.1007/s10967-010-0550-4

[11] P.P. Povinec, I. Sýkora, K. Holý, M. Gera, A. Kováčik, and L. Bresťáková, Aerosol radioactivity record in Bratislava/Slovakia following the Fukushima accident – A comparison with global fallout and the Chernobyl accident, J. Environ. Radioact. **114**, 81–88 (2012), http://dx.doi.org/10.1016/j.jenvrad.2012.05.008

[12] B. Lukšienė, A. Puzas, V. Remeikis, R. Druteikienė, A. Gudelis, R. Gvozdaitė, Š. Buivydas, R. Davidonis, and G. Kandrotas, Spatial patterns and ratios of ^{137}Cs, ^{90}Sr, and Pu isotopes in the top layer of undisturbed meadow soils as indicators for contamination origin, Environ. Monit. Assess. **187**, 268 (2015), http://dx.doi.org/10.1007/s10661-015-4491-9

[13] UNSCEAR, *Ionization Radiation: Sources and Biological Effects*. Report to the General Assembly, with annexes (UNSCEAR, New York, NY, 1982).

[14] S. Babel and T.A. Kurniawan, Low-cost adsorbents for heavy metals uptake from contaminated water: a review, J. Hazard. Mater. **97**(1–3), 219–243 (2003), http://dx.doi.org/10.1016/S0304-3894(02)00263-7

[15] F.E. Soetaredjo, A. Kurniawan, L.K. Ong, D.R. Widagdyo, and S. Ismadji, Investigation of the continuous flow sorption of heavy metals in a biomass-packed column: revisiting the Thomas design model for correlation of binary component systems, RSC Adv. **4**, 52856–52870 (2014), http://dx.doi.org/10.1039/C4RA06425A

[16] M.A. Barakat, New trends in removing heavy metals from industrial wastewater, Arabian J. Chem. **4**(4), 361–377 (2011), http://dx.doi.org/10.1016/j.arabjc.2010.07.019

[17] J. Wang and C. Chen, Biosorbents for heavy metals removal and their future, J. Biotech. Adv. **27**(2), 195–226 (2009), http://dx.doi.org/10.1016/j.biotechadv.2008.11.002

[18] M. Sravani, M. Manasa, V. Sridevi, and M.V.V. Chandana Lakshmi, A competitive effect of Cu (II) and Pb (II) on biosorption by Aspergillus Niger NCIM (616) using ICPMS, Res. J. Pharm. Biol. Chem. Sci. **3**(4), 1408–1416 (2012).

[19] B. Volesky and Z.R. Holan, Biosorption of heavy metals, Biotechnol. Progr. **11**(3), 235–250 (1995), http://dx.doi.org/10.1021/bp00033a001

[20] D. Park, Y.S. Yun, and J.M. Park, The past, present, and future trends of biosorption, Biotech. Bioprocess Eng. **15**(1), 86–102 (2010), http://dx.doi.org/10.1007/s12257-009-0199-4

[21] G. Crini and P.M. Badot, Application of chitosan, a natural aminopolysaccharide, for dye removal by aqueous solutions by adsorption processes using batch studies: a review of recent literature, Prog. Polym. Sci. **33**, 399–447 (2008), http://dx.doi.org/10.1016/j.progpolymsci.2007.11.001

[22] M. Eriksson, E. Holm, P. Roos, and H. Dahlgaard, Distribution and flux of ^{238}Pu, ^{239,240}Pu, ^{241}Am, ^{137}Cs and ^{210}Pb to high arctic lakes in the Thule district (Greenland), J. Environ. Radioact. **75**(3), 285–299 (2004), http://dx.doi.org/10.1016/j.jenvrad.2003.12.007

[23] M. Lusa, J. Lehto, A. Leskinen, and T. Jaakkola, ^{137}Cs, ^{239,240}Pu and ^{241}Am in bottom sediments and surface water of Lake Päijänne, Finland, J. Environ. Radioact. **100**(4), 468–476 (2009), http://dx.doi.org/10.1016/j.jenvrad.2009.03.006

[24] A. Moisejenkova, N. Tarasiuk, E. Koviazina, E. Maceika, and A. Girgždys, ^{137}Cs in lake Tapeliai, Lithuania, Lith. J. Phys. **52**(3), 238–252 (2012), http://dx.doi.org/10.3952/lithjphys.52308

[25] N. Tarasiuk, E. Koviazina, and V. Kubarevičienė, On seasonal variations of radiocesium speciation in the surface sediments of Lake Juodis, Lithuania, J. Environ. Radioact. **99**(1), 199–210 (2008), http://dx.doi.org/10.1016/j.jenvrad.2007.10.014

[26] V. Putyrskaya, E. Klemt, and S. Röllin, Migration of ^{137}Cs in tributaries, lake water and sediment of Lago Maggiore (Italy, Switzerland) – analysis and comparison with Lago di Lugano and other lakes, J. Environ. Radioact. **100**(1), 35–48 (2009), http://dx.doi.org/10.1016/j.jenvrad.2008.10.005

[27] P. Kalač, A review of chemical composition and nutritional value of wild-growing and cultivated mushrooms, J. Sci. Food Agric. **93**(2), 209–218 (2013), http://dx.doi.org/10.1002/jsfa.5960

[28] M. Rinaudo, Chitin and chitosan: Properties and applications, Prog. Polym. Sci. **31**, 603–632 (2006), http://dx.doi.org/10.1016/j.progpolymsci.2006.06.001

[29] V.N. Kosyakov, N.G. Yakovlev, and I.E. Veleshko, Application of chitin-containing fiber material "mycoton" for actinide adsorption, J. Nucl. Sci. Technol. **39**(3), 508–511 (2002), http://dx.doi.org/10.1080/00223131.2002.10875518

[30] M. Riazi, A.R. Keshtkar, and M.A. Moosavian, Batch and continuous fixed-bed column biosorption of thorium (IV) from aqueous solutions: equilibrium and dynamic modeling, J. Radioanal. Nucl. Chem. **301**(2), 493–503 (2014), http://dx.doi.org/10.1007/s10967-014-3129-7

[31] A.F. Seliman, Y.F. Lasheen, M.A.E. Youssief, M.M. Abo-Aly, and F.A. Shehata, Removal of some radionuclides from contaminated solution using natural clay: bentonite, J. Radioanal. Nucl. Chem. **300**(3), 969–979 (2014), http://dx.doi.org/10.1007/s10967-014-3027-z

[32] A. Lerman, D.M. Imboden, and J.R. Gat, *Physics and Chemistry of Lakes*, 2nd ed. (Springer-Verlag, Berlin, Heidelberg, 1995) p. 333, http://dx.doi.org/10.1007/978-3-642-85132-2

[33] G.N. Pshinko, L.N. Puzyrnaya, S.A. Kobets, V.M. Fedorova, A.A. Kosorukov, and V.Ya. Demchenko, Layered double hydroxide of Zn and Al, intercalated with hexacyanoferrate(II) ions, as a sorbent for removing cesium radionuclides from aqueous solutions, Radiochem. **57**(3), 259–265 (2015), http://dx.doi.org/10.1134/S1066362215030066 [in Russian].

[34] K.M. Krupka, D.I. Kaplan, G. Whelan, R.J. Serne, and S.V. Mattigod, *Understanding Variation in Partition Coefficient, K_d, Values. Vol. II: Review of Geochemistry and Available K_d Values for Cadmium, Cesium, Chromium, Lead, Plutonium, Radon, Strontium, Thorium, Tritium (^{3}H) and Uranium*, EPA 402-R-99-0048 (Office of Air and Radiation, Office of Solid Waste and Emergency Response, U.S. Environmental Protection Agency, Washington, D.C., 1999), http://www.epa.gov/radiation/technology/partition.htm

[35] Š. Palágyi and H. Vodičková, Sorption and desorption of ^{125}I$^-$, ^{137}Cs$^+$, ^{85}Sr^{2+} and ^{152,154}Eu^{3+} on disturbed soils under dynamic flow and static batch conditions, J. Radioanal. Nucl. Chem. **280**(1), 3–14 (2009), http://dx.doi.org/10.1007/s10967-008-7436-8

[36] D. Li, D.I. Kaplan, A.S. Knox, K.P. Crapse, and D.P. Diprete, Aqueous ^{99}Tc, ^{129}I and ^{137}Cs removal from contaminated groundwater and sediments using highly effective low-cost sorbents, J. Environ. Radioact. **136**, 56–63 (2014), http://dx.doi.org/10.1016/j.jenvrad.2014.05.010

[37] G. Lujanienė, S. Motiejūnas, and J. Šapolaitė, Sorption of Cs, Pu and Am on clay minerals, J. Radioanal. Nucl. Chem. **274**(2), 345–353 (2007), http://dx.doi.org/10.1007/s10967-007-1121-1

^{137}Cs IR PLUTONIO IZOTOPŲ SORBCIJOS-DESORBCIJOS BIOLOGINĖSE IR SINTETINĖSE MEDŽIAGOSE TYRIMAS

B. Lukšienė, Z. Žukauskaitė, N. Tarasiuk, E. Maceika, V. Filistovič, Š. Buivydas, L. Nedzveckienė, D. Jasinevičienė

Fizinių ir technologijos mokslų centras, Vilnius, Lietuva

Santrauka

Darbe pristatomi radionuklidų (^{137}Cs ir plutonio izotopų) sukoncentravimo iš gėlo vandens kietose matricose tyrimai. Ypatingas dėmesys skirtas inovatyviam fizikiniam-cheminiam radionuklidų pašalinimo iš vandeninės terpės procesui – adsorbcijai, naudojant naujo tipo sorbentus, gamtines medžiagas. Tyrimai atlikti į ežero vandens tirpalą įterpus ^{137}Cs ir ^{236}Pu izotopus. Ežero vandenyje nustatyti pagrindiniai anijonai, mikro ir makroelementai, pritaikant jonų ir atominės absorbcijos chromatografijos metodus. Radionuklidų sorbcijos-desorbcijos geba buvo įvertinta pagal α- ir γ-spektrometrinių matavimų duomenis. Gamtinių ir sintetinių sorbentų sorbcinė geba buvo palyginta taikant statinius (purtant) ir dinaminius kolonėlių (pratekant) metodus.

Pagal pašalinimo iš tirpalo efektyvumo rezultatus, iš visų tirtųjų gamtinių medžiagų, išskyrus molį, samanos gali būti laikomos geriausiu plutonio sorbentu, o samanų sorbcinė geba buvo didesnė net už sintetinius sorbentus. Didžiausiu ^{137}Cs iš ežero vandens tirpalo pašalinimo efektyvumu pasižymėjo granuliuotos aktyvintos anglies kolonėlės.

Pritaikius dinaminį metodą, filtruojant per samanų kolonėles natūralų ežero vandenį *in situ*, pasitvirtino statiniu metodu laboratorijoje gauti rezultatai apie ^{137}Cs grįžtamąją sorbciją samanose. Priešingai negu ^{137}Cs, toje pačioje samanų kolonėlėje plutonio izotopai buvo sorbuojami iš ežero vandens. Pagal samanų kolonėlėje akumuliuotų radionuklidų α-spektrometrinius matavimus po radiocheminės analizės nustatyta, kad $^{239,\,240}$Pu aktyvumo koncentracija ežero vandenyje yra 4,87±0,98 mBq/m^3, o ^{238}Pu – 0,67±0,21 mBq/m^3.

Gauti skirtingi ^{137}Cs ir plutonio sorbcijos rezultatai, naudojant samanas kaip sorbentą, nurodo skirtingas šių radionuklidų prisijungimo prie samanų savybes, todėl numatomi tolimesni adsorbcijos tyrimai.