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Plutonium and americium in sediments of Lithuanian lakes

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Abstract The assessment of contribution of the global and the Chernobyl NPP (Nuclear Power Plant) accident plutonium and americium to plutonium pollution in sediments of Lithuanian lakes is presented. Theoretical evaluation of activity ratios of $^{238}Pu/^{239+240}Pu$ and $^{241}Pu/^{239+240}Pu$ in the reactor of unit 4 of the Chernobyl NPP before the accident was performed by means of the ORIGEN-ARP code from the SCALE 4.4A program package. Non-uniform distribution of radionuclides in depositions on the Lithuanian territory after nuclear weapon tests and the Chernobyl NPP accident is experimentally observed by measuring the lake sediment pollution with actinides. The activity concentration of sediments polluted with plutonium ranges from 2.0 ± 0.5 Bq/kg d.w. (dry weight) in Lake Asavėlis to 14 ± 2 Bq/kg d.w. in Lake Juodis. The ratio of activity concentrations of plutonium isotopes $^{238}Pu/^{239+240}Pu$ measured by α-spectrometry in the 10-cm-thick upper layer of bottom sediment varies from 0.03 in Lake Juodis to 0.3 in Lake Žuvintas. The analysis of the ratio values shows that the deposition of the Chernobyl origin plutonium is prevailing in southern and south-western regions of Lithuania. Plutonium of nuclear weapon tests origin in sediments of lakes is observed on the whole territory of Lithuania, and it is especially distinct in central Lithuania. The americium activity due to ^{241}Pu decay after the Chernobyl NPP accident and global depositions in bottom sediments of Lithuanian lakes has been evaluated to be from 0.9 to 5.7 Bq/kg.

Key words plutonium isotopes • americium • sediments • isotopic composition

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Introduction

Plutonium and americium elements of the actinide family are most dangerous among nuclides from the point of view of radioecology and radiation safety. Plutonium is the most widespread element among transuranic elements represented by four isotopes (^{238}Pu, ^{239}Pu, ^{240}Pu and ^{241}Pu). The contribution of these isotopes to plutonium pollution on the Lithuanian territory before the Chernobyl NPP accident was global fallout (especially due to the nuclear weapon tests in Novaya Zemlya, the North of Russia) [7]. In the pre-Chernobyl time, there were no measurements of plutonium activity concentration in the environment of Lithuania or in the environment of Poland either. According to [4], the deposition of $^{239+240}Pu$ due to the global fallout on the territories of Denmark and Germany, was 38–63 Bq/m^2 and that of ^{238}Pu – about 1.2–2.8 Bq/m^2. During passage of the Chernobyl cloud, deposition of plutonium covered the most part of southern and south-western Lithuania [6]. After the Chernobyl NPP accident, the activity concentration of $^{239+240}Pu$ on the Lithuanian territory was measured in soil (0.05–1.30 Bq/kg d.w.), in moss (0.2–8.4 Bq/kg d.w.) [2], in bottom sediments of the Curonian Lagoon (0.04–1.70 Bq/kg d.w.), and of Kaunas man-made

reservoir (The Nemunas River) (0.09–0.90 Bq/kg d.w.) [9]. The values of $^{238}Pu/^{239+240}Pu$ activity ratio ranged from 0.34 to 0.45 in soil of Lithuania [1]. The determined activity concentrations of $^{239+240}Pu$ were 3.13–5.46 Bq/kg d.w. in sediments in the Gulf of Gdansk with a maximal value of 7.2 ± 0.6 Bq/kg d.w. in the Gdansk Deep and ranged from 0.72 to 1.17 Bq/kg d.w. in the Bornholm Basin, the Baltic Sea [17]. The average plutonium activity concentration was 0.93 Bq/kg d.w. in sandy areas and it was 6.50 Bq/kg d.w. in silty areas in the Gdansk Basin [16]. After a long period of global fallout of nuclear weapon tests and the Chernobyl NPP accident products, bottom sediments of the majority of lakes in Europe turned into a real repository of radionuclides. Furthermore, in the course of time when radionuclide run-off from drainage basins of water bodies became negligible due to their constant deepening into soil, sediment accumulation zones in running lakes became the source of the secondary contamination of the outflowing rivers.

The precise information about plutonium sources in the environment according to the activity ratios only of plutonium isotopes or those with other radionuclides can be obtained. The $^{238}Pu/^{239+240}Pu$ activity ratio values in global fallout from nuclear weapon tests and the satellite SNAP-9 catastrophe ranged from 0.024 to 0.041 [4], whereas the same ratio in the Chernobyl fallout was reported to be equal to 0.3 ± 0.05 [13], 0.55 [11] or 0.65 [14]. At present, ^{241}Am ($T_{1/2}$ = 432 y) activity concentration increases in all components of the environment, where plutonium is accumulated. Therefore, one more issue should be taken into consideration, i.e. the product of β decay of short-lived plutonium isotope ^{241}Pu ($T_{1/2}$ = 14.35 y). 90% of global fallout of ^{241}Pu and ~50% of the Chernobyl NPP accident fallout of ^{241}Pu both decayed to ^{241}Am.

The aim of this study was to assess the sediment pollution with actinides in the six selected for the study Lithuanian lakes and to determine the origin of actinides.

Materials and methods

Sampling

Six shallow running lakes from different parts of Lithuania were selected for the study. Sampling sites are presented in Fig. 1. Lakes Šventas and Asavėlis are in the 30-km zone of the Ignalina NPP (northern part of Lithuania), while Lake Juodis is in the vicinity of Vilnius city (eastern part of Lithuania). Lakes Žuvintas and Obelija represent southern part of Lithuania and Lake Germantas is in western part of Lithuania. Samples of sediments were collected in all six lakes on March 3–5, 1999 and they were taken with a special sediment sampler by cutting the 15×15 cm area and taking a 10-cm-thick upper layer of bottom sediments. For the determination of plutonium origin in selected six Lithuanian lakes, 10-cm thickness bottom sediment samples were taken, while for experimental evaluation of ^{241}Am concentration and their comparison with the calculated ones (from ^{241}Pu), bottom sediment samples were taken from deeper layers (depth 4–6 cm from sediment surface) in three lakes (Asavėlis, Žuvintas, Juodis).

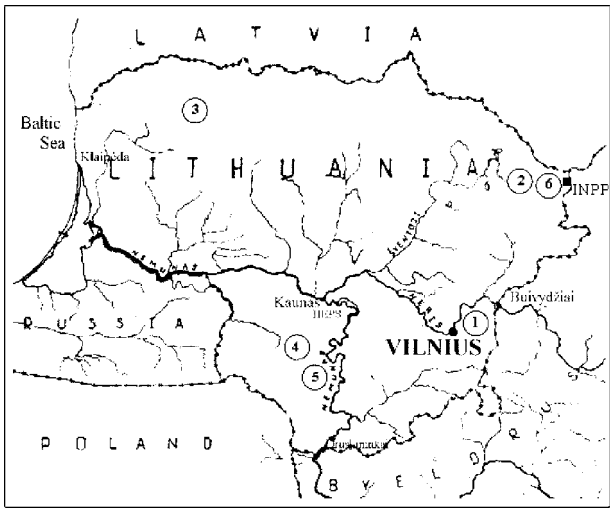

Fig. 1. Scheme of lake sampling sites: 1 – Juodis; 2 – Asavas-Asavėlis; 3 – Germantas; 4 – Žuvintas; 5 – Obelija; 6 – Šventas; ■ INPP – Ignalina NPP.

Analytical methods

Plutonium concentrations were analyzed by the α-spectrometry method with particular attention given to plutonium separation from ^{241}Am, whose α line overlaps the one of ^{238}Pu. The ion-exchange chromatography method, enabling extraction of trace amounts of the investigated radionuclides and distinguished for extreme selectivity, was chosen [1, 5]. For determination of the plutonium activity concentration, a dry bottom sediments sample was heated in a muffle furnace at 700°C for 2 hours. After adding ^{242}Pu and ^{243}Am as radiochemical yield tracers, the sediment sample was digested repeatedly with 8 mol/L HNO_3. Plutonium isotopes were separated and purified by radiochemical separation using a strong basic anion exchange resin DOWEX 1×8. Then it was electrodeposited on a stainless-steel disk from Na_2SO_4/H_2SO_4 electrolyte solution for 1 h using a current density of 0.6 A/cm^2 [18].

Eluate was collected from a DOWEX 1×8 resin column. Americium isotopes were purified by oxalic acid precipitation and radiochemical separation on an anion exchange resin DOWEX 1×4 and on a cation exchange resin DOWEX 50W × 8. Then, americium isotopes were electroposited in the same way as those of plutonium. This method ensures that americium and plutonium are extracted with an uncertainty less than 10%. The radiochemical yield of ^{242}Pu and ^{243}Am varied from 50% to 70%. The α spectrometric measurement chain consists of a CANBERRA PD detector (area – 450 mm^2, resolution is 19 keV (FWHM) at 4–6 MeV) and a SES-13 spectrometer. The α counting efficiency of the detector is 25%, the detection limit of $^{239+240}Pu$ and of ^{241}Am for α counting time of 86,400 s is about 10^{-3} Bq.

Modeling

In the presented study, the impact of the global and local sources of plutonium on the environment radioactive pollution is evaluated using the methods presented in [11]. A simple set of equations is used to separate plutonium of the Chernobyl or global origin from the total plutonium fallout:

$$(1) \qquad \begin{aligned} A_{239+240\mathrm{Pu}} &= A_g + A_{\mathrm{Ch}} \\ A_{238\mathrm{Pu}} &= \zeta A_g + \xi A_{\mathrm{Ch}}, \end{aligned}$$

where: $A_{239+240\mathrm{Pu}}$ and $A_{238\mathrm{Pu}}$ are the measured activity concentration values of plutonium isotopes in a sample; A_g and A_{Ch} are the $^{239+240}$Pu activity concentrations caused by the global and Chernobyl pollution, respectively; ζ and ξ are activity ratios of ^{238}Pu/$^{239+240}$Pu due to the global and Chernobyl pollution, respectively [10]. The solutions of Eq. (1) are as follows:

$$(2) \qquad \begin{aligned} A_{\mathrm{Ch}} &= (A_{238\mathrm{Pu}} - \zeta A_{239+240\mathrm{Pu}})(\xi - \zeta)^{-1} \\ A_g &= (\zeta A_{239+240\mathrm{Pu}} - A_{238\mathrm{Pu}})(\xi - \zeta)^{-1}. \end{aligned}$$

The percentage F of the Chernobyl fraction in the total activity can be defined as

$$(3) \qquad \begin{aligned} F &= \left[A_{\mathrm{Ch}} \left(A_{239+240\,\mathrm{Pu}} \right)^{-1} - \zeta \right] (\xi - \zeta)^{-1} \\ &= \left[\frac{A_{238\,\mathrm{Pu}}}{A_{238\,\mathrm{Pu}+239\,\mathrm{Pu}+240\,\mathrm{Pu}}} - \zeta \right] (\xi - \zeta)^{-1} \cdot 100\%. \end{aligned}$$

Time-dependent material concentrations were determined by using the SCALE 4.4A package code ORIGEN-ARP based on the point-depletion calculation utilizing the problem-dependent cross-section libraries. ORIGEN-ARP consists of the ARP code used to interpolate burnup-dependent cross-sections for all nuclides and the ORIGEN-S code which computes time-dependent concentrations and source terms of a large number of isotopes. Nuclides are simultaneously generated or depleted through the neutron transmutation, fission and radioactive decay. The following equation describes the actinide production:

$$(4) \qquad \begin{aligned} \frac{dN_i}{dt} &= \sigma_{c,i-1} N_{i-1} \Phi \sum_{i'} \lambda_{i'i} N_{i'} - \sigma_{c,i} N_i \Phi \\ &\quad - \sigma_{f,i} N_i \Phi - \lambda_i N_i, \end{aligned}$$

where: N_i is the number of nuclei of the i-th nuclide; $\sigma_{c,i}$ are the cross-sections of the neutron capture; $\sigma_{f,i}$ are the fission cross-sections; λ_i is a constant of the nuclide radioactive decay and Φ is the neutron flux in the reactor. In reality, the flux Φ (x,y,z;E;t) is dependent on the nuclide concentrations and the fuel burnup.

The analysis sequence SAS2 from the SCALE package was used for generating the burnup-dependent cross-section library. One reactor cell including fuel assembly (FA) and a 25 × 25 cm graphite column was simulated in cylindrical 1-D geometry. Fuel pins were arranged in the lattice cell geometry with the 1.6 cm triangular pitch. The cell averaged fuel, Zr + Nb cladding and, H_2O mixture, was distributed in a cylinder with the 0.75-cm inner diameter, the 3.95-cm outer diameter and the height of 686.2 cm. The graphite moderator was modeled as a cylinder with a 14.1-cm outer diameter and a density of 1.65 g/cm^3 around FA [15]. We used the SCALE 238-group neutron library based on ENDF-B/V data for calculation of neutron cross-sections.

The input parameters for the code ORIGEN were mass of uranium in FA, 114 kg, and masses of light elements: Zr – 39 kg, Nb – 1 kg, O – 15.3 kg, Fe – 2.3 kg. The initial uranium enrichment was 2.0% ^{235}U and the power per FA was 2 MW. The neutron capture and fission cross-section of the RBMK type reactor assembly were used in calculations. The coolant density ρ = 0.5 g/cm^3 was used. The axial power profile of FA in the RBMK core and distribution of FA burnup were taken into account for the calculation of actinides in irradiated nuclear fuel. In the calculations we used the average nuclear fuel burnup value of 10.9 MWd/kg of the Chernobyl NPP RBMK-1000 reactor [12].

Results and discussions

The α spectra of plutonium in sediment samples are presented in Fig. 2. The samples were taken from the 10-cm-thick upper layer of bottom sediments, i. e. from the sediment thickness which well represents the total pollution from the global fallouts as well as the Chernobyl NPP accident fallouts. In all α spectra, the ^{242}Pu (tracer), $^{239+240}$Pu and ^{238}Pu energy peaks of 4900, 5140 and 5441 keV, respectively, were registered. From the known amount of the introduced tracer ^{242}Pu, corresponding to 2 Bq/kg in the spectrum, ^{238}Pu and $^{239+240}$Pu activities in the sample and the activity ratio of these nuclides were calculated. An apparent difference in the plutonium amount and isotopic composition, indicating a different total radioactive pollution level and a different contribution of pollution sources, is observed. Non-uniform distribution of radionuclides in depositions on the Lithuanian territory after nuclear weapon tests and the Chernobyl NPP accident is experimentally observed by measuring the sediment pollution with plutonium and americium in the six lakes selected for this study. The activity concentration of sediments polluted with plutonium ranges from 2.0 ± 0.5 Bq/kg d.w. in Lake Asavėlis to 14 ± 2 Bq/kg d.w. in Lake Juodis. The ratio of activity concentrations of plutonium isotopes ^{238}Pu/$^{239+240}$Pu varies from 0.03 in Lake Juodis to 0.30 in Lake Žuvintas. The data show that plutonium fallout in bottom sediments in the studied lakes of Lithuania is from different sources.

First of all, referring to Eq. (3) and the experimental measurement values of ^{238}Pu and $^{239+240}$Pu activity concentrations, we calculated the contribution of plutonium of the Chernobyl NPP accident and of plutonium of nuclear weapon tests to the total activity concentration of this radionuclide.

The average activity ratios of ^{238}Pu to $^{239+240}$Pu isotopes in the reactor of unit 4 of the Chernobyl NPP before the accident were calculated. The obtained isotopic composition ratio value ξ = 0.51 (for the year 2000) with the experimental value obtained by analyzing α spectra of samples taken in the 10-km Chernobyl NPP

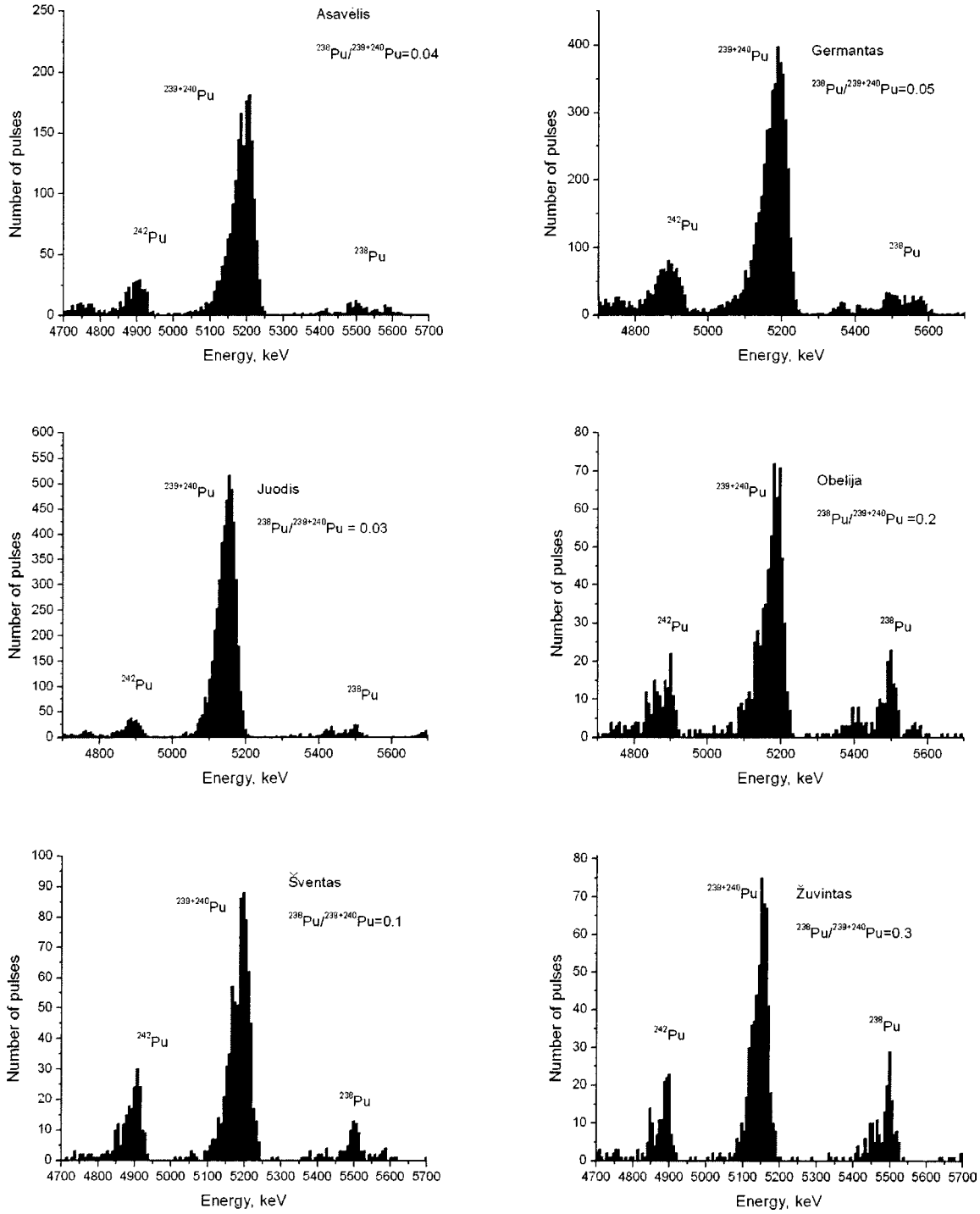

Fig. 2. α Spectra of plutonium in sediments of the Lithuanian lakes.

zone were compared. The experimental and theoretical values are coincident with the accuracy of 3%. The results of calculations are presented in the columns 3 and 4 of Table 1. The uncertainties of calculated ^{241}Am activity values in Table 1 have been calculated taking into account uncertainties of the experimentally measured plutonium activity. The ratios and uncertainties of plutonium isotopes activity for the Chernobyl and the global fallout $\xi = 0.51 \pm 0.03$ and $\zeta = 0.03 \pm 0.01$ for ^{238}Pu with $^{239+240}Pu$ and 50 ± 5 and 14 ± 3 for ^{241}Pu with $^{239+240}Pu$ have been used.

The next task was to calculate the ^{241}Am amount in bottom sediments of each lake referring to the plutonium activity concentration values. For this purpose, the ratio of ^{241}Pu activity to activities of $^{239+240}Pu$ isotopes measured by the α-spectrometry method was evaluated.

The ^{241}Pu (β emitter) amount of Chernobyl origin in samples (at the time of the accident) was calculated after theoretical evaluation of ratios of all plutonium isotopes in the reactor of unit 4 of the Chernobyl NPP. Using the above presented modeling it was obtained that the ratio of $^{241}Pu/^{239+240}Pu$ was equal to 50 ± 5.

Table 1. Activity concentrations of $^{239+240}$Pu, ^{238}Pu and ^{241}Am (Bq/kg) in bottom sediments of the lakes (experimental and calculated data; errors indicate standard deviations)

Lake	Experimental		Calculated				
	^{238}Pu	$^{239+240}$Pu	$^{239+240}$Pu (A_g)	$^{239+240}$Pu (A_{Ch})	^{241}Am (A_g)	^{241}Am (A_{Ch})	^{241}Am Total
Juodis	0.4 ± 0.2	13.6 ± 2.0	13.6 ± 2.0	0.0 ± 0.5	5.7 ± 1.2	0.0 ± 0.4	5.7 ± 1.3
Germantas	0.5 ± 0.2	9.3 ± 1.0	8.3 ± 1.1	1.0 ± 0.5	3.5 ± 0.7	0.8 ± 0.4	4.3 ± 0.8
Obelija	1.5 ± 0.3	7.5 ± 0.4	4.8 ± 0.7	2.7 ± 0.7	2.0 ± 0.5	2.2 ± 0.6	4.2 ± 0.8
Šventas	0.7 ± 0.2	7.3 ± 0.4	6.2 ± 0.6	1.1 ± 0.6	2.6 ± 0.6	0.9 ± 0.5	3.5 ± 0.8
Asavėlis	0.05 ± 0.02	1.9 ± 0.5	1.7 ± 0.5	0.2 ± 0.2	0.7 ± 0.2	0.2 ± 0.2	0.9 ± 0.3
Žuvintas	1.8 ± 0.5	6.2 ± 0.4	2.6 ± 1.0	3.5 ± 0.9	1.1 ± 0.4	3.0 ± 0.7	4.1 ± 0.8

The detailed analysis of plutonium isotope ratios in the RBMK reactor nuclear fuel is presented in [8]. The ratio of isotope activity concentration ^{241}Pu/$^{239+240}$Pu of the nuclear weapon tests origin was estimated experimentally. For experimental evaluation of ^{241}Am concentration and their comparison with the calculated ones (from ^{241}Pu), bottom sediment samples (depth 4–6 cm from the sediment surface) were analysed radiochemically and alpha spectrometrically. The obtained plutonium and americium spectra are shown in Fig. 3. The lines on the right side of the α spectrum correspond to tracers (^{242}Pu (4 Bq/kg) and ^{243}Am (1 Bq/kg)) introduced in the sample. The ratio of ^{238}Pu activity concentration to the one of $^{239+240}$Pu is 0.04 which shows that the sample well represents radioactive pollution of the nuclear weapon tests (Fig. 3a). According to the line intensity in the spectrum it has been determined that the activity concentrations of ^{241}Am and $^{239+240}$Pu are equal to 2.0 ± 0.2 Bq/kg and 4.5 ± 0.5 Bq/kg, respectively. Assuming that 90% of ^{241}Pu in sediments after the nuclear weapon tests decayed into the presently registered ^{241}Am, we calculated the initial amount of ^{241}Pu in fallout. By averaging the measurement results of samples taken in the lakes and adding about 10% of not yet decayed ^{241}Pu, we obtain that the ^{241}Pu/$^{239+240}$Pu ratio is 14 ± 3. This value rather well coincides with the 4.2 value presented in [10] for 1986 and the 4.0 value for 1988 [19]. Using the values of the activity concentration ratios ^{241}Pu/$^{239+240}$Pu determined theoretically and experimentally, corresponding to the Chernobyl and the global plutonium pollution, the americium concentrations in bottom sediments of Lithuanian lakes were calculated. The results evaluating the influence of both the Chernobyl and global pollution are presented in the columns 6 and 7 of Table 1. The total ^{241}Am activity concentration in bottom sediments of the lakes is shown in column 8. The calculations show that the amount of ^{241}Am caused by ^{241}Pu fission will increase, and it will reach the maximum value of 10 Bq/kg (Lake Žuvintas) after approximately 15 years. Activity concentrations of ^{241}Am measured by α-spectrometry in Lakes Juodis, Žuvintas, Asavėlis vary from 3.0 to 6.0 Bq/kg d.w., 3.0 to 5.0 Bq/kg d.w., 0.7 to 1.0 Bq/kg d.w., respectively, and these values agree with the calculated ones.

The diagram of contribution of the global and Chernobyl NPP accident plutonium to pollution in bottom sediments of the lakes is shown in Fig. 4. The plutonium pollution of Lakes Šventas, Juodis and Asavėlis was mainly caused by the global fallout after the nuclear weapon tests, while Lake Germantas was both in the global and the Chernobyl fallout impact zone. In Lakes Žuvintas and Obelija, in southern Lithuania, plutonium fallout of the Chernobyl accident is distinctly prevailing. These results correlate with the data of air mass transport after the Chernobyl accident

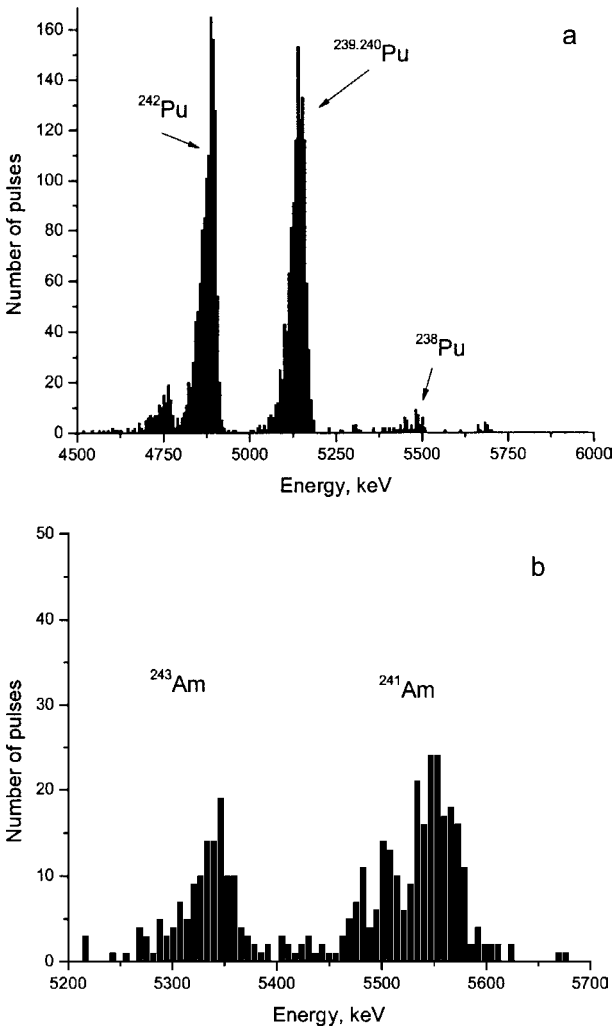

Fig. 3. α Spectrum represents actinides (plutonium (a) and americium (b) isotopes), whose origin is nuclear weapon tests fallout, in the same sample.

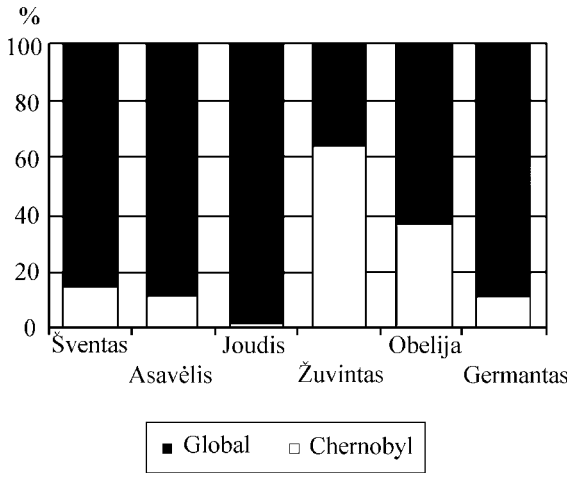

Fig. 4. The diagram of contribution of the global and the Chernobyl plutonium contribution to Lithuanian lakes.

[3] and with the data of the regional pollution with plutonium of the Polish north-eastern region bordering Lithuania [11].

Conclusions

Non-uniform distribution of radionuclides in depositions on the Lithuanian territory after nuclear weapon tests and the Chernobyl NPP accident is experimentally observed by measuring the sediment pollution with plutonium in the lakes. The activity concentration of sediments polluted with plutonium ranges from 2.0 ± 0.5 Bq/kg d.w. in Lake Asavėlis to 14 ± 2 Bq/kg d.w. in Lake Juodis. The ratio of activity concentrations of plutonium isotopes $^{238}Pu/^{239+240}Pu$ measured by α-spectrometry in the 10-cm-thick upper layer of bottom sediment varies from 0.03 in Lake Juodis to 0.30 in Lake Žuvintas. The preliminary data of the ratios of plutonium isotopes show that the contribution of the Chernobyl NPP accident pollution to bottom sediments of the lakes is prevailing in southern and south-western regions of Lithuania. The influence of the global plutonium fallout after the nuclear weapon tests in atmosphere, especially in Novaya Zemlya (North of Russia), is observed on the whole territory of Lithuania, and it is especially distinct in central Lithuania. The americium activity due to ^{241}Pu decay after the Chernobyl NPP accident and global depositions in bottom sediments of the Lithuanian lakes has been evaluated to be from 0.9 to 5.7 Bq/kg. Activity concentrations of ^{241}Am measured by α-spectrometry in Lakes Juodis, Žuvintas and Asavėlis vary from 3.0 to 6.0 Bq/kg d.w., from 3.0 to 5.0 Bq/kg d.w. and from 0.7 to 1.0 Bq/kg d.w., respectively, and these values agree with the calculated ones. The assessment shows that the amount of ^{241}Am caused by ^{241}Pu decay in lakes will increase, and the maximum value will be reached in a few decades.

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