VARIATION OF SPECIFIC ACTIVITY OF ^{137}Cs IN THE BOTTOM GROUND OF WATER RESERVOIRS AND WATERSIDE SOIL IN VILNIUS CITY, LITHUANIA*

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Abstract. *The present work analyses the problem of radiocesium contamination of water bodies. The main object of research is the dynamics of radiocesium in water and bottom sediments of three Lithuanian lakes: Tapeliai, Juodis and Lydekinis. Lake Tapeliai basin is of the glacier origin and has bottom sources. Lake Juodis is a running shallow lake with a thick layer of bottom sediments (over 7 m). Lake Lydekinis is a small humic lake with highly colored water. Sediments and water samples were analyzed for ^{137}Cs using a γ-spectrometric system. Investigation results deepen our knowledge on the processes of lake self-cleaning from anthropogenic pollutants and allow predicting the terms of super warm lake remediation after radioactive impacts. Estimating radioecological consequences of the radioactive impact to the natural water bodies, meromictic lakes are suggested as critical objects.*

Key words: *Radiocesium, specific activity, activity concentration, lake, bottom sediments*

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1. Introduction

The migration of radionuclides in aquatic ecosystem has been raising a huge interest for some years. Radiocesium is an important indicator showing the pollution of the aquatic ecosystem [1]. Radiocesium has been investigated for a quite long time. This artificial radionuclide gained attention of the researches after the accidents of the Chernobyl and Fukushima nuclear power plants. The activity level of the radiocesium is usually indicated in soil and sea water, however, for the past decade more and more attention has been drawn to the investigation of radiocesium in river and lake waters

It is known that close lakes and water objects with sufficiently high water retention perform as accumulators of long-lived radionuclides [2,3]. They can content a high concentration of long-lived radionuclides in the water and bottom sediments [4,5]. The bottom sediments of the lake are the most important element of the water environment acting as a “filter” of radionuclides or as a temporary sink of radionuclides [6]. The vertical profiles of radiocesium bottom sediments are used to restore the chronology of the radioactive pollution, to evaluate the general load of the bottom sediments and sedimentation rate (the deposition of suspended particles in water per year) [7].

It can be stated that the processes of radionuclides’ retention are concerned with their chemical and physical processes and bottom sediments. Having occurred in the water the radiocesium migrates to the bottom sediments by direct absorption or together with suspended particles, which reach the bottom sediments later. Usually the bottom sediments are presented as “filter” for radionuclides, however they can also be the source of radionuclides.

The main object of the study is a radiocesium behavior in a relationship with the seasonal variations of standard water parameters in Lithuanian lakes.

The main object of the study is to estimate the peculiarity of radiocesium dynamics in Vilnius city (Lithuania) lake waters, bottom sediments and near bottom water. The research has shown that the concentration of pollutants is ten times higher in lower stagnant water layer of close meromictic lakes than in the waters of dimictic lakes. Therefore such lakes must be considered as objects of increased ecological risk. The results of the research allow to explain the processes of lake self-cleaning and to predict radiocesium pollution remediation processes in lakes.

2. Materials and methods

Water samples (20 l each) were collected during the period of 2000–2010 from different lake depths using a Molchanov barometer (single water volume is ~4 l). The parameters of these water samples were averaged through the whole interval of 40 cm depth collector. Only the aerobic water samples were filtered in the laboratory through the filters of Filtrak 391 using vacuum filtering system. The hypolimnetic water samples, which had sufficient amount of iron oxide flock, were not filtered. Then aerobic and hypolimnetic water samples were evaporated to extract dry deposits for further analysis of radiocesium.

The bottom sediment samples were collected using an Ekman-Birge type bottom sampler made of metal with lockable bottom. The collector was attached with a rope to the manual or mechanical winch and was lowered to the selected depth with

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the open lid. When the required depth was reached a certain mass solid was released which effected the spring mechanism forcing to close the bottom lid.

Radiocesium specific activity of bottom sediment samples was measured using SILENA γ spectrometric system with highly pure germanium detector (HPGe) (42% relative efficiency, resolution - 1.8 keV / 1.33 MeV) by 661.62 keV ^{137m}Ba (^{137}Cs secondary nuclide) γ line. Measurements were carried out using a standard geometry dishes and based on efficiency estimated for the known density samples. The mixture of different densities radionuclides (1 and 1.45 kg/l) prepared in Russian Research Institute of Technical Physics and Radiometric Measurements (Moscow) was used for calibration. The error of radiocesium activity measurements in the samples were evaluated by GAMMAPLIUS software. They were less than 5 % (standard deviation) for high activity samples and did not exceed 15 % for much less active layers of bottom sediments. The samples activity amendment was not entered as the measurements were carried out immediately after sampling.

The dry deposits of water samples were analyzed using ORTEC scale – spectrometric system with a well HPGe detector (volume 170 cm^3, relative efficiency 38 %, resolution – 2.05 keV/1.33 MeV). The efficiency of this γ spectrometric system of samples measurement is higher than SILENA system.

Vilnius city lakes of different specifics were chosen for the research – Juodis, Lydekinis and Tapeliai (Fig. 1).

Figure 1. Location places of lakes: location of samples shown by points

Lake Juodis (54°46´49″N, 25°26´29″E) is a small (only 0.1 km^2) flowing shallow lake in a particular chain of lakes, which are tied up with each other by small rivers. Lake Juodis is eutrophic, it does not have the near-bottom water sources. This lake is of glacial origin. The southern part of the lake is broad and deep (up to 3.5 m), the northern - shallow bottom terrace (depth 1.0 to 1.7 m). The thickness of the bottom sediments in the northern (shallow) part is up to 7 m.

Lake Lydekinis (54°46´11″N, 25°27´23″E) belongs to the river basin as part of the dammed headwaters. This is an open water to the north, west and east sides, separated by the ramparts from the wetland areas. The average and the maximum depth of the lake are 1.6 and 4.5 m respectively. The water of Lake Lydekinis is coloured. Formation of coloured water influence large amounts of iron oxide in this water. Processes of gravitational mixing of the water column in lake Lydekinis are followed by abundant Fe_2O_3 floc creation and sedimentation. Lake consists of two different parts, which do not intermix: the upper part of the lake of some 300-cm thickness, which becomes completely aerobic due to the gravitational mixing in autumn and constantly anoxic stagnant bottom water layer of some 150-cm thickness. It means that this lake is meromictic.

Lake Tapeliai (54° 46´ 49″N, 25° 26´ 29″E) is of glacial origin and is composed of four parts: 1) the shallow southern terrace (4-5 m depth); 2) central deeper part of the lake (7-9 m depth); 3) the northern terrace with gradual bottom (depth ranges from 1.5 m to 6 m); 4) the bottom small terrace on the west side of the lake (depth of 5-6 m). The lake reaches a maximum depth of 9.5 m with a beaver dam. Lake Tapeliai also has bottom sources. They are located in three different areas: a) near the west coast of the southern terrace of approximately 4-4.5 m; b) in the southwestern area of the central part of the lake; c) at the southern edge of the lake near the old drainage channel.

3. Results

After measuring the specific activities of radiocesium in Lake Juodis, it was found that the highest specific activity was 300 Bq kg^{-1} at a 17 cm depth of bottom sediments (Fig. 2). This enables to estimate the sedimentation rate, which is equal to 0.37 cm year^{-1}. By analogy with the previous measurements of radiocesium vertical distribution in bottom sediments [8], the maximum values of radiocesium specific activity are assigned to the moratorium period of nuclear explosion (1963).

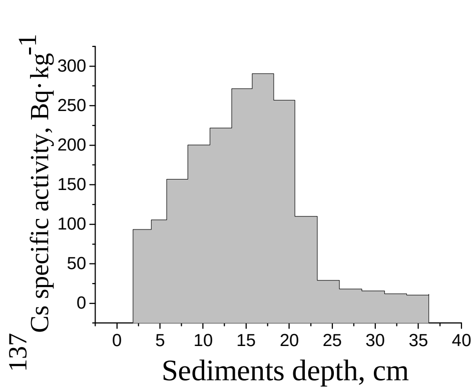

Figure 2. Radiocesium specific activity in Lake Juodis (depth ~3 m). Error of measurements 2 %

Vertical profiles of radiocesium specific activity of the bottom sediments taken from the deepest part of the lake Lydekinis are described by the highest specific activity values of bottom sediments near the surface (Fig. 3) and a marked decrease in activity in the deeper layers.

Sedimentation rate was evaluated according to the fact that the radioactive contamination in deeper areas of the Lake Lydekinis was caused only by global deposition. This rate is approximately 0.06 $cm \cdot y^{-1}$.

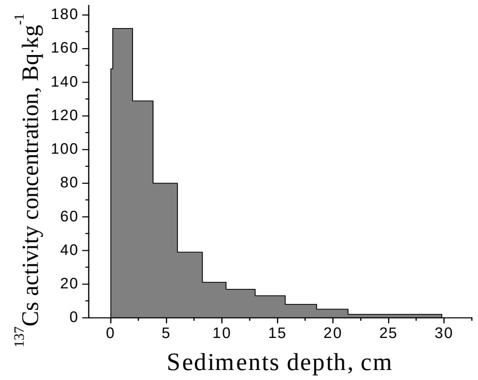

Figure 3. Vertical profiles of radiocesium specific activity in Lake Lydekinis (depth ~4,5 m). Error of measurements 3 %

Radiocesium specific activity of the vertical profiles of the bottom sediment samples taken at a certain distance from the deepest layer of Lake Lydekinis were different. They are described by the increased sedimentation rate (0.2–0.35 $cm \cdot y^{-1}$) and high radiocesium specific activity at 3.5-8 cm depth interval of bottom sediments. One of these vertical profiles shown in Fig. 4 was obtained from the bottom sediment samples collected from a depth of 3.9 m.

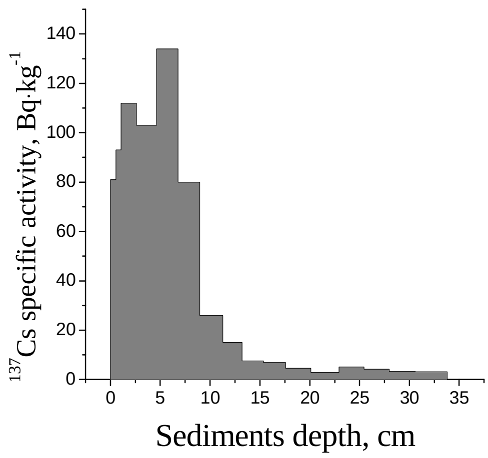

Figure 4. Vertical profiles of the radiocesium specific activity in Lake Lydekinis (depth ~ 3.9 m). Error of measurements 2 %

Using the Gaussian formula for vertical profiles slopes of radiocesium specific activity of the bottom sediments (below the peak of the profile), it was found that characteristic half-widths of these slopes vary in the range 3.4–4.5 cm. Compared with the primary vertical profile, these small shifts in the shape of slopes formed during the fast phase of radiocesium free-ion diffusion [8, 9]. This show limited possibility of radiocesium migration to deeper sediments under these conditions.

Given the relatively wide band of wetland areas at Lake Lydekinis, the bottom sediment samples were collected in the upper basin of the lake at 1.9-3.0 m depth (Fig. 5).

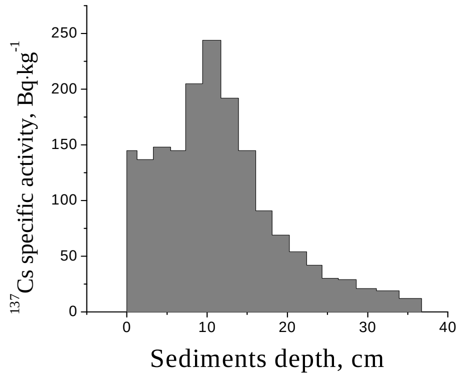

Figure 5. Vertical profiles of the radiocesium specific activity in Lake Lydekinis (depth ~1.9 m). Error of measurements 5 %

The maximum values were measured at the bottom sediments depths from 9.5 to 11.7 cm (Fig. 5), and from 8.5 to 11.1 cm. The characteristics of slopes' half-widths are also relatively high (6.2 and 5.7 cm respectively). The structure of the vertical profiles of radiocesium specific activity in all bottom sediments samples remained the same. However, the maximum density of bottom sediments was identified in the deepest layers: in the 26.4 to 31.1 cm (samples taken from 1.9 m depth of the lake) and in the 25.2 to 29.5 cm (samples taken from 3 m depth of the lake). Sedimentation rate was equal to ~0.5 and ~0.49 $cm \cdot y^{-1}$ respectively.

Having analyzed the gained data of radiocesium activity in Lake Tapeliai, it can be seen that the vertical profile of radiocesium specific activity characterizes its activity peaks on the surface of bottom sediments (Fig. 6).

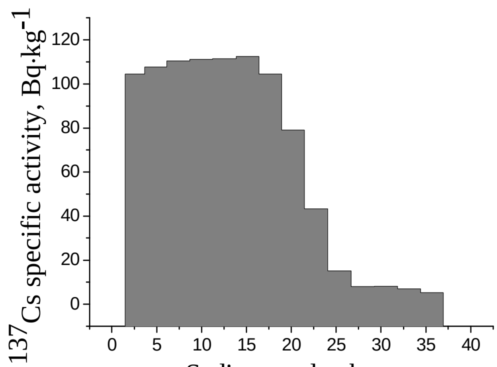

Figure 6. Vertical profiles of the radiocesium specific activity in Lake Tapeliai (~ 3.5 m depth). Error of measurements 2 %

We see that the specific activity of radiocesium at the superficial bottom sediment layer (at the depth of 3.5 m) is slightly higher than 111 $Bq \cdot kg^{-1}$, then, starting from the bottom sediment depth of 15 cm the specific activity decreases. The maximums of vertical profiles of bottom sediment density were at 15-17 cm of bottom sediment depth.

Meanwhile radiocesium specific activity data obtained at a depth of 5.1 m shows a slightly different radiocesium distribution in bottom sediments (Fig. 7).

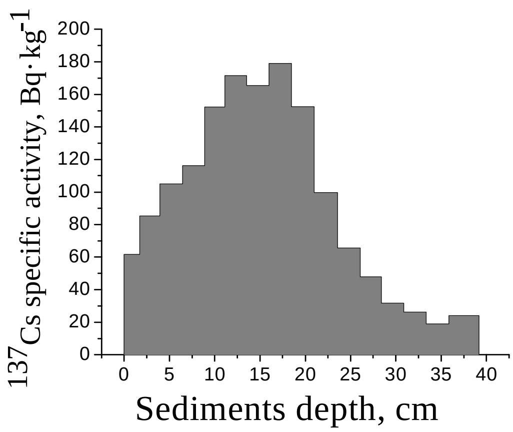

Figure 7. Vertical profiles of the radiocesium specific activity in Lake Tapeliai (depth ~ 5,1 m). Error of measurements 3 %

Given the relatively wide zone of marsh at Lake Lydekinis, the bottom sediment samples were taken in the upper basin of the lake at depth of 1,9-3.0 m (Fig. 5).

In this case, the main part of radiocesium is distributed at the bottom sediment depth of 10–20 cm. We can notice that the radiocesium specific activity was nearly 180 $Bq \cdot kg^{-1}$. The density of sediments solids' vertical profiles show growth starting at 20 cm depth. The distribution of radiocesium at different depths of Lake Tapeliai is irregular: higher values of radiocesium specific activity were indicated at deeper lake layers. Received profiles of radiocesium specific activity show radiocesium's spread deep into the bottom sediments.

The highest values of radiocesium specific activity, measured in the whole lake area, varied at the interval of 110–190 $Bq \cdot kg^{-1}$. Bottom sediment layer, characterized by the highest values of radiocesium was always located at 6 to 19.5 cm deep into the bottom sediments. Radiocesium specific activity of the surface layers of the bottom sediments was 20–120 $Bq \cdot kg^{-1}$. These values were measured in the southern and south-western part of the lake near the bottom sources (Fig. 8).

These data indicate that radiocesium migration in bottom sediments was lowest in the central part of the lake: the slope width ranged from 1.3 to 2.8 cm interval.

The data of bottom sediments load ($Bq \cdot m^{-2}$), considering the collection places of bottom sediments samples are presented in Fig. 9. The data was distributed into four groups according to their collection places: the north terrace, north-west terrace, central part of the lake, and south terrace.

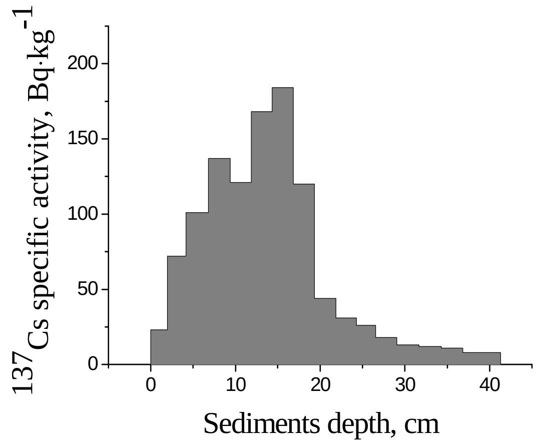

Figure 8. Vertical profiles of the radiocesium specific activity in Lake Tapeliai (depth ~7.7 m). Error of measurements 3 %

The maximum radiocesium inventory was recorded on the north-west terrace, it was around ~1520 $Bq \cdot m^{-2}$. Radiocesium inventory on the southern lake's terrace varied at 770–1470 $Bq \cdot m^{-2}$ interval, while the depth of the lake in that part was 4.2 to 5.4 m. Such radiocesium inventory distribution can be explained by the existence of lake's bottom sources and the influence of the currents of these sources on the spread of the pollutants. The radiocesium inventory on the shallow northern terrace is much smaller and is 850 $Bq \cdot m^{-2}$. The terrace is next to the stream flowing from the lake and open to the wind gusts. The radiocesium inventory in bottom sediments that are below 5.4 m depth of Lake Tapeliai is about 1.5-2 times lower than in the upper water layers. However, sedimentation rate is approximately equal.

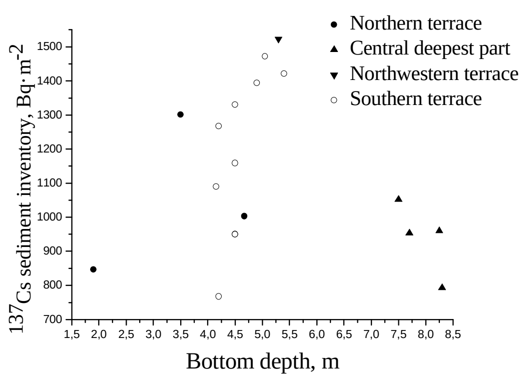

Figure 9. Distribution of radiocesium inventories ($Bq \cdot m^{-2}$) in the lake sediments core with the bottom depth. Error of measurements 5 %

It implies that the main process of sediment activation in the lake is related to the direct sorption of radiocesium onto the sediment surface. It also means that in the lake, which is thermally stratified for about 5 months during the warm period, radioactive pollution were mainly distributed and accumulated in the boundary sediments of the upper mixed water layer. These pollutions are able to reach the bottom sediments of the deepest areas of the lake under conditions of the full lake water mixing in spring or late autumn. Radiocesium inventory in the deepest part of the lake can also increase due to the landslides of the bottom sediments.

Processes of lake self-cleaning are taking place in lake Tapeliai, the same as in other lakes. The studies of Lake Juodis have shown that these processes during winter are thermodynamic and are associated with heat transfer processes during the formation of the ice cover. Depending on the amount of heat generated Lake Tapeliai can be attributed to moderately warm or super warm lakes [10].

During a long exploration period of Lake Juodis (since 2000), only during the winter of the year 2008-2009 the snow cover was low, causing long-term total water layer aerobic conditions for the presence of green algae and phytoplankton photosynthetic activity under the ice. Temperature and radiocesium vertical profiles (Fig. 10) are measured in the aerobic water; data showed that the near-bottom water temperature increase above 4 ° C, which is believed to be associated with an increase in mineralization due to its porous water buoyancy effect, was accompanied by the growth of radiocesium concentration.

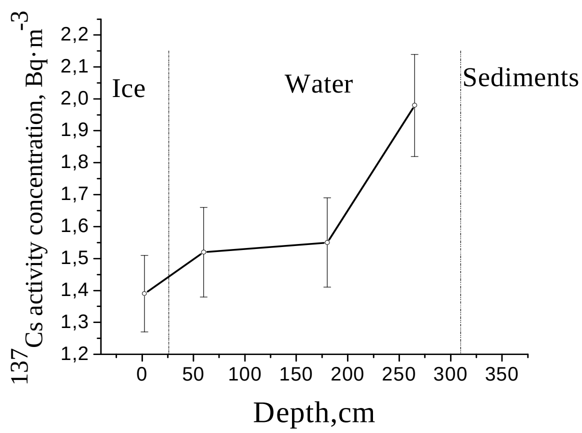

Figure 10. Vertical profiles of ^{137}Cs activity concentrations in Lake Juodis

The measurement data showed that radiocesium activity concentration in the lake water going deep down always increases (Fig. 11), also the tendency of the increase in dry deposits of water samples is observed.

The data shown in Fig. 11 represents approximately the entire range of volumetric activity in coloured water. The broadest range is dependent on surface watercolor: 1.6±0.3 Bq·m^{-3}–7.0±0.4 Bq·m^{-3}.

Radiocesium activity concentration increases with depth, and its range of variation at the depth interval of 3.1–3.6 m decreases almost twice: from 7.3±0.4 Bq·m^{-3} to 9.9±0.8 Bq·m^{-3}. Such variations can be induced by external effect, which is decreasing with depth.

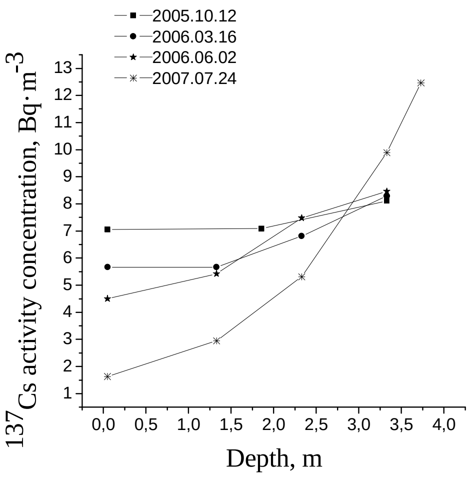

Figure 11. Vertical profiles of radiocesium activity concentrations in Lake Lydekinis water. Error of measurements 5 %

Fig. 12 presents the data of radiocesium activities concentration in surface waters of Lake Tapeliai during the years 2000–2010. The decreasing radiocesium activity concentration tendency in the outflowing river is obvious. Over time, the amount of radiocesium in the central part of the lake started to decrease as well, though the increase of radiocesium activity concentation in spring 2010 is observed. It can be related with the color marshy water access into the lake. Episodic radiocesium activity concentration measurements of inflow into the Lake Tapeliai of colored water stream shows that these concentrations ranging within limits of 4–6.4 Bq·m^{-3} in 2006–2010.

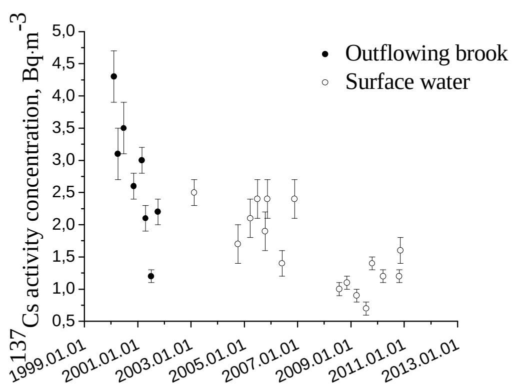

Figure 12. A course of radiocesium activity concentrations in surface water of Lake Tapeliai and in the outflowing brook in 2000–2010

Radiocesium activity concentration data obtained during 2009-2010 is only one-third of the concentration activities measured in the outflowing stream from the lake. However, data show some increase in the radiocesium activity concentration during spring and autumn periods. Colored water in the lake depends on the direction of the wind; under the northern winds, colored water can cover the entire surface of the south terrace.

Having analyzed the obtained data of radiocesium activities, we can see that the vertical profile of radiocesium specific activity describes its activity peaks at the bottom sediment surface.

Fig. 13 shows the radiocesium activity concentration distribution in Lakes Juodis, Tapeliai and Lydekinis.

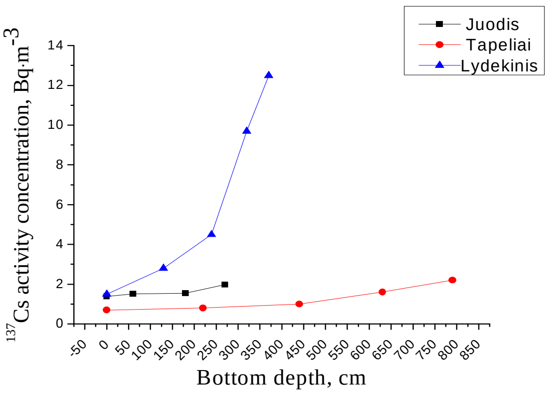

Figure 13. Comparison of the vertical profiles of the water soluble radiocesium activity concentrations in studied lakes in summer. Error of measurements 2 %

The lowest radiocesium activity concentration values are measured in Lake Tapeliai, just slightly higher values are measured in Lake Juodis. However, the radiocesium activity concentration of both lakes does not exceed 2 Bq·m^{-3}. Meanwhile the radiocesium activity concentration in the water of Lake Lydekinis is almost 7 times higher than the values measured in the ecosystems (near-bottom water) of Lakes Juodis and Tapeliai.

From the data, it is evident that radiocesium activity concentration is increasing extremely in near-bottom water. These data show processes of radiocesium release from bottom sediments into the near-bottom water.

Conclusions

1. A thermodynamic mechanism – buoyancy of the sediment interstitial liquids – is responsible for the near-bottom water enrichment in radiocesium in lakes in winter. The process reveals itself also in autumn under aerobic conditions and is a predecessor of the anaerobic zone formation in winter. The latter is proved by the vertical profile of radiocesium activity concentrations in water of shallow Lake Juodis. Under aerobic conditions, these concentrations increase from 1,5 to 2,1 Bq·m^{-3}.
2. The peak values of radiocesium activity concentration measured in the water of Lake Lydekinis is almost 7 times higher than the values recorded in the ecosystems of Lakes Juodis and Tapeliai, and reaches 13 Bq·m^{-3}, which implies that similar meromictic lakes should be treated as increased radioecological risk objects.
3. The temperatures of near-bottom water in lake Tapeliai exceeding 4 ^{o}C in winter and formation of thermally stratified lake water layered structures shows that the bottom sediments' interstitial liquids thermodynamic buoyancy mechanism induces processes of the surface sediment mixing and is involved in processes of lake self-cleaning.
4. The bottom sources of Lake Tapeliai decrease the sedimentation rate in the area of their existence and absorption of pollution through the bottom sediment surface.

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