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^{137}Cs IN LAKE TAPELIAI, LITHUANIA

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The results of an integrated study of the radiocesium behaviour in Lake Tapeliai by using not only conventional data on radiocesium activity concentrations in lake water and sediments but also a complex data set on seasonal variations and vertical profiles of standard water variables are presented. Radiocesium activity concentrations in lake water and a vertical structure of the water column considerably depend on radiocesium-enriched inflows of the coloured waters from the lake's swampy watershed. The global fallouts are mainly responsible for radiocesium inventory in lake sediments, where maximum values of radiocesium are found in the upper part of the water column above the ~5.4 m depth. The maximum values of radiocesium inventories in lake sediments are consistent with the respective densities of its deposits in the nearest forest soils. The main process of sediment activation is a direct sorption of radiocesium onto the sediment surface. Sedimentation rates in the lake mainly vary in the range of 3.5–5 mm yr^{-1}. The lake bottom feeding sources located mainly on the southern terrace as well as their related near-bottom flows reduce respective sedimentation and radiocesium inventories in sediments. The thermal regime of the near-bottom water in Lake Tapeliai in winter strongly depends on the meteorological conditions in autumn and may be classified as varying from super warm to moderately warm. Under conditions of a super warm regime, the elevated radiocesium concentrations in the near-bottom waters may be partially due to the thermodynamic mechanism of its release from the bottom sediments.

Keywords: radiocesium, lake, water, standard variables, sediments

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1. Introduction

The problems of lake contamination by radiocesium are still the focus of many studies [1-9]. It is related to the fact that lakes are a repository of fresh water and its mostly accessible source. Reviewing the results of simulations of the behaviour of radionuclides in fresh water ecosystems, the authors [10] stated an advantage of holistic (lumped) models predicting radionuclide concentrations in water averaged over the entire volume of the lake. These models relate the behaviour of radionuclides in lakes to the relevant environmental characteristics and use generic values for the transfer parameters which are not specific to physical and chemical fundamental processes. Radiocesium concentrations in water averaged over the entire volume of the lake are further used in the modelling of its migration processes in lake sediments [11-13]. However, earlier Santschi et al. [14, 15] evidenced that after the Chernobyl fallouts in deep thermally stratified lakes, the processes of radiocesium direct sorption onto surface sediments in shallow epilimnetic parts of lakes could significantly eliminate its flux to hypolimnion. It implied the necessity of knowledge of the lake thermal structure variations in evaluating the peculiarities of radiocesium loads in bottom sediments. Evidently, the absence of such information assuming a very limited number of sediment cores [1, 2] makes the respective data not completely representative [16]. This comment is very urgent in analysing the state-of-the-art situation in lake radiological studies in Lithuania [17-20], which were not related to the thermal structure of the studied water bodies. As it was stated earlier [21], a simultaneous recording of water standard variables (pH, temperature, oxygen concentrations, and conductivity) could highly improve the significance of radiological data. Thus, a long-term permanent study of seasonal variations of the vertical structure and radiological data (on radiocesium) in Lake Juodis (54°46′49″N, 25°26′29″E) made it possible to determine that carbonate deposits, which were formed in shallow areas of the lake, were acting as a radiocesium barrier [22]. Further, it became possible to evidence that in autumn and winter a thermodynamic mechanism was responsible for the enrichment in radiocesium of the near-bottom water of the lake [23]. A thick layer of organics-rich sediments and their proximity to the water surface made this lake a splendid scientific object allowing fruitful investigations of the processes at the near-bottom water-sediment interface. However, some features of the lake: abundant water plants restricting horizontal mixing [24] and a surrounding pine forest sheltering the lake from wind, may be treated as a somewhat drawback.

The aim of the present work is an integrated study of radiocesium behaviour, including comprehensive information on the variations of standard water parameters in the Lake Tapeliai, which is deeper, larger and not completely wind-sheltered compared with the neighbouring Lake Juodis.

2. Materials and methods

2.1. Object of the study

Lake Tapeliai (54°46′28″N, 25°26′45″E) is located 17 km northeast of Vilnius city, in a wooded region at 136.1 m a. s. l. (Fig. 1).

It is a source water body in the lake chain connected by a brook. The lake is eutrophic. Its banks are rush-grown with a marshy zone formed in a brook outflow area. The outflows are seasonally dependent and vary in the range of 7–80 L s^{-1}. Sometimes, a small beaver dam controls the water level of the lake. The basin of the lake is of the glacier origin (tunnel valley lake, groove type). It consists of four sections: (a) southern shallow terrace (almost flat bottom, ~4–5 m depth), (b) central deepest part of the lake (~7–9 m depth), (c) northern terrace with a gradual bottom deepening from ~1.5 down to ~6 m depths, and (d) small bottom terrace on the western side of the lake (~5–6 m depth). Apart from the watershed, the lake is fed by the bottom feeding sources. They are located in three areas: (a) on the southern terrace at the 4–5 m depth, (b) at the south-western corner of the central part of the lake, and (c) near the southern edge of the lake in the vicinity of the old drainage channel, which earlier connected Lake Tapeliai with Red Lake (54°46′11″N, 25°27′23″E). During flood periods (in spring and after long-term rains) the lake is additionally fed (sometimes for some months) by the temporal discharge with coloured water (shown by a dotted arrow, Fig. 1) from a swamp. The surface area of the lake is equal to ~0.126 km^2, drainage basin ~0.7 km^2 [25], mean depth ~3.3 m, water retention time ~1 year. A typical amount of dry substances in the surface layer of the sapropelic type sediments in the lake mainly varies in the range of 0.020–0.036 kg L^{-1}. The organic content of the sediments determined as a loss on ignition is shown to increase with the sediment depth (down to 30–35 cm) and varies in the range of 50–70% (southern platform). The thickness of the sediment layer measured in the northern terrace increases with the bottom depth. It varies from ~1.5 m near the northern rush-grown bank up to ~4 m at the 6 m depth. The concentrations of total dissolved

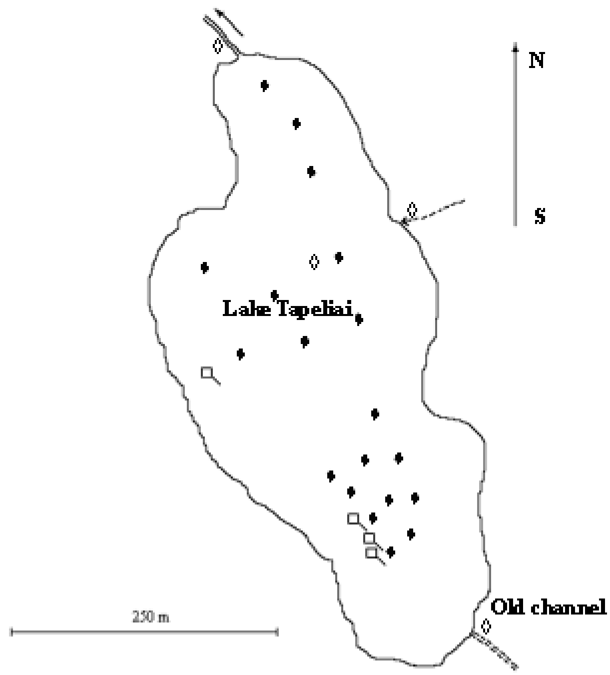

Fig. 1. Scheme of Lake Tapeliai: (♦) sampling sites of sediment cores, (◊) water sampling sites, (⌕) location of bottom feeding sources.

solids (TDS) in lake water determined on evaporation (dry deposits) are mainly in the range of 180–220 mg L^{-1}.

2.2. Parameterisation

The vertical profiles of standard water parameters (pH, temperature, oxygen concentration, and conductivity) were episodically measured in the lake water column from July 2006 to February 2011 with a pause from August 2007 until March 2008. The aim of the study was to estimate lake mixing conditions and seasonal variations of the lake vertical structure. A portable device ProfiLine Multi 197i (*WTW*) with 10 m cables allowed carrying out these measurements down to the lake bottom. During a warm period, measurements were conducted from an inflatable boat stabilised by an anchor. In winter, holes were drilled in ice. As a rule, measurements were carried out at the deepest site of the central part of the lake.

2.3. Water and sediment sampling

Four series of water samples of 20 L volume (20 samples in total) were taken at different depths (0–7.8 m) of the deepest central part of the lake on 25 July 2008, 20 March 2009, 28 July 2009, and 17 March 2010. Surface water samples were taken from the layer of some 5 cm thickness episodically in 2003–2006 and in autumn (on 7 November 2008, 19 October 2009, 25 October 2010, and 11 November 2010) when the water column was partially or completely mixed. The Molchanov type bathometer was used for deeper water samples. In this case, the parameters of water samples were averaged over the 40-cm depth interval of the sampler. The water samples of 15 L volume from the swamp brook (Fig. 1) were taken on 17 and 30 April 2006, 4 and 9 April as well as 23 September 2010. On 18 November 2008, a water sample was taken from a pit dug in the old channel connecting Lake Tapeliai with Red Lake (Fig. 1). After delivery to the laboratory, only the aerobic surface water samples were passed through the Filtrak 391 type filters using a vacuum pump system. Hypolimnetic water samples, where on exposure to air an iron oxide floc was created, were not filtered. Further, surface water aliquots and hypolimnetic water samples were evaporated on a water bath to get dry deposits (further cited as total dissolved solids (TDS)), which were analysed for the radiocesium content. Radiocesium activity concentrations associated with the suspended particles in water samples from the central part of the lake were always below the detection limit (~0.010 Bq) and were not considered. Sediment cores were taken in 2003–2009 using the Ekman–Birge type sampler (19 samples in total). It was a steel tubing with a square cross-section and a manually operated spring bottom shutter. Two versions of this sampler were used: an ordinary one of 20 cm in height (2 sediment cores) and the improved version of ~40 cm in height (17 sediment cores), with cross-sections of 15 × 15 cm and 14 × 14 cm, respectively. The sampling was carried out with the weight compensation, where an additional float controlled the depth the sampler sank into the sediments. Sediment samples without the water layer above the sediment surface were discarded. Sediment cores were sliced into layers of about 2–2.5 cm thickness. Considering that the sampler was not waterproof, the slicing was conducted in shallow waters near the bank using a special spoon to fill the plastic bottles of standard volume and gradually moving the sampler up to the bank. The bottles were held for some time to settle the sediments, and real sediment volumes were determined. Sediment samples were air-dried at room temperature. Their weights and those of dry deposits of water samples were determined using scales VLV-100 (former SU device) where samples were held under thermostatic conditions (in the 40–50 °C temperature interval) up to the constant weight. Measurements showed that dry deposits of water samples were hygroscopic and could change their weight in ambient air in the range of ±5%.

2.4. Radiocesium measurements in sediment and water samples

The sediment samples were analysed for ^{137}Cs using a SILENA γ-spectrometric system with a HPGe detector (42% relative efficiency, resolution 1.8 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 radionuclide mixtures (^{152}Eu + ^{137}Cs) of different densities (1 and 1.45 kg L^{-1}) prepared by the Russian Scientific

Research Institute of Physical-Technical and Radiometric Measurements (Moscow) were used for efficiency calibrations. Measurement errors of radiocesium activity concentrations in the samples were evaluated by the GAMMAPLUS software program. They were less than 5% (standard deviation) for active samples and not larger than 15% for the deepest less active layers of sediment cores. Activity corrections to the sampling date were not made because measurements were carried out shortly after sampling.

The dry deposits of water samples were analysed for ^{137}Cs using an ORTEC γ-spectrometric system with a HPGe well-type detector (sensitive volume 170 cm^3, relative efficiency 38%, resolution 2.05 keV/1.33 MeV). The density corrected calibration was made using four standards prepared in non-liquid matrices in the density range of 0.4–1.7 kg L^{-1} on the basis of the Amersham standard solution [26]. Measurement errors of radiocesium activity concentrations in samples were evaluated by the GAMMAVISION software program and did not exceed 15%.

3. Results

3.1. Parameterisation results

The data of the measurements of vertical profiles of standard water variables showed that Lake Tapeliai was dimictic. The water column of the lake becomes totally oxygenated for a very short period in spring (in April) at water temperatures near 4 °C and for a long-term period in autumn (October or November) due to the cooling processes, inducing intense gravitational mixing. Seasonal courses of oxygen concentrations and conductivity of the surface water, thickness of the oxygenated layer of the water column in 2008–2009 are presented in Fig. 2(a-c). In Fig. 2, measurement points are connected by lines only for visual convenience and do not represent any trends in parameter variations between measurements. However, the spring overturns of the lake when the whole water column is oxygenated are not shown (blank intervals in the respective courses). Those events occurred somewhere before the time of the first measurement point on 27 April 2008 and during the period between 26 March and 20 April 2009, Fig. 2. Before the spring overturn, elevated oxygen concentra-

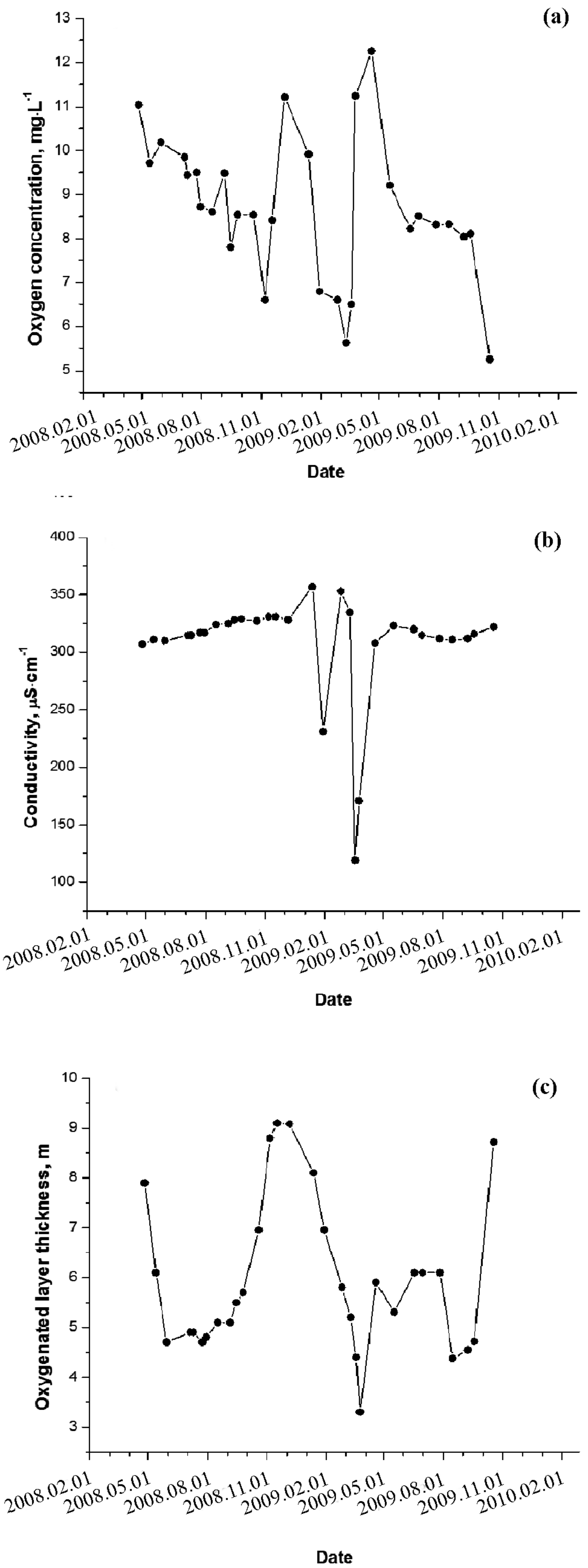

Fig. 2. Seasonal courses of oxygen concentrations in the water column (a), conductivity of surface water (b), and thicknesses of the oxygenated water layer (c) in 2008–2009.

tions in the surface waters are due to an increase in the photosynthetic activity of phytoplankton under the transparent ice cover. After the spring overturn, lake surface waters are enriched in nutrients, which also induce phytoplankton bloom followed by high oxygen concentrations in the surface water exceeding saturation levels, Fig. 2(a) The beginning of the gravitation mixing in autumn incites the dilution of oxygen concentrations due to the involvement of the anaerobic water layers. However, enrichment in nutrients of the surface waters due to gravitation mixing under favourable weather conditions (elevated ambient air temperatures) can also induce elevated photosynthetic activities of phytoplankton.

In the period of 2008–2009, conductivity of the surface waters mainly varied in the range of 310–352 μS cm^{-1}, Fig. 2(b). Two gaps in the conductivity course were due to the appearance of the melting water, while the second gap was related to the beginning of the period of the total lake clearing from the ice cover. However in 2010, due to large inflows of coloured water from the swamp during flood periods, the conductivities of the surface water decreased remarkably and varied mainly in the range of 150–220 μS cm^{-1}. The data of the measurements of conductivities of the swamp brook waters carried out in spring and autumn of 2010 showed them to vary in the range of 63–69 μS cm^{-1}; pH of the coloured water was ~4.9. Thus, during the thaw on 6 February 2011 the surface water under ice in the lake's central part became coloured and its conductivity decreased down to 18–25 μS cm^{-1}; water pH varied in the range of 4.52–4.62.

The data on the seasonal course of the thickness of the oxygenated layer (cut-off value of oxygen concentrations ~0.1 mg L^{-1}) of the lake water column (Fig. 2(c)) show the depth below which the zone of elevated mineralisation begins. In the anaerobic water layer, radiocesium can be released from the sediments due to the decomposition of its potentially mobile physico-chemical forms (carbonate, Fe and Mn oxide, and organic ones). Therefore, the size of the anaerobic water layer implies a potential ability of the lake self-cleaning from radiocesium. The thickness of the oxygenated layer in summer may be significantly larger than that of the completely mixed layer and may be defined in the case of very thick metalimnion as the depth of the maximum gradient of the slope of the vertical profile of oxygen concentrations. Thus, during summer samplings on 25 July 2008 and 28 July 2009, the thicknesses of the oxygenated layers were ~3.7 and ~6.1 m, respectively. At that point, the estimated values of mixed layer thicknesses were ~2.7 and 3.5 m, respectively. However, in the case of radioactive effluence or precipitation over the lake surface, the spread of those contaminants would only be at first possible within that mixed layer [14, 15].

Large inflows of the coloured swamp water in 2010 led to decrease in the thickness of the oxygenated layer in spring down to ~2 m on 17 March and down to ~3.6 m in summer on 27 July 2010. The influences of coloured water inflows on the transparency of the lake surface water to solar radiation and on the thickness of the photic layer had been studied earlier [27].

3.2. Radiocesium activity concentrations in lake water

The course of water-soluble radiocesium activity concentrations in surface waters of Lake Tapeliai in the period of 2000–2010 is presented in Fig. 3. The first eight points represent the data of respective concentration measurements in water samples taken in the outflowing brook [28]. The latter ones show water-soluble radiocesium activity concentrations in surface water samples taken in the centre of the deepest part of the lake. It is easy to see that the mean concentration of the water-soluble

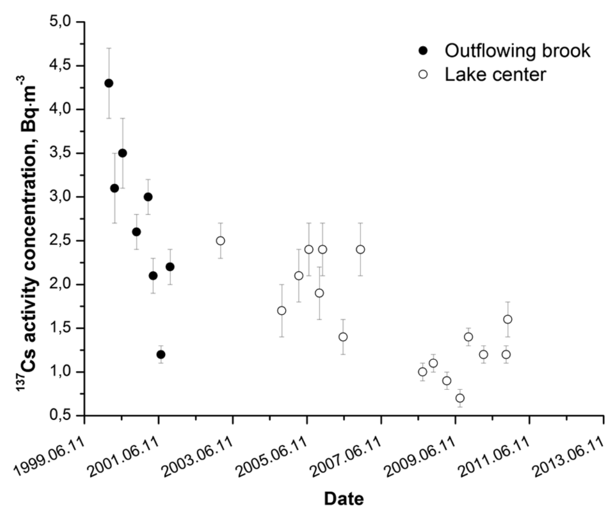

Fig. 3. A course of water-soluble radiocesium activity concentrations in (○) lake and (●) outflowing brook water in 2000–2010.

radiocesium activity in the measured samples in the period of 2009–2010 amounts to only one-third of that in the outflowing brook in 2000. However, the course of the data shows a somewhat irregular rise in those concentrations in spring and autumn, which may be also related to the appearance in surface waters of inflows from the swamp brook. Thus, the elevated water-soluble radiocesium activity concentrations in the range of 4–6.4 Bq m^{-3} were always measured in brook water during the period of 2006–2010.

The spread of the coloured water depends on wind direction, and at northern winds it can cover the total surface of the southern terrace. In the case of southern winds, only the northern part of the lake surface is affected. The mineralisation of the coloured brook water in spring in the period of 2006–2010 varied in the range of 137–185 g · m^{-3}. However, in autumn 2010 it increased up to 240 g · m^{-3}. Due to a relatively small density of the swamp water, water must be located on the surface in a thermally stratified water column. However, in windy weather it is distributed in a completely mixed water layer. Presumably, the influence of old channel water on water-soluble radiocesium activity concentrations in the lake is very small: this concentration in water of the pit is of the ~1 Bq · m^{-3} order.

The vertical profiles of the water-soluble radiocesium activity and TDS concentrations as well as of temperature in the water column in summer on 25 July 2008 and 28 July 2009 are presented in Fig. 4(a-c)). Data show that water-soluble radiocesium activity concentrations vary with depth in summer in a rather narrow range of 0.7 ± 0.1–2.2 ± 0.2 Bq · m^{-3}, Fig. 4(a). These variations were also small in the upper partially mixed layer (down to the ~4.5 m depth): a range of 1 ± 0.1–1.1 ± 0.1 Bq · m^{-3} on 25 July 2008 and a range of 0.7 ± 0.1–1.1 ± 0.10 Bq · m^{-3} on 28 July 2009. The data on TDS concentrations presented in Fig. 4(b) may slightly differ from those of salinity due to the loss on evaporation of volatile components of water admixtures. Variations of TDS concentrations with depth were in the range of 198.4–207.7 g · m^{-3} on 25 July 2008 and in the range of 182.2–226.8 g · m^{-3} on 28 July 2009. The vertical courses of temperature profiles (Fig. 4(c)) show a rather strong diurnal stratification of the mixed layers. It seems that only an uppermost part of mixed layers is affected by wind and, presumably, the mixing processes are mostly effective at night due to decrease in ambient air temperatures.

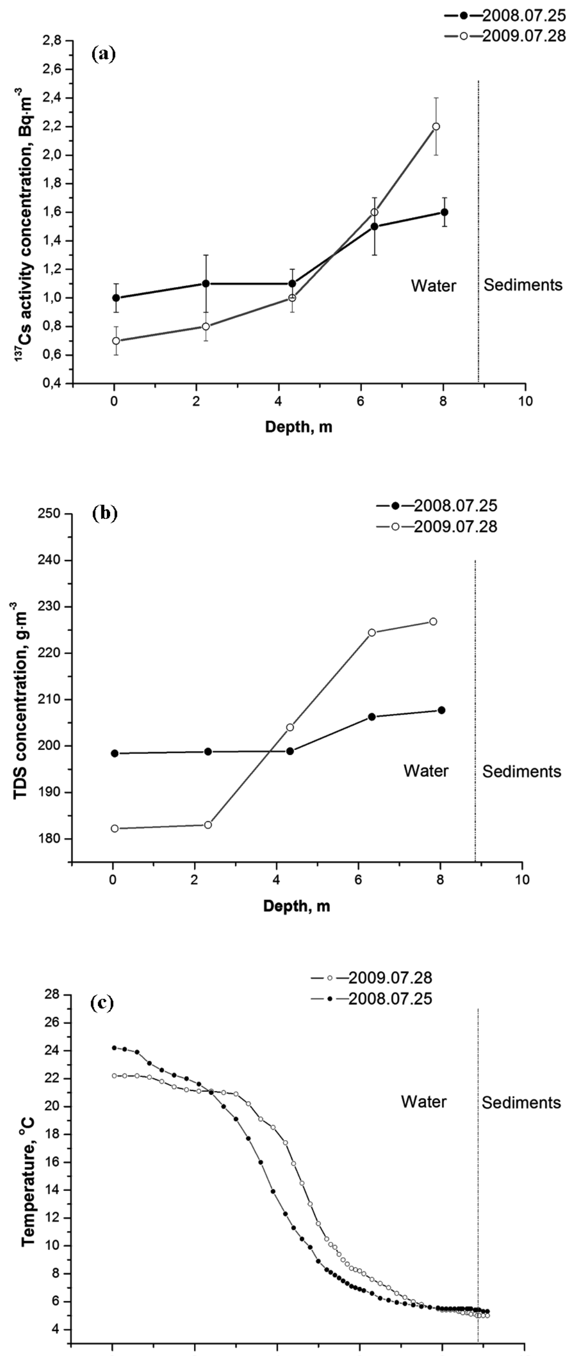

Fig. 4. Vertical profiles of (a) radiocesium activity and (b) TDS concentrations in lake water and temperature in summer 2008 and 2009.

The vertical profiles of water-soluble radiocesium activity and TDS concentrations as well as of temperature in the water column in winter on 20 March 2009 and 17 March 2010 are presented in Fig. 5(a-c). As in the summer case, a range of variations of water-soluble radiocesium activity concentrations with depth in winter was narrow. The concentrations varied in the range of 0.9 ± 0.1–1.7 ± 0.1 Bq · m^{-3} on 20 March 2009 and in the range of 1.1 ± 0.1–1.3 ± 0.1 Bq · m^{-3} on 17 March 2010. The respective variations of TDS concentrations on 20 March 2009 were rather large (127.4–215.3 g · m^{-3}) due to the presence of melting water under ice. It was also the cause of a rather low concentration of the water-soluble radiocesium activity in surface waters (0.9 ± 0.1 Bq · m^{-3}).

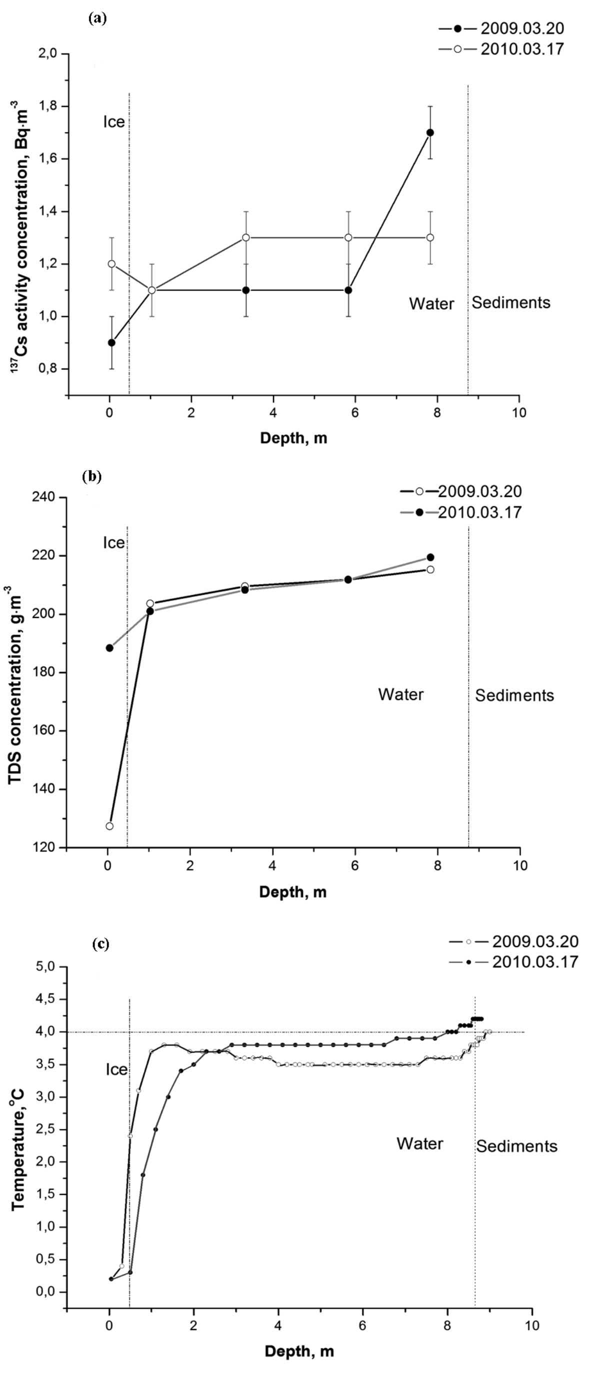

Fig. 5. Vertical profiles of (a) radiocesium activity and (b) TDS concentrations in lake water and temperature in winters 2008/2009 and 2009/2010.

In the interval of depth of 2–7.8 m, variations of TDS concentrations on 20 March 2009 and on 17 March 2010 were about the same. It is the zone of very small temperature gradients (Fig. 5(c)), which can evidence an unstable thermohalinic situation in the water column, especially in the 1.6–4 m depth interval where even small negative temperature gradients (~–0.3 °C · m^{-1}) were measured on 20 March 2009.

3.3. Radiocesium in the lake bottom sediments

The vertical profiles of radiocesium activity concentrations in bottom sediments were distinguished for their elevated deepening, and they did not fit even to the ~40 cm height of our improved sampler. However, estimations of the radiocesium load losses due to the “tails” of its vertical profiles, which did not fit to the sampler height, showed them to be small (≤10%) [27]. Typical forms of the vertical profiles of radiocesium activity concentrations (Bq · kg^{-1}, Bq · L^{-1}) in sediments as well as those of the density of sediment solids in the cores taken on the southern terrace are presented in Fig. 6(a-c). The maximum values of radiocesium activity concentrations (Bq · kg^{-1}, Bq · L^{-1}) in the vertical profiles measured in sediment cores all over the lake varied in the range of 110–190 Bq · kg^{-1} and 4.6–8.7 Bq · L^{-1}, respectively (the data are decay-corrected to the date of the measurements of the sediment samples from the latest core in November 2009). The sediment layers distinguished for their maximum radiocesium activity concentrations were always located beneath the sediment surface in the sediment depth interval of 6–19.5 cm. Measurement data showed that radiocesium activity concentrations (Bq · kg^{-1}, Bq · L^{-1}) in the uppermost surface layer of the sediments varied in the range of 20–120 Bq · kg^{-1} and 1.5–5.4 Bq · L^{-1}, respectively.

The minimum values were measured in sediment cores taken in the southern and south-western parts of the central deepest area of the lake adjacent to the southern terrace with its bottom feeding sources, Fig. 7(a,b). Radiocesium migration abilities in sediments were estimated by fitting the slopes of vertical profiles of radiocesium activity concentrations in sediment cores below its peak activities to the Gauss shape and determining a characteristic width of slopes at their half-heights. Those data showed that migration of radiocesium was minimal in the sediments of the deepest central area of the lake with the widths of the slopes varying in the range of 1.3–2.8 cm. These values were rather elevated in the respective vertical profiles measured in sediment cores from the northern terrace (3.7–4.9 cm) in the bottom depth interval of 1.9–4.7 m. A range of such variations was especially wide (~3.8–7.5 cm) for radiocesium vertical profiles measured in the sediment cores taken on the lake's southern terrace. The elevated migration abilities of radiocesium in the bottom sediments may be explained by bioturbation due to red worms (*Chironomid* larvae), which were found even below the 30 cm sediment depth in some cores taken in autumn 2009.

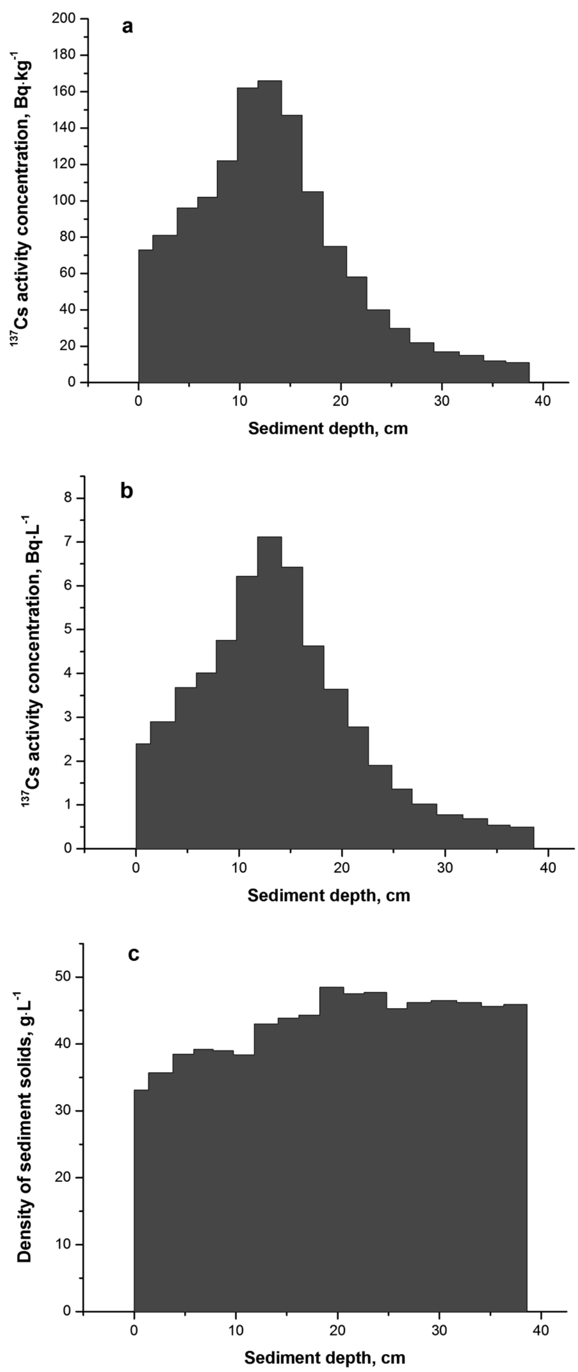

Fig. 6. Vertical profiles of radiocesium activity concentrations measured in (a) Bq · kg^{-1} and (b) Bq · L^{-1} and (c) density of sediment solids in sediment cores taken on the southern terrace on 9 September 2009; 4.5 m depth.

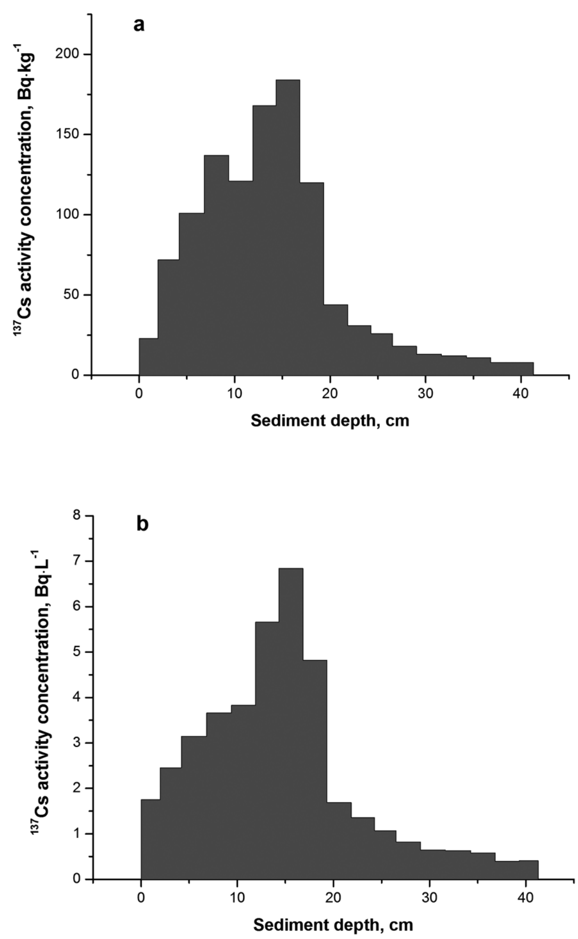

Fig. 7. Vertical profiles of radiocesium activity concentrations measured in (a) Bq · kg^{-1} and (b) Bq · L^{-1} in sediment cores taken in the deepest part of the lake on 15 September 2008; 7.7 m depth.

The data on the total radiocesium inventory (deposition per unit area, Bq · m^{-2}) in all sediment cores decay-corrected to the date of the measurements of the sediment samples from the last core in November 2009 were plotted as a function of the bottom depth in . 8.

The results were subdivided into four groups according to their sampling areas: the northern, north-western, and southern terraces, and the deepest zone of the lake. Radiocesium load was maximum in a core taken on the north-western terrace (~1520 Bq · m^{-2}). Radiocesium inventories in the cores taken on the southern terrace were distributed in a wide range of 770–1470 Bq · m^{-2}, although the bottom depth interval was rather narrow (~4.2–5.4 m). Such distribution may be explained by the influence of bottom feeding sources and of their related near-bottom currents on the southern terrace. A radiocesium inventory in the core taken in the shallow area of the northern terrace (~1.9 m depth) was rather low (~850 Bq · m^{-2}). This area is in the proximity of an outflowing brook and is open to the southern winds. The resuspension of fine sediments due to wind stresses stirring up the shallow water increases suspended matter concentrations in the outflowing brook and reduces the respective radiocesium inventory in sediments. Radiocesium inventories of the cores from the deepest central zone of the lake were also comparatively low (below ~1050 Bq · m^{-2}) and did not show a "focusing effect" [2, 29] with depth.

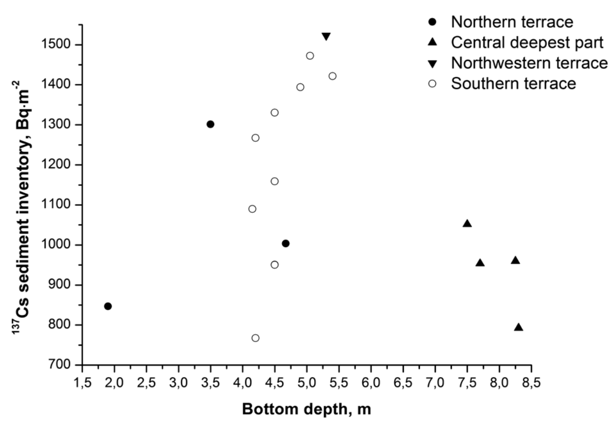

Fig. 8. Distribution of radiocesium inventories (Bq · m^{-2}) in sediment cores with bottom depth.

We suggested that peak activity concentrations of radiocesium in their vertical profiles in sediment cores were attributed to the period of notification of the nuclear weapon test moratorium (1963). The main reasons for this conclusion were our earlier considerations [22] and the fact that an impact of contaminated air masses from Chernobyl on the area of Vilnius city was too short-term and not followed by precipitation contrary to global fallouts. The effects of Chernobyl deposits can be seen in the vertical profiles of radiocesium activity concentrations in sediment cores as small elevations or a somewhat plateau at sediment depths above the main peak, Figs. 7(a,b), 9.

Such sediment dating was further used in the assessments of sedimentation rates in the lake. In this case, the sedimentation rate was calculated as

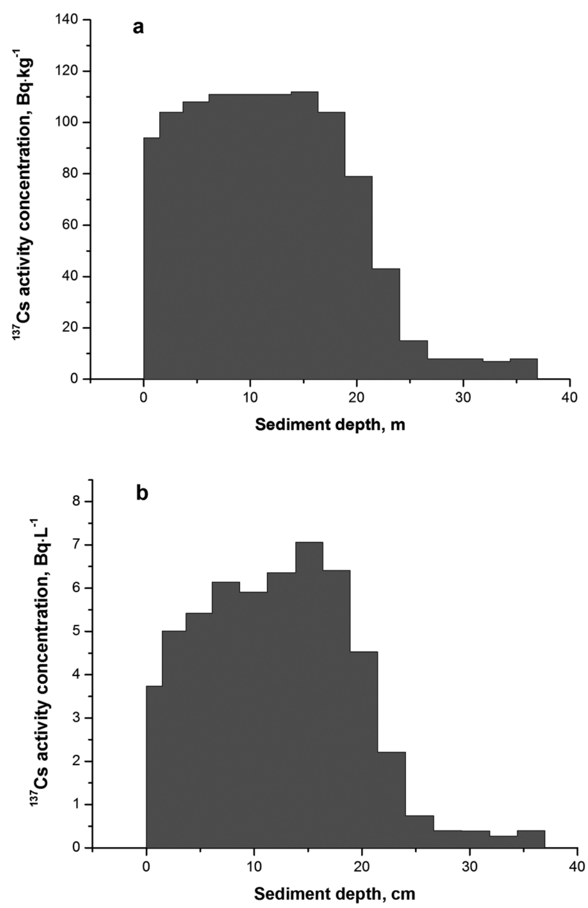

Fig. 9. Vertical profiles of radiocesium activity concentrations measured in (a) Bq · kg^{-1} and (b) Bq · L^{-1} and (c) density of sediment solids in sediment cores taken on the northern terrace on 11 July 2008; 3.5 m depth.

a ratio of the dry mass of the sediment layer in the core above the peak depth to the density of sediment solids of the surface layer (Fig. 10) and to the time interval since the event.

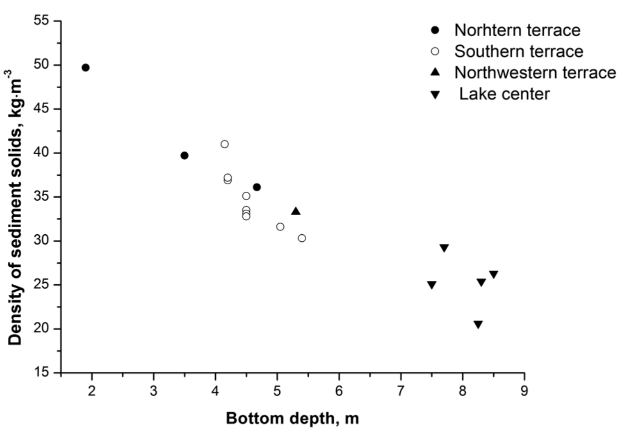

Fig. 10. Distribution of density of sediment solids in their surface layer with the bottom depth.

The density of sediment solids in the surface layer of sediment cores taken in the central deepest area of the lake shows rather large variations (~21–29 kg · m^{-3}). Presumably, it is related to the processes of sediment sloughing in some areas of the central deepest part of the lake due to rather steep slopes of the bottom relief [6, 30]. As it was mentioned above, due to the processes of fine sediment resuspension by southern winds in the shallow zone of the northern terrace and further withdrawal of the resuspended substances with flushing waters, the density of surface sediments in that zone was elevated, Fig. 10. The estimations of sedimentation rates in the lake are shown in Fig. 11. It is easy to see (Fig. 11) that according to the main part of sediment cores, the sedimentation rate in the lake varies in the range of ~3.5–5 mm · yr^{-1}. Some parts of the bottom areas on the southern terrace are affected by the bottom feeding sources and by their related near-bottom flows, which reduce the respective sedimentation rates. Sedimentation rates in the proximity of the outflowing brook may be not only seemingly low due to losses of fine sediments in the course of resuspension. A rather thin water layer (~1.9 m) in the area, which is less than

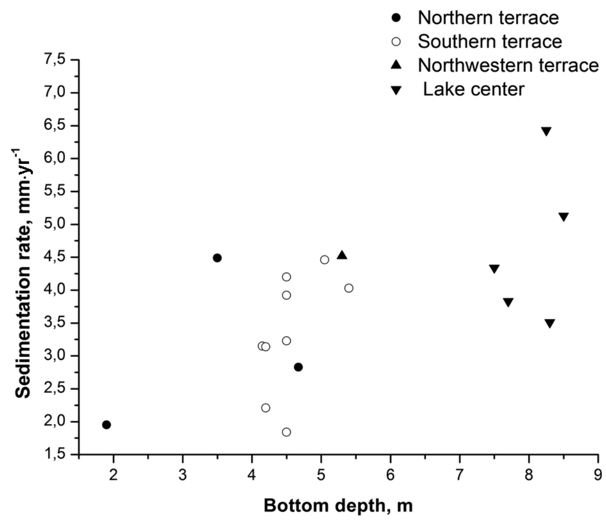

Fig. 11. Distribution of the estimated values of sedimentation rates with bottom depth.

that of a photic zone, implies a respective decrease in sedimentation rates due to dead phytoplankton.

However on the other hand, additional sedimentation in eutrophic shallow waters is possible due to the photosynthetic activity of green algae covering the bottom surfaces [22]. Sedimentation rates calculated using reference peaks of radiocesium activity concentrations in its vertical profiles due to Chernobyl deposits were in good agreement with other assessments consistent with the used sediment dating.

4. Discussion

It seems that the data on the course of the radiocesium activity concentrations in surface waters presented in Fig. 3 do not show any further evident decrease in their values with time. In turn, the data on vertical profiles of radiocesium activity concentrations in Figs. 4, 5 imply somewhat oscillations with depth: small values in the surface waters are always related to elevated radiocesium activity concentrations in the near-bottom water and vice versa. It seems that the total radiocesium inventory of the water column does not change. In this case, information on radiocesium activity concentrations in surface waters in autumn when the water column is completely mixed is very useful. Thus, in November 2005 and 2006, radiocesium activity concentrations in surface waters were the same

(~2.4 ± 0.3 Bq · m^{-3}). In November 2008, they decreased down to ~1.1 ± 0.1 Bq · m^{-3}. In a completely mixed water column on 19 October 2009, the concentrations amounted to ~1.4 ± 0.1 Bq · m^{-3}. Two samples of the surface water were taken in autumn 2010, on 25 October and 11 November. On 25 October 2010, when the water column was mixed only down to the ~8.5 m depth, the radiocesium activity concentration in the surface water amounted to ~1.2 ± 0.1 Bq · m^{-3}. In a completely mixed water column on 11 November 2010, this concentration increased to ~1.6 ± 0.2 Bq · m^{-3}. Evidently, the latter value was due to the inflow of coloured water from the swamp brook with its elevated radiocesium activity concentrations. Obviously, Lake Tapeliai is recurrently supplied with radiocesium from its boggy watershed during flood periods. In autumn, under conditions of gravitational and wind stress induced mixing, this additional amount of radiocesium is equally distributed over the whole water column increasing its total inventory in the lake. This phenomenon is well known in the literature [31, 32].

A comparison of radiocesium inventories in lake sediments with its load in the watershed soils shows that down to some 5.4 m depth these inventories are about of the same order. Thus, the radiocesium load in watershed soils according to its inventory in a soil core, which was sampled on 6 November 2000 near Lake Tapeliai [33], amounted to ~2130 Bq · m^{-2} (~1730 Bq · m^{-2} in November 2009). It is consistent with the average value of radiocesium density of deposits after the Chernobyl accident in undisturbed meadow soils in the middle and eastern regions of Lithuania in 1992–1993 (~1740 Bq · m^{-2} in November 2009) [34, 35]. This value in forest soils with litter amounted to ~3.3 ± 1.3 kBq · m^{-2} (~2.3 kBq · m^{-2} in November 2009). However, we think that the estimation of density of radiocesium deposits on the surface of the lake using the inventory measured in the soil core taken near Lake Tapeliai on 6 November 2000 is rather overestimated. The main reason for this conclusion is related to the fact that the sampling site was surrounded by pine forest and the soil surface was covered with forest litter. A distinct peak of the radiocesium activity concentration of Chernobyl origin in the respective vertical profile in the soil core, Fig. 12 [33], belongs to the litter layer (densities ~200–300 kg · m^{-3}). Apparently, not only the direct radiocesium deposits after Chernobyl fallouts, but also further washout processes in pine forest were responsible for activities of this peak in the soil core. As it was mentioned above, radiocesium inventories in lake sediment cores taken below some 5.4 m depth were about 1.5–2 times as small as those of the overlying water layer, Fig. 8. However, sedimentation rates in both water layers were of the same order. 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 fallouts of global as well as of Chernobyl origin were mainly distributed and accumulated in the boundary sediments of the upper mixed water layer. These products were able to reach the sediments of the deepest bottom areas of the lake under conditions of the completely mixed water column in late autumn and spring.

In Lake Tapeliai, as in other lakes which after global fallouts and the Chernobyl impact turned into the repository of artificial radionuclides, the processes of sediment self-cleaning take place. Earlier studies carried out in Lake Juodis showed that in winter these processes are of thermodynamic origin [23] and are related to the amount of heat accumulated by sediments until the time of the formation of the ice cover. Depending on the amount, Lake Tapeliai may be treated as a super warm or

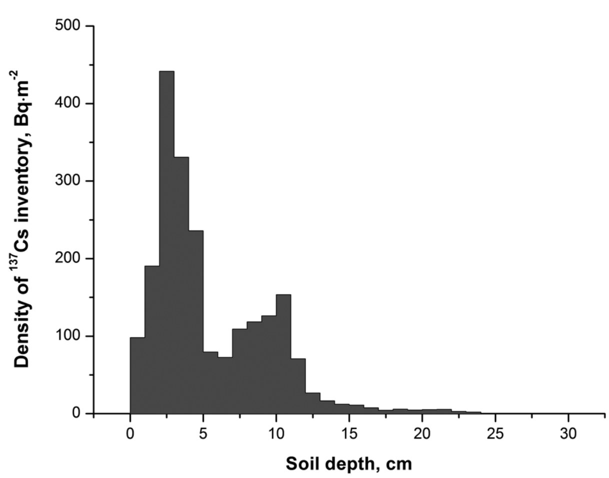

Fig. 12. Vertical profile of radiocesium inventory (Bq · m^{-2}) in 1 cm thickness layers in soil cores taken near Lake Tapeliai on 6 November 2000.

moderately warm lake in some years [36, 37]. Investigations showed that the temperature of the surface sediments in the deepest areas of the lake is maximal in autumn. Obviously, a mechanism of such additional heating in autumn is related to the complete stirring of the water column induced by wind stresses imposed onto the lake surface during the processes of gravitational mixing at water temperatures exceeding those of the sediments [38, 39]. Thus, it is known [40] that due to strong winds and elevated temperatures of ambient air, the temperature of the sediment surface in deep bottom areas in Lake Landvettersjön (Sweden, maximum depth ~21 m) during a short convection period in autumn increased up to about 10 °C. Evidently, the amount of heat accumulated in sediments of deep bottom areas of lakes depends not only on the duration of this short-term heating period but also on further weather conditions. Considering that a completely mixed water column is isothermal and unstable [39], cold weather and further winds may lead to a significant overcooling of the water column below the temperature of water maximum density (~4 °C). Thus, in Nordic lakes, average temperatures of the water column before the formation of the ice cover drop down to ~0.5 °C [41, 42]. In this case, a 4 °C isotherm must be deepened below the sediment surface due to elevated thermal diffusivity of the sediment surface layer, which is affected by the buoyancy of interstitial liquids [23, 43]. Therefore, the significant overcooling of the water column induces respective heat losses in the sediments. In turn, the buoyancy of interstitial liquids enriched in radiocesium is followed by its release into the completely mixed water column. Therefore, it implies that radiocesium will be partially removed with the flushing waters. However, the processes of sediment self-cleaning are not finished at this stage. The thermal diffusivity of the water column, which becomes temperature stratified before ice formation, decreases significantly due to the absence of convective motions. In turn, it leads to the decrease in the earlier elevated thermal diffusivity of the sediment surface layer, which was induced by buoyancy forces. The process is followed by the strengthening of the temperature gradient below the sediment surface due to the appearance of an obstacle (decreased thermal diffusivity of the sediment surface layer) to the permanent heat flux from the sediment proper directed upwards. In the case of large amounts of heat accumulated in sediments at critical heat and salinity Rayleigh numbers, the secondary buoyancy events are possible in surface sediments [23]. They are followed by the additional intrusions of highly mineralised and warm interstitial liquids to the near-bottom water and by the increase in the near-bottom water temperatures above 4 °C. Under these conditions, a specific layered structure of the water column is formed. Possibilities of such events in Lake Tapeliai were confirmed in the winter of 2009/2010 [36]. The intrusions of sediment interstitial liquids enriched in reduced ions together with the processes of organics decomposition in the surface sediments induce the formation of an anaerobic zone in the near-bottom waters. Evidently, both processes are related to the radiocesium enrichment in the near-bottom waters. Nevertheless, investigations carried out in Lake Juodis [23] allowed showing that in super warm lakes the radiocesium enrichment due to the interstitial liquid intrusions might be possible under aerobic conditions and it was a precursor of the anaerobic zone formation. Presumably, under conditions of a moderately warm regime in Lake Tapeliai in winter, elevated radiocesium activity concentrations in the anaerobic near-bottom waters may be mainly due to processes of sediment organics decomposition. In any case, this statement may be checked studying the seasonal course of radiocesium physico-chemical forms in surface sediments of the deepest bottom area of the lake [24].

5. Conclusions

The present investigations of the radiocesium behaviour in Lake Tapeliai showed an advantage of an integrated approach using not only conventional data on radiocesium activity concentrations in lake water and sediments but also a complex data set on seasonal variations and vertical profiles of standard water variables. These complex data show that radiocesium activity concentrations in lake water as well as a vertical structure of the water column considerably depend on the inflows of the coloured water enriched in radiocesium from the lake swampy watershed. Radiocesium activity concentrations in lake sediments are mainly due to the global fallouts and their radiocesium inventory is maximum in the upper part of the water column above the ~5.4 m depth. The maximum values of radiocesium inventories in lake sediments are consistent with the respective densities of its deposits in the nearest forest soils. The main process of sediment activation is a direct sorption of radiocesium onto the sediment surface. Sedimentation rates in the lake mainly vary in the range of 3.5–5 mm · yr^{-1}. The lake bottom feeding sources located mainly on the southern terrace as well as their related near-bottom flows reduce sedimentation and radiocesium inventories in the sediments. A thermal regime of the near-bottom water in Lake Tapeliai in winter strongly depends on the meteorological conditions in autumn and may be classified as varying from super warm to moderately warm. Under conditions of a super warm regime, radiocesium elevated concentrations in the near-bottom waters may be partially due to the thermodynamic mechanism of its release from bottom sediments.

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RADIOCEZIS TAPELIŲ (LIETUVA) EŽERE

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Santrauka

Darbe yra tiriama radiocezio elgsena Tapelių ežere panaudojant ne tik įprastinius duomenis apie radiocezio aktyvumo koncentracijas ežero vandenyje ir dugno nuosėdose, bet ir vandens standartinių parametrų vertikalių profilių ir jų sezoninių variacijų matavimo rezultatus. Parodyta, kad pelkės spalvoto vandens, praturtinto radioceziu, pritekėjimas yra labai reikšmingas ežero vertikaliai struktūrai ir vandens radioaktyviajai taršai. Tyrimai rodo, kad radiocezio atsargos ežero dugno nuosėdose susiformavo dėl jo globalinių iškritų ir šių atsargų didžiausias kiekis sukauptas seklesnėje ežero dalyje (iki ~5,4 m gylio). Maksimalūs radiocezio kiekiai dugno nuosėdose atitinka jo iškritų tankį šalia esančiame miško dirvožemyje. Darbe parodyta, kad pagrindinis dugno nuosėdų aktyvacijos radioceziu mechanizmas yra jo tiesioginė sorbcija. Sedimentacijos greitis ežere buvo įvertintas 3,5–5 mm/metus. Ežero dugniniai šaltiniai pietinėje ežero dalyje ir jų veikiamos priedugninio vandens srovės silpnina sedimentaciją ir mažina šių dugno nuosėdų užterštumą radioceziu. Priedugninio vandens terminis režimas žiemą ypač priklauso nuo meteorologinių sąlygų rudenį ir gali svyruoti nuo *itin šiltojo* (temperatūra per 4 °C) iki *vidutiniškai šiltojo* (temperatūra žemiau 4 °C) tipo. Tikėtina, kad priedugninio vandens padidintas radiocezio koncentracijas *itin šiltojo* režimo sąlygomis žiemą iš dalies lemia šio nuklido išskyrimas termodinaminiu mechanizmu iš dugno nuosėdų.