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ORIGINAL PAPER

On the radiocesium behavior in a small humic lake (Lithuania)

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Abstract. Peculiarities of radiocesium contamination of a small humic lake, which became meromictic some thirty-five years ago due to the inflow of a large amount of humic water, are presented. The lake consists of two separate water layers, which do not intermix. A lower water layer of the lake below some 3-m depth is stagnant and anaerobic, and radiocesium load of the sediments is mainly caused by nuclear weapons fallout. The radiocesium load of the sediments of the upper monomictic water layer is significantly larger due to additional contamination after the Chernobyl accident. Radiocesium activity concentrations in lake water increase with depth, and even in the surface layer, they are commonly the largest among the neighboring lakes with transparent water. It is shown that bottom areas of the monomictic part of the lake with the elevated radiocesium deepening into sediments are related to the favorite sites of the tench (*Tinca tinca*) winter torpor. Sediment bioturbation and redistribution due to tench activities distort naturally formed radiocesium vertical profiles and they cannot be used for estimations of sedimentation rates and sediment chronology. The studied lake can be useful as an analogous model in analyzing structural and radiological consequences of humic water inflows to closed lakes. Concerning extreme radiological situations in closed humic lakes related to their specific vertical structure, they may be treated as critical objects in assessing the risk to humans after radionuclide deposition events.

Key words: radiocesium • lake • closed • humic • sediment • tench

Introduction

It is known that closed lakes and water bodies with the comparatively long water retention times act as accumulators of long-lived radionuclides [2, 13, 19, 23]. They are distinguished for the highest radionuclide activity concentrations in water and some of them are still of serious radiological concern [9, 23]. In modeling of the long-term behavior of radiocesium in closed lakes it has been commonly assumed that runoff from the lake catchments is negligible. The other impact of catchments on lakes is related to sedimentation due to possible washout of soil particles by surface runoff. The main result of these simulations is an exponential decrease in radiocesium activity concentrations with time in lake water due to its decay [13].

In fact, the catchment's influence on the radionuclide behavior in closed lakes is not limited by the above-mentioned model assumptions. Closed lakes are especially sensitive to high inputs of the allochthonous organic matter from their catchments [18]. The allochthonous organic matter from the terrestrial surroundings may be in the form of dissolved organic carbon, (DOC) produced from decaying plant tissue in terrestrial soils [1, 11]. Humic substances in DOC provide a specific color to lake water and reduce its transparency for ultraviolet (UV – wavelengths of 320–400 nm) and

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photosynthetic available radiation (PAR – wavelengths of 400–800 nm) [3, 14, 25]. Absorption of UV and PAR in lake surface waters leads to their overheating and to changes in the temperature stratification of the water column. Simultaneously, it reduces annual integral primary and phytoplankton productivity expressed on an areal basis due to a decrease in the thickness of the euphotic zone with color [10, 15]. All these self-sustaining processes turn humic lakes into dystrophic water bodies with a growing volume of anaerobic bottom zone, large temperature gradients in the metalimnion blocking upward nutrient fluxes and, as a result, with the thin layer of oxygenated surface waters.

It is known [16, 17] that information on the vertical structure of lakes during radionuclide fallout after the Chernobyl accident was very useful. It allowed disclosing that epilimnetic boundaries of temperature stratified lakes acted first as sinks of Chernobyl radiocesium from the upper water column and later they served as its source for deeper lake zones [16, 17].

The aim of the present work is to describe the peculiarities of the radiocesium behavior in a small lake, which due to imprudence of people turned into a humic one some thirty-five years ago, before the Chernobyl accident. This study also aims at showing those peculiarities to be related to the specific vertical structure of humic lakes and their inhabitants. We intend to show that the lake may be treated as an analogous model of closed humic lakes where an additional emergency mechanism of self-cleaning from radiocesium is possible.

Object of study

A small (2.3 ha) humic lake (54°46'11" N, 25°27'23" E) (Fig. 1a) belongs to the catchment area of the Varžuva River (41.2 km^2) [5] as part of a source impoundment (6.9 km^2) (Fig. 1b). Water of the lake is highly colored. We denominated it further as Red Lake due to the distinct reddish color of its sand bottom in the shallow area. It is located 19 km to the northeast of Vilnius in a small depression (~137.4 m above sea level) surrounded by a pine forest. From the north, west and south its open water basin (~1 ha) is separated from the banks by marshy zones. The lake mean and maximum depth amounts to ~1.6 and 4.5 m, respectively. Aforetime, it was an ordinary eutrophic lake with transparent water and bottom feeding sources. It was connected by outflowing ditches with Lake Tapeliai (136.1 m a.s.l.) and the catchment of the Varžuva River (Fig. 1b). The main sources of the Varžuva River impoundment are a small brook from the swamp (*Ch1*) and the outflow of the drainage ditch (*Ch2*) from a large meadow. Water of both sources is also highly colored. Some thirty-five years ago, a source zone of the Varžuva River was dammed and an outflowing ditch connecting Red Lake and Lake Tapeliai was blocked by a sand buffer. During the filling of the depression, a thick layer of some 70 cm of humic water covered the surface of Red Lake. Consequences of this event were dramatic. Apparently, bottom-feeding sources of the lake were blocked and the regime of vertical mixing changed. Finally, Red Lake turned into the repository of humic water.

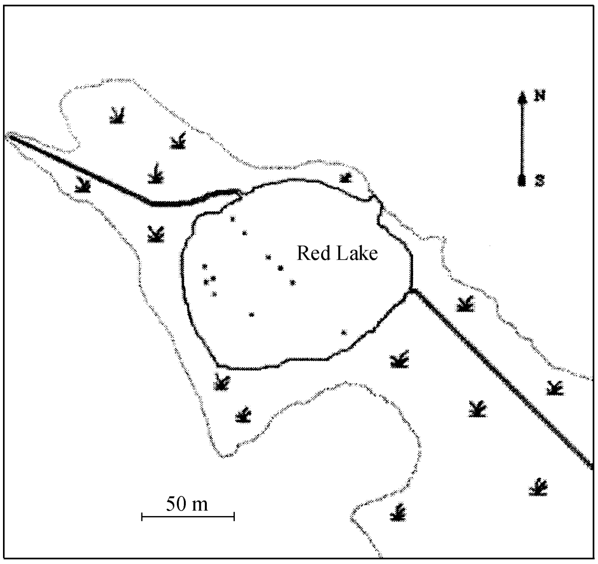

Fig. 1a. Scheme of Red Lake; ▪ – a site of water sampling; × – sites of sediment sampling.

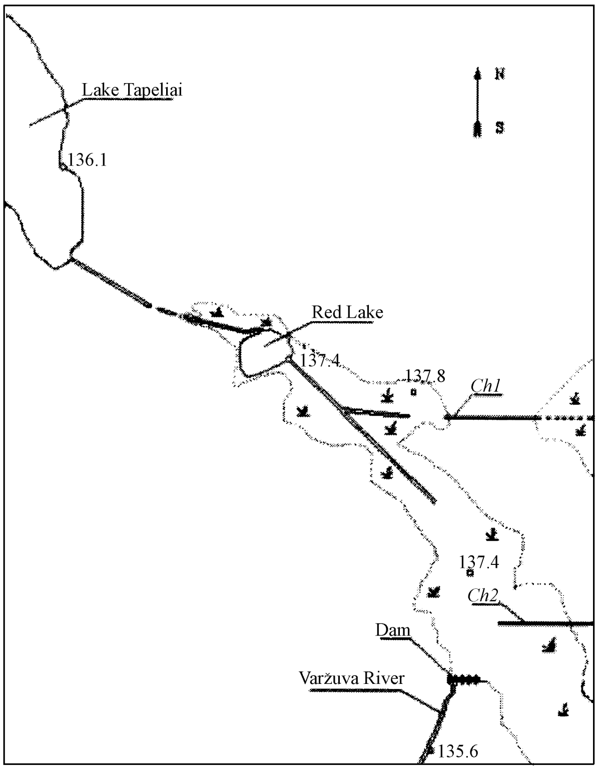

Fig. 1b. Scheme of the source impoundment of the Varžuva River; 137.4 – elevation in meters above the sea level; *Ch1* – an outlet of the swamp brook; *Ch2* – an outlet of the drainage ditch.

Experimental methods

Parameterization

Vertical profiles of standard water parameters (pH, redox potential, temperature, oxygen concentration, and conductivity) were periodically measured in the lake water column from January 2004 to July 2007. The aim of the study was to estimate lake mixing conditions and seasonal variations of the lake vertical structure.

A portable device HAND MULTILAB 12 (SCHOTT) with 3-m cables was used from the very beginning, and later (from November 2006) the 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, stabilized by mooring to a 120-m line extended across the open water basin. In winter, holes were drilled in ice. As a rule, measurements were carried out in the center of the open water basin, but sometimes, for comparison, sites at a 5–10 m distance from the marshy zone were chosen.

Water and sediment sampling

Water samples (10-L volume) (28 samples in all) were taken episodically at different depths (0–3.7 m) in 2004–2007. Surface water samples were taken from the layer of some 5-cm thickness. The Molchanov type bathometer was used for deeper water samples. In this case, parameters of water samples were averaged over the 40-cm depth interval of the sampler. In April 2007, after the end of the flood period, water samples (10-L volume) were taken from the swamp brook (*Ch1*) and from the drainage ditch (*Ch2*) (Fig. 1b). At 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 a dissolved solid concentration) that were analyzed for radiocesium content. Radiocesium activity concentrations associated with the suspended particles were always below the detection limit (~0.010 Bq) and were not considered.

Sediment cores were taken in 1998–2007 using the Ekman–Birge type sampler (13 samples in all). It was a steel tubing with a square cross-section, and had a manually operated spring bottom shutter. Two versions of this sampler were used: an ordinary one of the 20-cm height (3 sediment cores) and the improved version of the 40-cm height (11 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. 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 as well as weights 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 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%.

Radiocesium measurements in sediment and water samples

Sediment samples were analyzed for ^{137}Cs using the SILENA γ-spectrometric system with the 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 known efficiencies according to 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 the radiocesium concentrations in samples were evaluated by the GAMMAPLUS software program. They were less than 5% (standard deviation) for active samples and were 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.

Dry deposits of water samples were analysed for ^{137}Cs using an ORTEC γ-spectrometric system with the HPGe well-type detector (a sensitive volume of 170 cm^3, a relative efficiency of 38%, resolution – 2.05 keV/1.33 MeV). 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 Amersham standard solution [6]. Measurement errors of radiocesium concentrations in samples were evaluated by the GAMMAVISION software program and they did not exceed 10% for active and ~30% for samples of low activity.

Experimental results

Parameterization results

A time-course of maximum oxygen concentrations in Red Lake water

Data on the time-course of maximum oxygen concentrations present in its vertical profiles in the water column measured from 22 October 2004 are shown in Fig. 2. It means that the depths of these maximum concentrations in different profiles may not coincide. Measurement points are connected by lines only for visual convenience and do not represent any trends in oxygen concentration variations between measurements (Fig. 2). During warm seasons, these concentrations are typical of the thin surface layer of 50–60-cm thickness. At the beginning of winter, vertical profiles of oxygen concentrations peak often at the 1–2-m depth and indicate their minimum values in the surface water layer in the immediate vicinity to ice. Data in Fig. 2 show maximum oxygen concentrations gradually decreasing with time in winter. At its end, in March 2005 and 2006, oxygen was completely depleted in the water column. A distinct smell of H_2S from the holes in ice indicated that measured minimum oxygen concentrations in under-ice water were mainly induced by atmospheric air during the hole drilling. Winter of 2006/2007 was very mild and the ice cover in

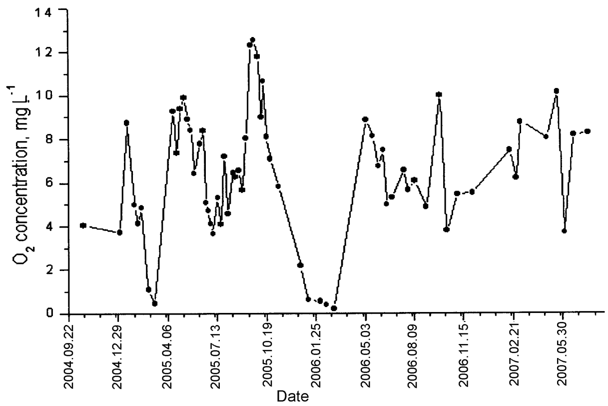

Fig. 2. A time-course of maximum oxygen concentrations present in its vertical profiles in the water column measured from 22 October 2004 to 19 July 2007.

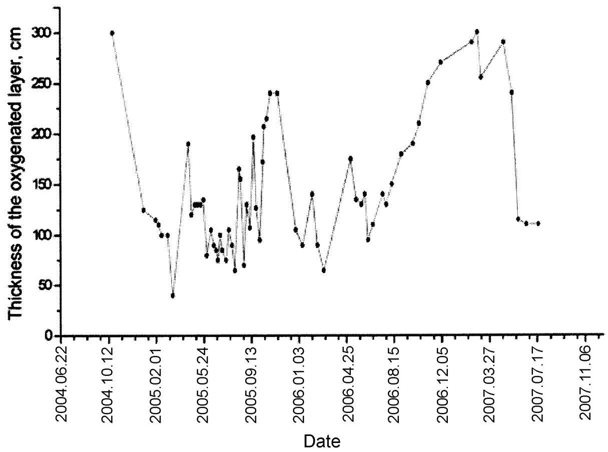

Fig. 3. A time-course of the thickness of the oxygenated water layer for the period of 22 October 2004 – 19 July 2007.

Red Lake formed at the end of January 2007. However, overcooling of shallow sediments during the long fall (when their temperature decreased below 4°C) led to reduction in sediment organics decomposition and, respectively, to a decrease in oxygen consumption. Therefore, at the end of winter on 8 March 2007, a rather thick upper layer of the water column was oxygenated and oxygen concentrations in water down to the 2-m depth varied in the range of 5–7.5 mg·L^{-1}. In April and May, colored surface waters of Red Lake were abundant in nutrients still remaining in that layer. This period is distinguished for rather large photosynthetic activity of phytoplankton and for elevated oxygen concentrations in water up to 9.9 mg·L^{-1}. In summer, surface waters of Red Lake behave as a nutrient-limited system [7, 8], and the respective oxygen concentrations were low: 3.65–5.30 mg·L^{-1} in 2005 and 5.0–6.56 mg·L^{-1} in 2006. Some rise in oxygen concentrations in surface waters in summer takes place after the arrival of rather cool air masses and the respective renewal of gravitational mixing. These processes became especially intensive in fall when the inflow of nutrients to the surface waters leads to the appearance of elevated oxygen concentrations above their saturation level in water (~12.56 mg·L^{-1} on 23 September 2005).

The thickness of the oxygenated water layer in Red Lake

The time-course of the thickness of the oxygenated water layer in Red Lake (the cutoff oxygen concentration – 0.1 mg·L^{-1}) is presented in Fig. 3. Data show that this thickness reaches its maximum values of some 250–300 cm in late fall due to gravitational mixing of the water column. Its elevated values (up to 190 cm on 17 April 2005) are also typical in spring. Surface water after ice melting becomes transparent and promotes the photosynthetic activity in the lower water layers. However, it lasts for a short period. Absorption of sun radiation by humic lower waters leads to their overheating and promotes an overturn and mixing of the whole water layer. Therefore, a surface water layer soon becomes colored and due to the rise in temperature gradients in metalimnion – again nutrient-limited.

The presented data on the time-course of the thickness of the oxygenated water layer in Red Lake show that this lake is meromictic. It consists of two different parts, which do not intermix: the upper monomictic part of Red Lake of some 300-cm thickness, which becomes completely aerobic due to the gravitational mixing in late fall and an anaerobic stagnant bottom water layer of some 150-cm thickness.

A time-course of conductivity of Red Lake surface waters

Data on the time-course of conductivity of the surface waters are presented in Fig. 4. Apparently, maximum conductivity values on 11 March 2005 (232 µS·cm^{-1}) and on 2 March 2006 (333 µS·cm^{-1}) in this course indicate a rise in ion concentrations under reductive conditions in the under-ice surface water. The respective maximum on 22 October 2004 (293 µS·cm^{-1}) may be due to the extreme mixing of the water column of some 300-cm thickness when near-bottom water layers with a large ionic content are involved. Minimum conductivity values in summer 2005 (104–120 µS·cm^{-1}) may be explained by peculiarities of weather conditions which suppressed vertical mixing of the upper water layer and preserved conductivity values typical of the end of spring flood. Elevated water conductivities were measured in summer 2006 (191–228 µS·cm^{-1}). It is a matter of fact that the impoundment dam was damaged in June 2006

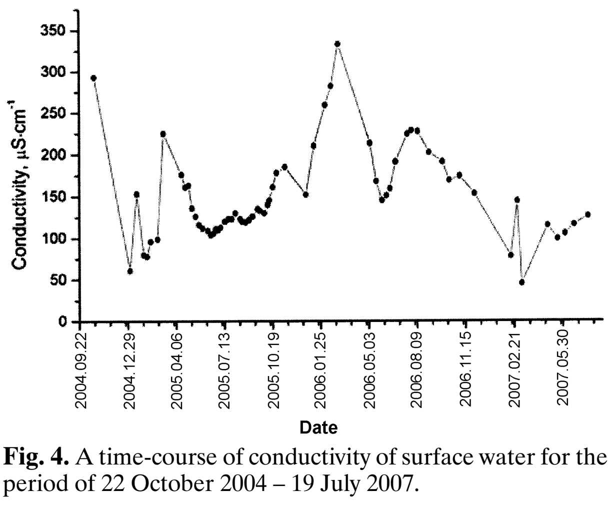

Fig. 4. A time-course of conductivity of surface water for the period of 22 October 2004 – 19 July 2007.

and later, the respective vertical profiles in Red Lake were measured at the decreasing water level. It dropped by some 40 cm below the ordinary summer level on 10 August 2006. After emergency repairs, this water level was restored at the beginning of September. Apparently, inflows from the swamp brook (*Ch1*) and the drainage ditch (*Ch2*) were responsible for this effect. A size of the impoundment is considerably larger than that of Red Lake, and under normal conditions it serves as a specific buffer suppressing water level variations in the lake. The respective characteristics of the impoundment sources are very different. Thus, organic acids were dominating in *Ch1* in April 2007. Its humic water was distinguished for low values of pH (~4.4), conductivity (~ 38–40 μS·cm^{-1}) and the dissolved solid concentration (~135 mg·L^{-1}). However, its discharge was very low (~1 L·s^{-1}). The elevated mineral content (the dissolved solid concentration ~348 mg·L^{-1}, conductivity ~391 μS·cm^{-1}) and pH ~7.54 (in April 2007) were characteristic of water in *Ch2*. Its discharge exceeded significantly that of *Ch1* and was equal to some 200 L·s^{-1}. Therefore, it is evident that the *Ch2* source was mainly responsible for restoration of the summer water level in Red Lake in August 2007.

Sedimentation

It is known [22] that in small lakes processes of sedimentation are more related to the topography of the basin, distance to the shore and the water depth. Site-specific sedimentation rates in Red Lake were estimated by studying peculiarities of the respective radiocesium vertical profiles in sediment cores. Visual observations showed that the under-water part of the marshy zone mat separating the open-water basin from banks consists of curtains of abundant algae, which are always destroyed in anaerobic waters in winter. The remainder of the dead algae is a crumbly black mass that sinks in water and promotes abundant sedimentation in the nearest bottom areas. At sites distanced from the marshy zone, sedimentation may be induced by the water column mixing. In this case oxygen penetration to the lower anaerobic water layers changes their status from reductive to oxidational one. Therefore, processes of gravitational mixing of the water column in Red Lake are followed by abundant Fe_2O_3 floc creation and sedimentation. However, sinking particulates are able only to reach the bottom areas belonging to the mixed water column and may be dissolved in the lower anaerobic water layers.

Radiocesium activity concentrations in Red Lake water

Measurement data show that radiocesium activity concentrations in the water column increase with depth (Fig. 5) and follow the same tendency of the dissolved solid concentration in water samples (Fig. 6). Data in Fig. 5 show practically the whole range of variations of these concentrations in colored water for the whole period of measurements in 2004–2007. The widest range of variations is related to the colored surface waters: 1.6±0.3 Bq·m^{-3} (on 2007.07.24) – 7.9±0.4 Bq·m^{-3} (on 2004.07.22) (not shown in Fig. 5). This range decreases with depth and radiocesium activity concentrations in the depth interval of 3.1–3.5 m vary from 7.3±0.4 Bq·m^{-3} (on 2006.03.16) to 9.9±0.8 Bq·m^{-3} (on 2007.07.24). It implies that such variations may be induced by the outside influence decreasing with depth. Data of the measurements carried out on 24 July 2007 are rather different. Although, radiocesium activity concentration in the water sample taken from the deepest depth interval (3.5–3.9 m) was the highest (~ 12.5±0.8 Bq·m^{-3}), the dissolved solid concentration was comparatively low (~286 mg·L^{-1}). Low radiocesium activity concentrations in surface water (1.6±0.3 Bq·m^{-3}) may be explained as effected by a number of factors. As it was mentioned above, an inflow from the impoundment source *Ch2* (Fig. 1b) due to its large discharge was mainly responsible for restoration of the water level in Red Lake in August 2006. Measurements of radiocesium activity

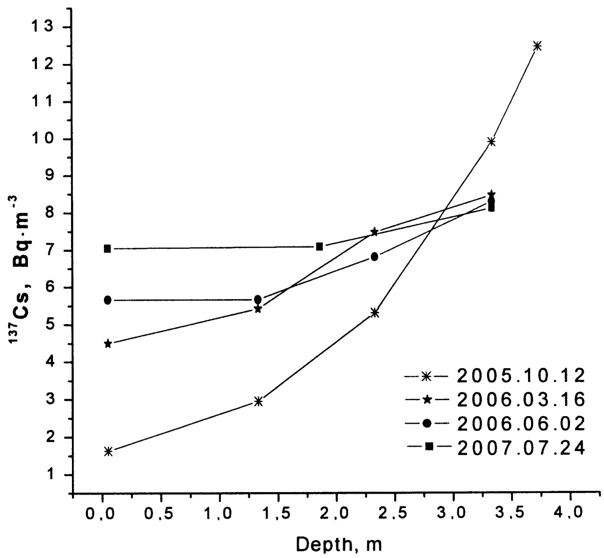

Fig. 5. Vertical profiles of radiocesium activity concentrations (Bq·m^{-3}) in the water column. Water samples were taken on 12 October 2005 (✳); 16 March 2006 (★); 2 June 2006 (•); 24 July 2007 (▪).

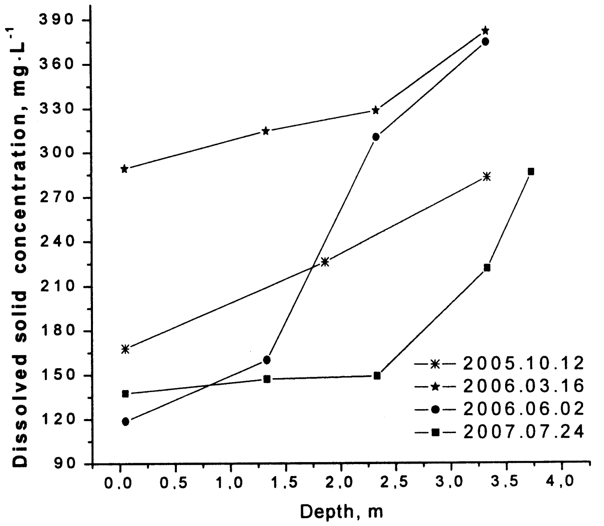

Fig. 6. Vertical profiles of dissolved solid concentrations (mg·L^{-1}) in the water column. Water samples were taken on 12 October 2005 (✳); 16 March 2006 (★); 2 June 2006 (•); 24 July 2007 (▪).

concentrations in *Ch1* and *Ch2* waters in April 2007 showed that they were equal to ~ 5.1±0.4 $Bq \cdot m^{-3}$ and ~ 1.0±0.3 $Bq \cdot m^{-3}$, respectively. Therefore, the inflow of less active waters from the drainage ditch *Ch2* in August 2006 caused the dilution of radiocesium activity concentrations in surface waters of Red Lake.

An analysis of the vertical profiles of standard water variables showed that the second factor that promoted low radiocesium activity concentrations in the upper layer of Red Lake (on 2007.07.24) was related to the abnormally high level of transparent melt-water in the lake at the beginning of spring mixing in 2007. Usually, seasonal variations of the water level in the lake for the period of our observations of 2004–2005 were rather low (±10 cm) with its maximum values in spring and minimum – in summer. Apparently, under conditions of the usual hydraulic regime of the impoundment, some part of melt-water with zero radiocesium activity concentrations was flushed before the beginning of spring vertical mixing in the lake. However, in spring 2007, an obstacle (a fallen pine tree) partially blocked the flushing water channel through the impoundment dam and raised the melt-water level in the lake. Because of spring mixing, and due to dilution, conductivity of the surface waters decreased down to 99 $\mu S \cdot cm^{-1}$ on 19 May 2007. Later, due to flushing, the water level in the lake slowly decreased. However, on 24 July 2007, it remained elevated (by ~7 cm) as compared with the usual spring water. Apparently, in both cases, the processes were followed by the removal of radiocesium from the lake and might be treated as its emergency self-cleaning mechanism. However, the vertical profile of radiocesium activity concentrations in Red Lake on 24 July 2007 (Fig. 5) showed that vertical mixing and dilution processes did not affect the situation in the lower water layer.

Data in Fig. 6 show that dissolved solid concentrations in colored surface water are minimum in summer (~119 $mg \cdot L^{-1}$ on 2 June 2006) and are maximum at the end of winter (~289 $mg \cdot L^{-1}$ on 16 March 2006). Their averaged values in the depth interval of 3.1–3.5 m were significantly larger: ~374 $mg \cdot L^{-1}$ on 2 June 2006 and ~382 $mg \cdot L^{-1}$ on 16 March 2006. The respective value on 24 July 2007 was smaller and out of the range (~221 $mg \cdot L^{-1}$). Possibly, a decrease in the dissolved solid concentrations in the lower water layer was related to specific conditions of oxygen migration down from the upper part of the lake water column in winter of 2006/2007 and formation of Fe_2O_3 floc deposits due to oxidation-reduction processes taking place under these conditions. Measurements showed that until 27 February 2007 the upper part of the water column was oxygenated down to the 3-m depth (the cutoff oxygen concentration ~0.1 $mg \cdot L^{-1}$). Oxygen penetration to the lower water layer was seen on the respective vertical profile as traces of its concentration (of the 0.08–0.09 $mg \cdot L^{-1}$ order) down to the 3.7-m depth.

By combining data on vertical profiles of the radiocesium activity and dissolved solid concentrations in the water column (Fig. 7) it is easy to see that in the surface layer radiocesium is mainly bound to water-soluble organic complexes. Thus, measurement data on 12 October 2005 showed a rise with depth in the dissolved solid concentration in the oxygenated water

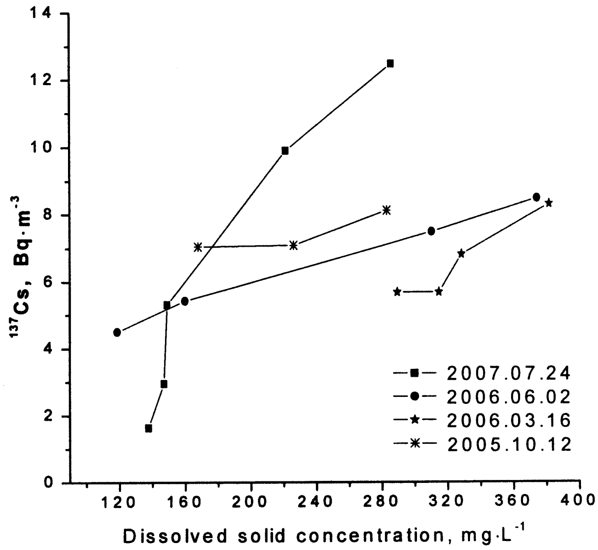

Fig. 7. Radiocesium activity and dissolved solid concentration relationship in water samples taken on 12 October 2005 (✳); 16 March 2006 (★); 2 June 2006 (•); 24 July 2007 (▪).

column with the almost constant radiocesium activity concentration. The same conclusion can be drawn studying the radiocesium activity concentration course with depth on 24 July 2007: a rise in its activity in the water layer of about constant dissolved solid concentration. This inference may also be evidenced in Fig. 8, where the vertical profile of dissolved solid concentrations on 24 July 2007 is presented as dependent on the respective data on water color. Color of water samples is expressed in Pt units (Pt, $mg \cdot L^{-1}$). It is estimated according to the recommendations of Cuthbert *et al.* [3] using light absorption coefficients of humic water at wavelength of 440 nm (the Beckman spectrophotometer UV 5270). Data in Fig. 8 show that a rather small increase in dissolved solid concentrations and significantly growing water color as well as the respective concentrations of humic substances [15] are characteristic of upper waters of Red Lake down to the 2.5-m depth. Presence of compounds of radiocesium with humic substances in dry deposits may also be evidenced due to temperature dependence of radiocesium content of the samples.

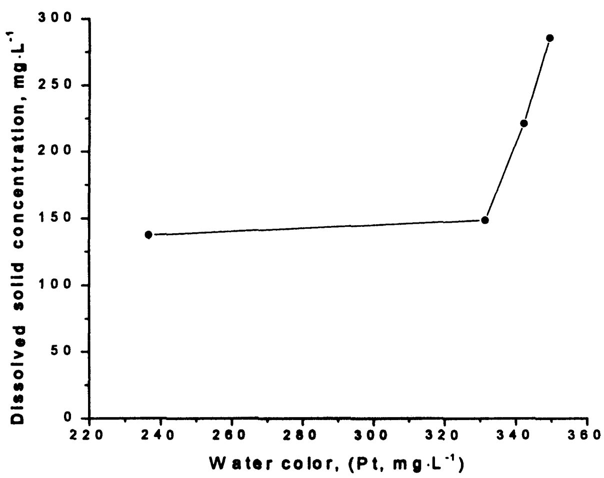

Fig. 8. Dissolved solid concentration ($mg \cdot L^{-1}$) and water color (Pt, $mg \cdot L^{-1}$) relationship in water samples taken on 24 July 2007.

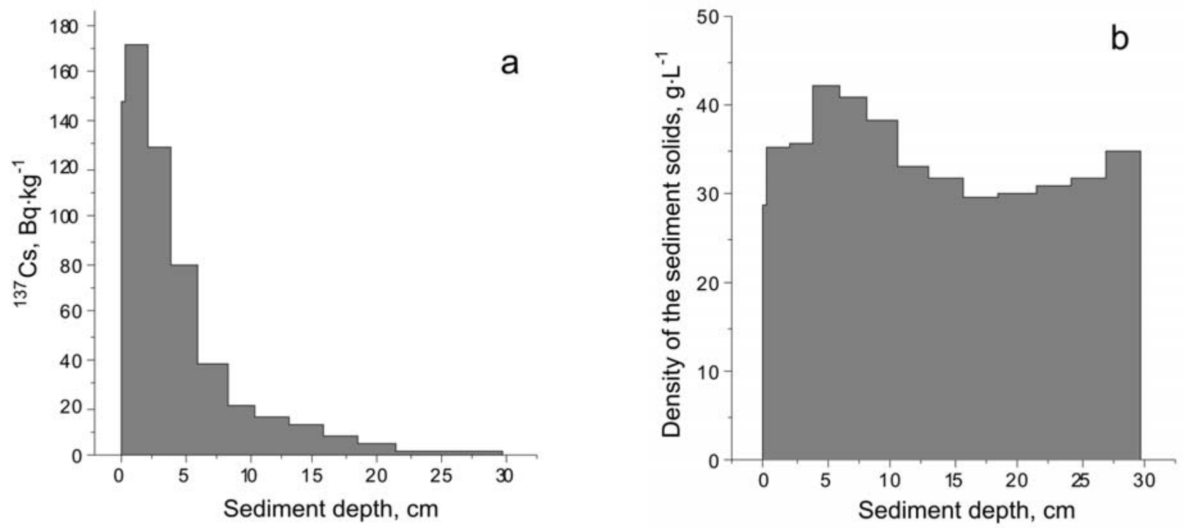

Fig. 9. Vertical profiles of radiocesium activity (Bq·kg^{-1}) (a) and sediment solid concentrations (g·L^{-1}) (b) in the sediment core sampled at the 4.5-m depth on 29 August 2004.

Thus, short-term heating of the samples of dry deposits up to 150°C reduces their radiocesium activity concentrations by the order of some 10%. Apparently, it is due to evaporation of the most volatile organic complexes associated with radiocesium.

Radiocesium activity concentrations in Red Lake sediments

Vertical profiles of radiocesium activity concentrations in sediments below the 3-m depth

Vertical profiles of radiocesium activity concentrations (Bq·kg^{-1}) in sediment cores (2 samples) taken in the deepest part of Red Lake are distinguished for its maximum activities in the sediment surface layer (Fig. 9a) and a strong decrease in these concentrations in deeper layers. A small amount of floc in the water layer above the sediment surface in the core taken on 29 August 2004 was deposited and further treated as an additional surface layer due to possible sedimentation. The sedimentation rate at this site was estimated by considering that the global fallout was responsible only for radioactive contamination of the deeper part of Red Lake. This rate amounted to ~0.06 cm·a^{-1}. However, this value may be significantly overestimated. Radiocesium load of two sediment cores was equal to ~370 and ~440 Bq·m^{-2}, respectively, and corresponded to the contamination level of the Lithuanian territory before the Chernobyl accident [4].

A small maximum at the sediment depth of 4–8 cm in the vertical profile of the density of sediment solids in the core taken on 29 August 2004 (Fig. 9b) may be explained by the appearance of the remainders of shells – evidence of the former transparent water regime of Red Lake.

Vertical profiles of radiocesium activity concentrations (Bq·kg^{-1}, Bq·L^{-1}) in sediment cores (3 samples) taken at some distances from the deepest site of Red Lake are different. Elevated sedimentation rates (~ 0.21–0.35 cm·a^{-1}) and the deepening of radiocesium peak activities to the 3.5–8.5-cm depth interval are characteristic of them. These vertical profiles in one of the cores taken at the 4.1-m depth on 24 July 2007 are shown in Figs. 10a and 10b. A typical maximum in the vertical profile of the density of sediment solids (Fig. 10c) moved to the 12.8–19.5-cm depth interval. The remainders of shells were seen in the eighth sediment layer (12.8–14.9-cm depth interval). Radiocesium loads of the cores were estimated to be in the range of ~ 460–640 Bq·m^{-2}.

By fitting the slopes of vertical profiles of radiocesium activity concentrations in sediment cores below its peak activities (Bq·kg^{-1}) to the Gauss shape it was determined that a characteristic width of slopes (at their half-heights) varied in the range of 3.4–4.5 cm. These small shifts in the shape of slopes from the

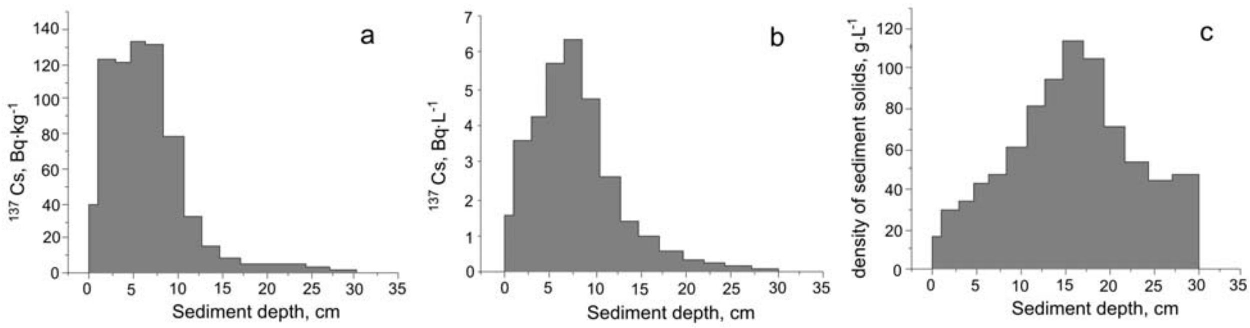

Fig. 10. Vertical profiles of radiocesium activity (Bq·kg^{-1}) (a), (Bq·L^{-1}) (b) and sediment solid concentrations (g·L^{-1}) (c) in the sediment core sampled at the 3.9-m depth on 24 July 2007.

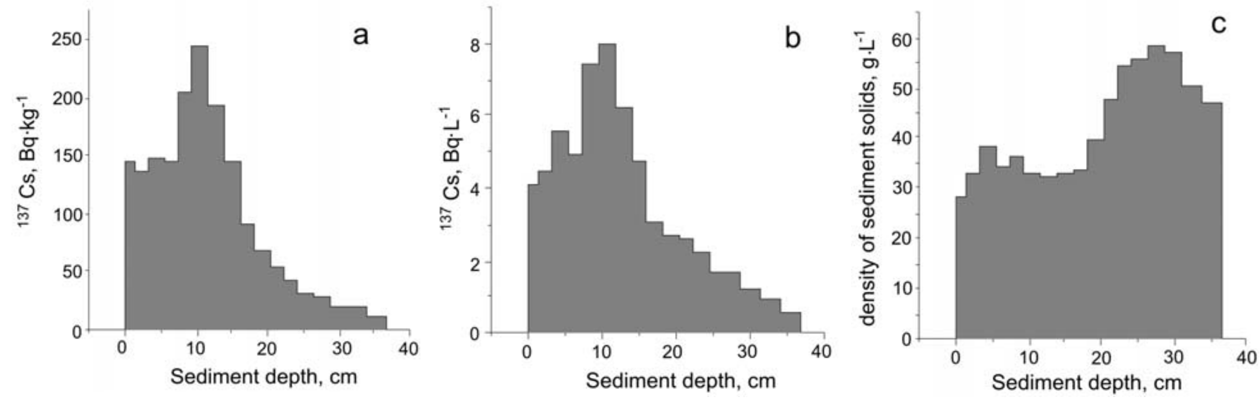

Fig. 11. Vertical profiles of radiocesium activity (Bq·kg^{-1}) (a), (Bq·L^{-1}) (b) and sediment solid concentrations (g·L^{-1}) (c) in the sediment core sampled at the 1.9-m depth on 6 August 2007.

primary vertical profiles formed during the fast phase of radiocesium free-ion diffusion [12, 21] show rather limited possibilities of radiocesium migration to deeper sediments under these conditions.

Vertical profiles of radiocesium activity concentrations in sediments above the 3-m depth

Concerning the presence of abundant marshy zones in Red Lake, sediment cores (7 samples) were taken in bottom areas of the upper water layer at 1.9–3-m depth. Vertical profiles of radiocesium activity concentrations in these sediment cores are distinguished for their elevated deepening in sediments. Therefore, only two of them – taken at the 1.9-m depth near the marshy zone on 6 August 2007 (Figs. 11a, 11b) and taken at the largest depth (~3 m) on 2 June 2006 – fitted to the height of our sampler (~40 cm). Maximum radiocesium activity concentrations in the respective vertical profiles of the two latter cores were measured in the 9.5–11.7-cm (Figs. 11a, 11b) and into the 8.5–11.1-cm depth intervals. The characteristic width of slopes was also larger (~6.2 and ~5.7 cm, respectively). The structure of vertical profiles of the density of sediment solids remained. However, maximum densities of sediment solids were measured in deeper layers: in the 26.4–31.1-cm depth interval in the sediment core taken at the 1.9-m bottom depth (Fig. 11c) and in the 25.2–29.5-cm depth interval in the core taken at the 3-m bottom depth. Estimated sedimentation rates were equal to ~0.5 and ~0.49 cm·a^{-1}, respectively. Radiocesium loads in these cores were also elevated ~1300 and ~1720 Bq·m^{-2}, respectively. It means a significant (~ 3–4-fold) increase in this load as compared to the core taken in the deepest area of Red Lake.

Broadened peaks of radiocesium activity concentrations shifted to larger (11.1–20.6 cm) depth intervals are characteristic of the respective vertical profiles measured in the other sediment cores taken at the 2–2.5-m bottom depth (Figs. 12a and 12b). These profiles decline gently in deeper sediments with the specific width of their slopes in the range of 7–12.2 cm showing elevated radiocesium mobility in these cores. We tried to estimate the loss of the radiocesium load due to "tails" of its vertical profiles that did not fit to the height of the sampler. For this aim, one of the cores was sampled in the same bottom area without weight compensation with the loss of the surface sediment layer of about 14–15-cm thickness. These measurement data showed that the radiocesium load in the ordinary lost "tails" of the profiles may be of the ~10% order. Therefore, radiocesium loads measured in the latter five cores (~ 1530–2150 Bq·m^{-2}) were underestimated by this order. Contrary to the former data, vertical profiles of the density of sediment solids (Fig. 12c) showed only a slight rise with the sediment depth due to the compaction indicating them to be adequately mixed. Sedimentation rates in these sediment cores were in the range of ~ 0.62–0.96 cm·a^{-1}.

The appearance of the bottom zones in the 2–2.5-m depth interval with the enhanced radiocesium migration may be explained by the elevated bioturbation induced by tench (*Tinca tinca*) – the main inhabitant of Red

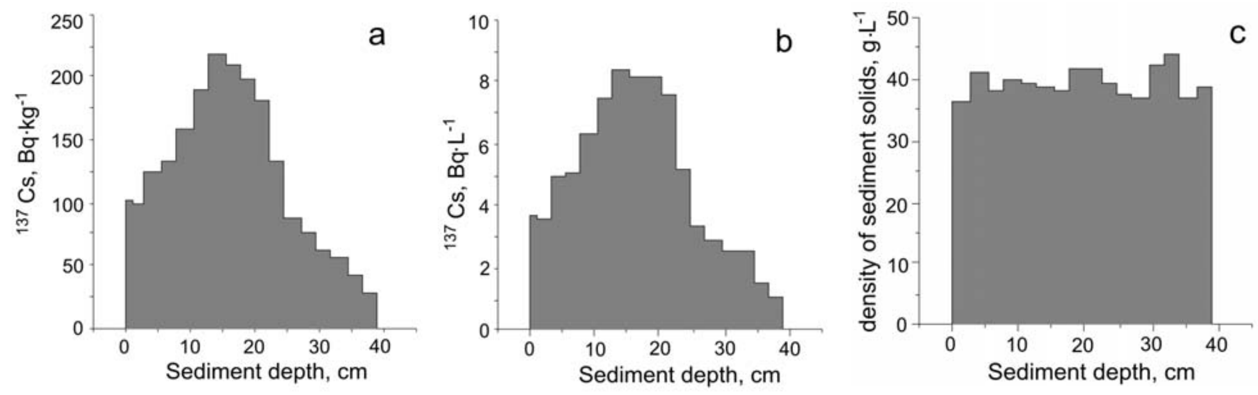

Fig. 12. Vertical profiles of radiocesium activity (Bq·kg^{-1}) (a), (Bq·L^{-1}) (b) and sediment solid concentrations (g·L^{-1}) (c) in the sediment core sampled at the 2.5-m depth on 30 June 2005.

Lake. It is known [24] that the biological cycle of this fish includes a period of the anabiotic winter torpor. In the late fall when the water column of the upper layer is completely oxygenated, tench sink to the bottom and settle in the silt sediments. Tench become active in spring when temperature of the sediment surface rises above 4°C. At first, bottom zones of the anabiotic winter torpor of tench were determined by our portable meter in late fall as areas of anomalous instability of standard water parameters indicating intense movements in the near-bottom waters. Bioturbation of the surface sediments induced by tench is followed by their resuspension and redistribution over the other below lying bottom areas along the bottom slope. Simultaneously, it leads to broadening and smoothing of vertical profiles of radiocesium activity concentrations and the density of sediment solids. These processes result in a decrease in the maximum radiocesium activity concentrations in sediments, their shifts to deeper sediments and overestimation of sedimentation rates if this calculation is based on the shape of radiocesium vertical profiles. Resuspended materials deposited in the lower water layer may be wrongly treated as a sedimentation flux of sinking particles formed in the lake surface layer and the water column.

Discussion

Modeling studies on the radiocesium behavior in closed lakes [2, 13, 19] did not account for all important effects related to the impact of lake catchments (humic water inflows, the behavior of radiocesium associated with the dissolved humic substances). The present study carried out in a small humic lake shows that the impact of the humic water inflow on the transparent water system can dramatically change its vertical structure. This shallow lake became meromictic. Formation of the lower stagnant anaerobic water layer separated from the upper one before the Chernobyl accident led to its lower radiocesium load only due to nuclear weapons fallout. Further nuclear weapon and Chernobyl fallouts increased significantly the radiocesium contamination of the upper lake waters and its load in the respective sediments.

The results of the study show that occasional changes of the impoundment hydraulic regime – the retarded discharge in spring as well as the enhanced short-term flushing – induce an emergency mechanism of Red Lake self-cleaning from radiocesium. However, this self--cleaning mechanism can only act in the upper part of the lake water column and does not influence elevated radiocesium activity concentrations in the lower water layer. Data show that these events are not able to change the specific vertical structure of Red Lake. Apparently, the spring dilution mechanism is typical of closed humic lakes inciting large variations of radiocesium activity concentrations in the surface water. It seems that in the case of a normal hydraulic regime of the impoundment when melt-water is partially flushed before the beginning of spring vertical mixing, Red Lake may be treated as a "conservative" system for radiocesium activity concentrations in the surface layer and as an analogous model of closed humic lakes. Apparently, as in the case of Red Lake, the vertical structure of closed humic lakes after severe accidents may induce the formation of the extreme radiological situations in limited layers of the water column. As a result, closed lakes with humic water turn into the repository of highly mobile radiocesium in the ionic form and associated with humic substances. Measurements showed that radiocesium water-soluble concentrations in Red Lake were the highest among the neighboring lakes [20, 21].

Resuspension of sediments due to the tench activity at the bottom of the upper water layer in the late fall and spring led to their redistribution distorting originally formed vertical profiles of radiocesium in sediments. These disturbances in the formation of radiocesium vertical profiles in sediments distort assessments of its migration, diffusion coefficients and estimates of sedimentation rates. Therefore, elevated sedimentation rates (~ 0.62–$0.96\ \mathrm{cm \cdot a^{-1}}$) estimated using radiocesium vertical profiles in sediment cores sampled in the bottom areas of the tench winter torpor are doubtful. Occasional may also be information on sedimentation rates in the other bottom areas of the upper and lower lake layers affected by processes of sediment redistribution along the bottom slope. Apparently, knowledge of seasonal variations of the lake vertical structure, which is mainly controlled by water transparency and color, as well as the most possible description of sampling sites and fish type inhabiting the lake is urgent during the modeling studies.

Crucian carp (*Carassius carassius*) may induce the same effects at the bottom in other lakes [24]. It is known that tench and crucian carp are hobby fishes of amateur anglers. They are also the object of industrial fishing in Lithuania. Considering their long-term winter torpor in radionuclide-contaminated sediments, the estimation of radiological consequences related to such behavior is urgent. Closed lakes with humic water are, as it was shown above, of special concern. It is evident that radionuclide behavior in these water bodies needs a detailed study.

Events of humic water impacts are typical of neighboring lakes Tapeliai, Juodis and Balžis during the spring flood and rainy periods. These events are always followed by distinct short-term distortion in the lake vertical structure inducing the formation of stagnant conditions in the water column. However, processes of surface water exchange related to their rather large flushing fluxes help them to recover.

Conclusions

Among the other water bodies affected by the same radiological impact, closed humic lakes may represent the hazardous case of radioecological consequences for their inhabitants. A distinct vertical structure of such lakes allows the formation of extreme radionuclide concentrations in the water column and boundary sediments in limited parts of these water bodies. Assessing the radiological risk to humans after radionuclide deposition events, closed humic lakes may be treated as critical objects.

Estimations of sedimentary chronology and sedimentation rates in lakes based on the vertical profiles of radiocesium activity concentrations in sediments may be inaccurate if they are not accounting for their possible distortion by tench and crucian carp. The largest deepening of radiocesium into the sediments is related to bioturbation produced by ground fishes at their favorite sites of the winter torpor. Radiocesium migration abilities into deeper sediments are minimum in the lower anoxic layers of Red Lake. Sedimentation in the deepest site of Red Lake may be mainly provided by floc deposits formed due to oxidation of anaerobic waters in its upper layer. Elevated sedimentation at the intermediate depths (~ 2–4 m) may be only seeming and related to redistribution of the surface sediments along its bottom slope from favorite bottom areas of the ground fish winter torpor.

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References

1. Andersson E, Sobek S (2006) Comparison of a mass balance and an ecosystem model approach when evaluating the carbon cycling in a lake ecosystem. Ambio 35;8:476–483
2. Bulgakov AA, Konoplev AV, Smith JT *et al.* (2002) Modelling the long-term dynamics of radiocaesium in closed lakes. J Environ Radioact 61;1:41–53
3. Cuthbert ID, Giorgio P (1992) Toward a standard method of measuring color in freshwater. Limnol Oceanogr 37;6:1319–1326
4. Dauskurdis S, Tamulėnaitė O, Nedveckaitė T (1989) The ^{137}Cs and ^{90}Sr distribution in edible mushrooms on the Lithuanian SSR territory. Atmospheric Phys 14:119–127 (in Russian)
5. Grižienė G, Jablonskis J, Januševičius S *et al.* (1993) Hydrography of the Neris river. Energetika 1:20–41 (in Lithuanian)
6. Gudelis A, Remeikis V, Plukis A, Lukauskas D (2000) Efficiency calibration of HPGe detectors for measuring environmental samples. Environ Chem Phys 22;3/4:117–125
7. Guildford SJ, Hecky RE (2000) Total nitrogen, total phosphorus, and nutrient limitation in lakes and oceans: is there a common relationship? Limnol Oceanogr 45;6:1213–1223
8. Howarth RW, Marino R (2006) Nitrogen as the limiting nutrient for eutrophication in coastal marine ecosystems: evolving views over three decades. Limnol Oceanogr 51;(1, part 2):364–376
9. Ilus E, Saxén R (2005) Accumulation of Chernobyl-derived ^{137}Cs in bottom sediments of some Finnish lakes. J Environ Radioact 82;2:199–221
10. Jones RI (1992) The influence of humic substances on lacustrine planktonic food chains. Hydrobiologia 229:73–91
11. Jonsson A, Meili M, Bergström AK, Jansson M (2001) Whole-lake mineralization of allochthonous and autochthonous organic carbon in a large humic lake (Örträsket, N. Sweden). Limnol Oceanogr 46;7:1691–1700
12. Kirikopoulos IL, Ioannides KG, Karamanis DT, Stamoulis KC, Kondoura EM, Mantzios AS (1994) Kinetics of radiocesium sorption in lake sediments. Health Phys 66;1:36–42
13. Monte L, Grimani C, Desideri D, Angeli G (2005) Modelling the long-term behaviour of radiocesium and radiostrontium in two Italian lakes. J Environ Radioact 80:105–123
14. Morris DP, Zagarese H, Williamson CE *et al.* (1995) The attenuation of solar UV radiation in lakes and the role of dissolved organic carbon. Limnol Oceanogr 40;8:1381–1391
15. Nürnberg GK, Shaw M (1999) Productivity of clear and humic lakes: nutrients, phytoplankton, bacteria. Hydrobiologia 382:97–112
16. Santschi PH, Bollhalder S, Farrenkothen K, Lueck A, Zingg S, Sturm M (1988) Chernobyl radionuclides in the environment: tracers for the tight coupling of atmosphere, terrestrial, and aquatic geochemical processes. Environ Sci Technol 22:510–516
17. Santschi PH, Bollhalder S, Zingg S, Lück A, Farrenkothen K (1990) The self-cleaning capacity of surface waters after radioactive fallout. Evidence from European waters after Chernobyl, 1986–1988. Environ Sci Technol 24:519–527
18. Sobek S, Söderbäck B, Karlsson S, Andersson E, Brunberg AK (2006) A carbon budget of a small humic lake: an example of the importance of lakes for organic matter cycling in boreal catchments. Ambio 35:8:469–475
19. Smith JT, Belova NV, Bulgakov AA *et al.* (2005) The "AQUASCOPE" simplified model for predicting ^{89,90}Sr, ^{131}I, ^{134,137}Cs in surface waters after a large-scale radioactive fallout. Health Phys 89;6:628–644
20. Tarasiuk N, Koviazina E, Kubarevičienė V (2008) On seasonal variations of radiocesium speciation in the surface sediments of Lake Juodis, Lithuania. J Environ Radioact 99;1:199–210
21. Tarasiuk N, Koviazina E, Kubarevičienė V, Shliahtich E (2007) On the radiocesium carbonate barrier in organics-rich sediments of Lake Juodis, Lithuania. J Environ Radioact 93;2:100–118
22. Terasmaa J (2005) Bottom topography and sediment lithology in two small lakes in Estonia. Proc Estonian Acad Biol Ecol 54;3:171–189
23. Vakulovski SM, Gaziev YaI, Kolesnikova LV, Petrenko GI, Tertyshnik EG (2006) ^{137}Cs and ^{90}Sr in the surface water bodies of Bryansk region. Atomnaya Energiya 100;1:68–74 (in Russian)
24. Virbickas J (2000) Fishes of Lithuania. Trys Žvaigždutės, Vilnius (in Lithuanian)
25. Wrona FJ, Prowse TD, Reist JD *et al.* (2006) Effects of ultraviolet radiation and contaminent-related stressors on Arctic freshwater ecosystems. Ambio 35;7:388–401