VILNIUS GEDIMINAS TECHNICAL UNIVERSITY

Anastasija MOISEJENKOVA

RESEARCH OF DYNAMICS OF RADIOCESIUM AND HYDROPHYSICAL PARAMETERS IN THE LITHUANIAN LAKES

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Introduction

Topicality of the problem

Nuclear test fallouts and atmospheric deposits after nuclear power plant accidents (Chernobyl, Fukushima and so on) converted many water bodies all over the world into a repository of radionuclides. It is known that after the radioactive impact radionuclides are quickly (for some months) removed from the lake water and accumulated in the bottom sediments. However, that radionuclide burial in lake sediments is not terminal. With time, due to processes of lake self-cleaning, running through lakes turn into a secondary source of radionuclides for water systems. This problem is very urgent for Lithuania, which is a laky country where there are almost 6000 lakes and impoundments. Besides, it is known that in the nearest future Lithuania may occur in the environment of three nuclear power stations. The one way to tackle ecological consequences of radionuclide inputs to the open water bodies is a thorough study of the radionuclide behavior and of mechanisms of lake and impoundment self-cleaning.

The present work is related to radiocesium (^{137}Cs) behavior in some Lithuanian lakes and especially in super warm ones. In making choice of the water bodies for a study, we followed the lake thermal classification suggested by K. Kilkus. This classification is related to the processes adjusting temperatures of the near-bottom water in winter. It defines the lakes with elevated temperatures (above 4 °C) of the near-bottom water as super warm ones. Our choice is not occasional. Elevated temperatures of the lake near-bottom water consider also its elevated mineralization and a somewhat mechanism of the water enrichment in impurities.

It is known that during the winter in most lakes a near-bottom anaerobic zone is formed. An appearance of the zone is related to processes of oxygen consumption in the thermally stratified water column due to bottom sediment organics decomposition. For many years, it was believed that elevated mineralization (as well as elevated radiocesium concentrations) of near-bottom waters in winter were only related to the anaerobic zone formation. However, some data showed that elevated concentrations of pollutants in the water column were also possible under aerobic conditions in autumn. Evidently, near-bottom waters with elevated concentrations of pollutants are the main self-cleaning potential of the running through water bodies. Thus, during spring and autumn overturns of dimictic lakes, water pollutants are evenly distributed in the water column and partially can leave the lake with the flushing waters. It means that running through super warm lakes can act as a most powerful secondary source of radiocesium for outflowing rivers.

Object of the research

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

Aim and tasks of the work

The aim of the research is a dynamics of radiocesium in a relationship with the seasonal variations of hydrophysical parameters in bottom sediments and water of super warm Lithuanian lakes.

1. To study the seasonal variations of standard water parameters in a relationship with the water-soluble radiocesium activity concentrations in a dimictic super warm lake (lake Juodis (54°46'49"N, 25°26'29"E) for the aim to determine a mechanism of the enrichment in radiocesium of near-bottom water in winter.
2. To study a radiocesium behavior in a super warm meromictic lake (lake Lydekinis (54°46'11"N, 25°27'23"E) in a relationship with the seasonal variations of standard water parameters.
3. To study the seasonal variations of the thermal structure in lake Tapeliai (54°46'28"N, 25°26'45"E).
4. To study the radiocesium behavior in lake Tapeliai for the aim to evaluate the possibility of the participation of the sediment interstitial liquid buoyancy mechanism in the near-bottom water enrichment in radiocesium in winter.
5. Refine of the measurements of the experimental hydrophysical parameters of water and water and sediment sampling techniques.

Methodology of research includes ion-exchange method using for water hydrophysical parameter measurement and gamma spectrometry method used for determination of radiocesium distribution in sediments and water.

Scientific novelty

For the first time, it is shown that a mechanism of the near-bottom water enrichment in radiocesium in super warm lakes in winter is of the thermodynamic origin and is related to buoyancy of the sediment interstitial liquids. It indicates that lake self-cleaning from pollutants is closely related to the thermal processes occurring in the lake.

For the first time, it is shown that this mechanism starts under aerobic conditions and becomes one of the causes of the formation of the anaerobic zone in super warm lakes.

Practical value

Detection of the thermodynamic mechanism of the lake near-bottom water enrichment in radiocesium allows explaining a super warm lake phenomenon and their feature as a secondary radiocesium and the other anthropogenic pollution source for water systems. 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.

Defended propositions

1. A thermodynamic mechanism – buoyancy of the sediment interstitial liquids – is responsible for the near-bottom water enrichment in radiocesium in super warm lakes in winter. Estimating radioecological consequences of the radioactive impact to the natural water bodies, meromictic lakes must be treated as critical objects.
2. This mechanism reveals itself also under aerobic conditions in autumn and is one of the causes of the anaerobic zone formation in the near-bottom waters in winter. Buoyancy itself is responsible for surface sediment mixing and the shifts of the beginning of the temperature gradient in sediments.
3. The thermal lake classification suggested by Kilkus is quite formal and must be improved involving a specific category of lakes with the intermittent thermal regime of the near-bottom water in winter.

The scope of the scientific work

The dissertation consists of an introduction, 5 chapters, general conclusions and recommendations, list of quoted references, and the author′s publications on dissertation issues.

The scope of the work is 128 pages, 27 numbered formules used in the text, 74 figures and 3 tables, used 118 literature sources.

1. Analysis of radiocesium dynamics in lake water and bottom sediment

An analysis of the radiocesium behavior in water bodies (literature data) shows that this nuclide is presented in a number of different physico-chemical forms. In a water column, it is known to be in a water-soluble form and associated with the suspended matter. In water of studied lakes, concentrations of suspended particles were always below 1 $g \cdot m^{-3}$ and the radiocesium activity concentrations related to the suspended substances were always below the detection limit (~0,010 Bq), respectively. Therefore, in our study a radiocesium water-soluble form was only treated.

In sediments, apart from the very small radiocesium water-soluble fraction this radionuclide is presented in exchangeable (mobile), potentially mobile fractions (associated with carbonates, iron and manganese oxides and organics) and in the residual (associated with the sediment matrix) fraction. Due to their large accumulation abilities, lake sediments are the main repository of radiocesium. With time, due to the presence in the sediments of water-soluble, mobile and potentially mobile radiocesium fractions and the acting sediment self-cleaning mechanism, lakes turn into the secondary source of this radionuclide.

2. Water and bottom sediment sampling and methodology of radiocesium determination

Lake Tapeliai (54°46'28"N, 25°26'45"E) (Fig. 1.1) consists of four sections: a) a southern shallow terrace (depths ~4−5 m); b) a central deepest part of the lake (depths ~ 7−9 m); c) a northern terrace with a gradual bottom deepening from ~1,5 down to ~6 m depths; d) a small bottom terrace in the western side of the lake (depths ~5−6 m).

Lake Juodis (54°46'49" N, 25º26'29"E) is located 16 km to the northeast from Vilnius city in a wooded region (Fig. 1.1). It is a small (~0,1 km^2) running shallow lake in the lake chain connected by a brook. Its banks are rush-grown, with large marshy zones formed at the brook inflow and outflow areas. The Lake Juodis basin is of the glacier origin (tunnel valley lake, groove type) and consists of two parts. The southern part of the lake is wider and deeper (up to 3,5 m deep). The northern one is a shallow bottom terrace (depth ~1,0−1,7 m).

All studied lakes are mainly screened from the wind by the forestry environment and, as a rule, an only small undulation is possible in Lake Tapeliai at southern winds.

Water of the small (2,3 ha) humic Lake Lydekinis (54°46'11"N, 25°27'23"E) (Fig. 1.1) is highly colored. It was denominated as a 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.

Vertical profiles of standard water parameters (pH, temperature, conductivity, oxygen concentration) were episodically measured in the lakes water. 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, stabilized by an anchor. In winter, holes were drilled in ice.

The Molchanov type bathometer was used for water samples (~20 l in total). In this case, parameters of water samples were averaged over the 40 cm depth interval of the sampler. After delivery to the laboratory, only the aerobic 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 flock 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), which were analyzed for the radiocesium content.

Fig. 1.1. Location places of lakes

Sediment cores were taken using the Ekman-Birge type sampler. It was 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 and the improved version of the ~40 cm height 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 (further as total dissolved solids – TDS) 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 ±5%.

Sediment samples were analyzed 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). Dry deposits of water samples were analyzed for ^{137}Cs using an ORTEC γ-spectrometric system with a HPGe well-type detector (a sensitive volume of 170 cm^3, a relative efficiency of 38%, resolution – 2,05 keV/1,33 MeV). Measurement errors of radiocesium activity concentrations in samples were evaluated by the GAMMAVISION software program.

3. Analysis of radiocesium behaviour correlation with the seasonal variations of hydro physical water parameters

3.1. Research of vertical structure of super warm lake Juodis

J. S. Turner (1973) explained the presence of a layered structure in the water column in some lakes in winter by the specific feature of the transfer of buoyancy convective motions. He reviewed the theory and results of experiments on convective motions of fluids in double-diffusive systems of temperature and salinity. Such systems with positive gradients (with depth) of those components were considered as a simulation of a stagnant water column – sediment system. However, those modeling studies dealt with heated solid surfaces whereas the sediment surface is a porous media. In this study, using numerous vertical profiles of temperature, conductivity and oxygen in super warm Lake Juodis, we have attempted to elaborate causes and principles of thermodynamic processes related to radiocesium behavior during autumn-winter seasons.

During warm seasons, lake sediments act as a heat accumulator. As a rule, serious interruptions in sediment heating begin in August after the arrival of cool air masses. Further, following the onset of permanently cooling weather in autumn, a positive temperature gradient (with depth) is established in the sediments (Fig. 3.1). Due to strong cooling, the downward temperature gradient may be also positioned below the sediment surface. An arrival of warm air masses (these events are very frequent in autumn in Baltic countries) is followed by the formation of thermal stratification of the water column. It leads to a decrease in the heat transfer through the water column (thermal diffusivities of water column due convective motions are significantly larger than molecular ones) and heat accumulation in the sediment surface layer. According to Turner (1973), a steep temperature gradient in the sediment-bottom water interface reaching some critical value induces buoyancy of the sediment interstitial liquid enriched in dissolved materials stimulating upward convective motions and intrusions of the interstitial liquid to the near-bottom water. It is known that radiocesium activity concentrations in anaerobic interstitial water of surface sediments are significantly larger than those in the near-bottom water. Therefore, intrusions of interstitial liquids to the near-bottom areas must induce elevated radiocesium concentrations in the respective waters. It explains elevated radioactivity concentrations (~6,8 Bq·m^{-3}) measured in the outflowing brook in autumn (~2 Bq·m^{-3} in summer) (Tarasiuk *et al.*, 2002).

Transferring consequences of these rather short-term events to the long-term winter period when the lacustrine system is thermally isolated by its ice cover, it is easy to see that a rise in dissolved material in the near-bottom waters allows stable density stratification at temperatures higher than 4 °C.

When the beginning of the temperature gradient is positioned below the sediment surface in the absence of convective motions in a temperature stratified water column, a thin overlying layer of sediments becomes also a significant obstacle to heat flux. This, in turn, must lead to the strengthening of the temperature gradient followed by buoyancy of the interstitial liquids. In this case, buoyancy itself inducing upward motions of the pore water increases thermal diffusivity of the surface sediments and allows the upward movement of the beginning of the temperature gradient.

According to Turner (1973), consequences of buoyancy effects in a stagnant water column induce its specific layered structure. This structure in Lake Juodis was also determined. The data suggest that a stagnant water column consists of a set of evenly mixed water layers, which always disappear during last stages of heat redistribution processes.

An analysis of the measurement data shows that intrusions of the sediment interstitial liquids to the near-bottom water and the formation of the near-bottom zone with elevated mineralization (elevated radiocesium activity concentrations) are a precursor of the near-bottom anoxic zone formation. Thus, the thickness of the overheated near-bottom water layer (a layer of warmer water below the 4 °C isotherm) and that of anoxic waters in winter (a cutoff concentration of oxygen at 0,1 mg·l^{-1}) do not coincide. The depth of the 4 °C isotherm defining the upper water level affected by sediment interstitial liquid intrusions is, as a rule, above the anoxic zone. This implies that elevated radiocesium concentrations may also be typical of aerobic water layers up to the depth of the 4 °C isotherm. Data on 24 March 2006, where the thickness of the anaerobic water layer was larger than that of the overheated water, were atypical. They were measured under conditions of a substantial decrease in the photosynthetic activity of phytoplankton incited by a thick layer of snow (~21 cm) and the ice cover (~46 cm). Generally, data presented in Fig. 3.1 imply that the processes of vertical transfer of interstitial liquids through the water column preceded those of anoxic zone formation and exceeded them in rate. Therefore, it is highly probable that the anaerobic zone in super warm lakes in winter is mainly due to the intrusions of sediment pore water with its elevated concentrations of reduced ions. We succeed estimating the possible consequences of interstitial liquid intrusions on the radiocesium activity concentrations in the near-bottom waters under conditions of the oxygenated water column.

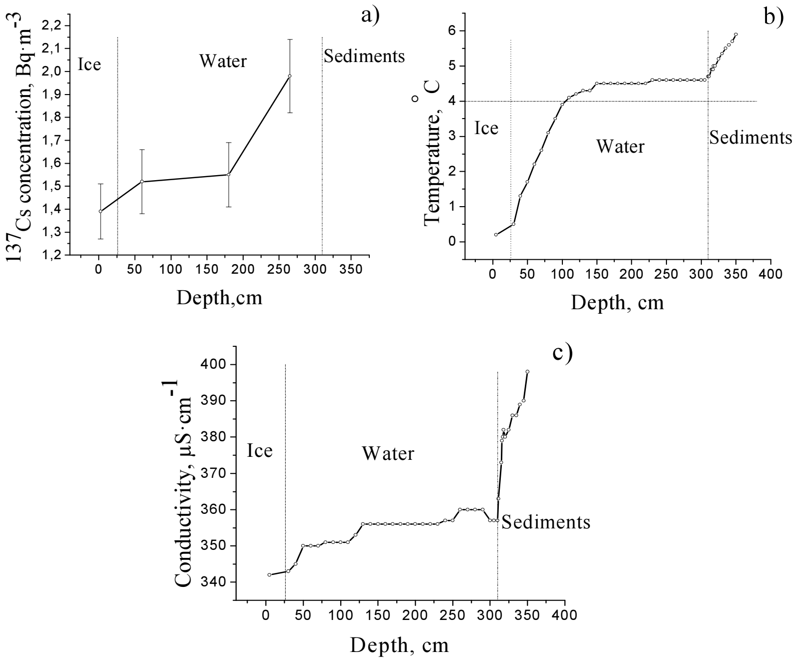

Fig. 3.1. Vertical profiles of ^{137}Cs activity concentrations ($Bq \cdot m^{-3}$) (a) as well as of temperature (b) and conductivity ($\mu S \cdot cm^{-1}$) (c) measured in the stagnant water column at station S2 (the bottom depth ~310 cm) on 12 February 2009; (the thickness of ice – 26 cm, snow layer thickness – ~1 cm).

Water samples from different depths were taken in lake Juodis on 12 February 2009 in the temperature stratified water column under aerobic conditions induced by extreme photosynthetic activity under a thin layer of snow and the ice cover (Fig. 3.1 (a–c)). The vertical profile of ^{137}Cs activity concentrations (Fig. 3.1 (a)) shows a distinct (~28 %) rise corresponding with a water temperature of 4,6 °C (Fig. 3.1(b)) in the 2,45−2,85 m depth interval. The ^{137}Cs activity concentrations within the 0,6−1,0 and 1,6−2,0 m depth intervals are about equal (in the 1,5−1,6 $Bq \cdot m^{-3}$ interval), as are the concentrations of TDS in those layers (~210 $mg \cdot l^{-1}$). This may imply that some water layer below the 4 °C isotherm is metastable and only affected by heat flux without any TDS rise.

The respective concentration of TDS in the 2,45–2,85 m interval was equal to ~214 $mg \cdot l^{-1}$. Some decrease in conductivity in the near-bottom water layer (Fig.3.1 (c)) is often due to the elevated photosynthetic activity of green algae covering the sediment surface. A vertical profile of oxygen concentrations due to a sensor cable failure was measured later under worse oxygenation condition on 15 February 2009. Data showed that all water column was aerobic.

These data evidenced that the enrichment in radiocesium of the near-bottom water due to the sediment interstitial liquid intrusions in super warm lakes in winter precedes the anaerobic zone formation.

3.2. Radiocesium dynamics perculiarity in super warm coloured water of lake

Seasonal courses of vertical profiles of standard water parameters in Lake Lydekinis show that the lake is meromictic and belongs to a super warm type. It consist of two different parts, which do not intermix: the upper monomictic part of some 300 cm thickness becomes completely aerobic in late autumn due to the gravitational mixing and an anaerobic stagnant bottom water layer of some 150 cm thickness.

It was determined that global fallout was only responsible for radioactive contamination of the deepest part of the lake. Radiocesium load of two sediment cores taken in the lake center was equal to ~370 and ~440 $Bq \cdot m^{-2}$, respectively, and corresponded to the contamination level of the Lithuanian territory before the Chernobyl accident. Vertical profiles of radiocesium activity concentrations in those cores were distinguished for its maximum activities in the sediment surface layer and limited radiocesium migration to deeper sediments. The radiocesium contamination of the sediments in the upper lake part is additionally effected by Chernobyl fallouts and radiocesium load show a significant (~ 3−4 fold) increase as compared to that measured in the deepest area of the lake. Theses sediments are characteristic of the elevated bioturbation and of the enhanced radiocesium migration.

Measurement data show that radiocesium activity concentrations in the water column from Lake Lydekinis increase with depth (Fig. 3.2) and follow the same tendency of the dissolved solid concentration in water samples.

Data in Fig. 3.2 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. 3.2). 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).

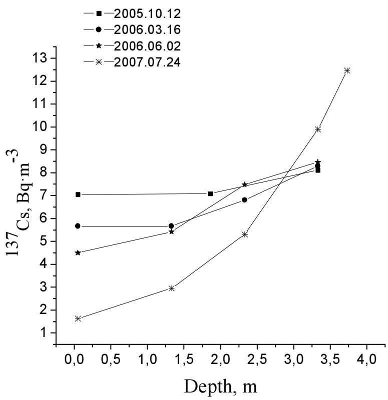

Fig. 3.2. Radiocesium concentration in lake water

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. Considering a permanent thermal stratification of the surface waters and bad mixing conditions, the main cause for that effect may be a large input of the melting water after a rather snowy 2006/2007 winter.

Measurement data show that radiocesium water-soluble concentrations in the lake are the highest among the other neighboring water bodies. It implies that radioactive fallouts may induce the formation of the extreme radiological situations in limited layers of the water column. It means that evaluating radiological consequences of radioactive accidents meromictic lakes must be treated as critical radioecological objects.

3.3. Radiocesium behavior in Lake Tapeliai

Data of the measurements of the 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-term period in spring (in April) at water temperatures near 4 °C and for a long-term period in autumn (in October or November) due to cooling processes inducing intense gravitational mixing.

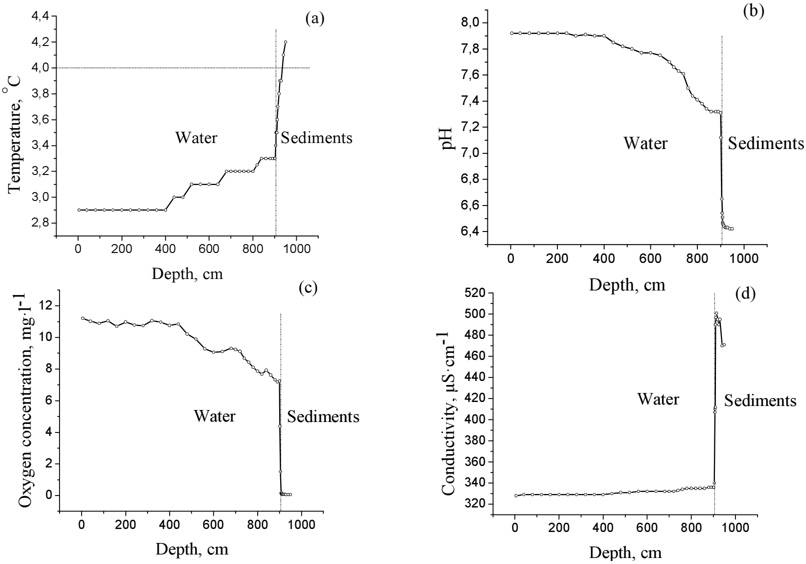

Fig. 3.3 (a, b, c, d). A layered structure of the water column measured on 7 December 2008: vertical profiles of temperature (a), pH (b), oxygen concentrations (c) and conductivity (d); depth of the bottom – ~905 cm

A classification of lakes based on the thermal regime of near-bottom water in winter must be related to thermodynamic peculiarities of the possible heat redistribution between sediments of the deep bottom area of the lake and the water column during that period. Measurements carried out in Lake Tapeliai show that depending on the amount of heat accumulated in sediments of the deep bottom areas of the lake until the beginning of winter, the lake may be assigned to the super-warm or to the moderately-warm type.

A shift of the 4 °C isotherm beneath the sediment surface in winter by some tens of centimeters measured on 7 December 2008 may imply the consequences the convective motions induced by buoyancy forces in surface sediments. A layered structure of the water column clearly seen in the vertical profiles of temperature, pH, oxygen concentration and conductivity (Fig. 3.3 (a–d)) proves the effects of buoyancy forces.

A course of water-soluble radiocesium activity concentrations in surface waters of Lake Tapeliai for the period of 2000−2010 is presented in Fig. 3.4. The first eight points represent data of the respective concentration measurements in water samples taken in the outflowing brook. The latter ones – in surface water samples taken in the center of the deepest lake part. It is easy to see that the mean concentration of the water-soluble radiocesium activity in measured samples for the period of 2009−2010 amounts to only one-third of that in the outflowing brook in 2000.

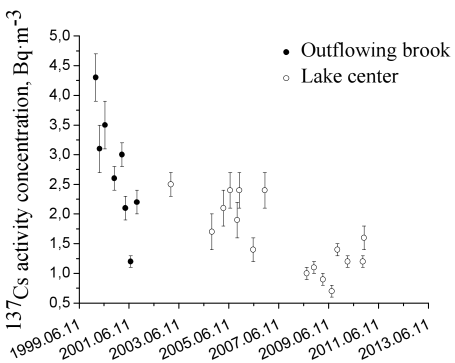

Fig. 3.4. A course of water-soluble radiocesium activity concentrations in lake (○) and outflowing brook (●) water in 2000−2010

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, elevated water-soluble radiocesium activity concentrations in the range 4,0–6,4 Bq·m^{-3} were always measured in brook water during the period of 2006−2010. 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 latest core in November 2009 were plotted as a function of the bottom depth in Fig. 3.5. The results were subdivided in four groups according to their sampling areas: the northern, northwestern and southern terraces, and the deepest lake zone. Radiocesium load is a maximum in a core taken on the northwestern terrace (~1520 Bq·m^{-2}).

Radiocesium inventories in cores taken on the southern terrace are distributed in a wide range 770−1470 Bq·m^{-2}, although, the bottom depth interval is rather narrow – ~ 4,2−5,4 m. Such distribution may be explained due to the influence of bottom feeding sources and of their related near-bottom currents at the southern terrace.

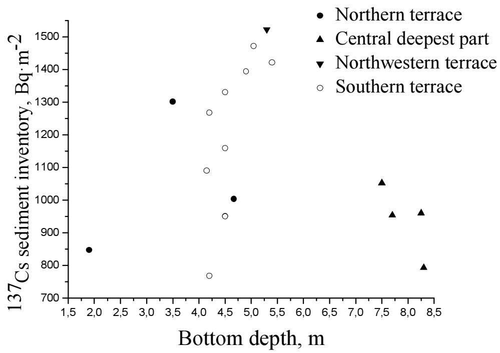

Fig. 3.5. Distribution of radiocesium inventories (Bq·m^{-2}) in the sediment cores with the bottom depth

A radiocesium inventory in the core taken in the shallow area of the northern terrace (~1,9 m depth) is rather low (~ 850 Bq·m^{-2}). This area is in the proximity of an outflowing brook and is open to the southern winds. Resuspension of fine sediments due to wind stirring of the shallow water increases suspended matter concentrations in the outflowing brook and decreases the respective radiocesium inventory in sediments. Radiocesium inventories of the cores from the deepest central zone of the lake are also comparatively low (below ~1050 Bq·m^{-2}) and do not show any "focusing effect" with depth.

4. Modeling of radionuclides migration in water and bottom sediments

In chapter 4, the transformations in the course of time of the radiocesium vertical profiles measured in two cores taken in the meromictic super warm lake and in a water body with the intermittent thermal regime, respectively, were depicted using a solution of the diffusion equation - the expression (4.1):

$$C(x,\ t)=\frac{C_0}{2\sqrt{\pi Dt}}\exp\left(-x^2/4Dt\right) \qquad (4.1)$$

Where C (x, t) − radionuclide activity in sediments at a depth x, C_0 − radionuclide activity at the sediments surface, D − an effective diffusion coefficient, t − time.

5. Development of measurements methodology of hydrophysical parameters of water

Automatic equipment (a complex of echo-sounder, of a ProfiLine Multi 197i portable device and of a laptop computer) is suggested in carrying out the measurements of vertical profiles of standard water parameters (vertical structure of the water body). The equipment is embedded at an inflatable boat.

General conclusions

1. A thermodynamic mechanism – buoyancy of the sediment interstitial liquids – is responsible for the near-bottom water enrichment in radiocesium in super warm 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 super warm Lake Juodis measured on 12 February 2009. Under aerobic conditions, these concentrations increase from 1,5 till 2,1 $Bq \cdot m^{-3}$ according to the temperature rise up to 4,6 $^{\circ}C$ in the 2,45-2,85 m depth interval.
2. Buoyancy of the sediment interstitial liquids induces processes of the surface sediment mixing and is responsible for the deepening of the beginning of the temperature gradient in sediments in autumn.
3. A thermal regime of the near-bottom water in Lake Tapeliai in winter depends on the amount of heat accumulated in sediments of its deepest bottom areas during warm seasons. This amount and temperatures of surface sediments of the deep bottom areas of the lake reach their maximum in autumn and their values depend on the particular meteorological situation. For this reason in some years, according to temperatures of the near-bottom water in the lake in winter, the lake may be assigned to the super warm or to the moderately warm type. A thermal classification of lakes suggested by K.Kilkus must be improved involving lakes of the intermittent thermal regime.
4. Maximum radiocesium activity concentrations measured in the sediments in Lake Tapeliai varied in the range of 110-190 $Bq \cdot kg^{-1}$ and of 4,6-8,7 $Bq \cdot l^{-1}$, respectively. Radiocesium activity concentrations in sediments reached their maximum values in the 6-19.5 cm sediment depth interval.
5. Maximum radiocesium activity concentrations measured in the sediments belonging to the bottom stagnant water layer of meromictic Lake Lydekinis varied in the range of 130−170 $Bq \cdot kg^{-1}$ and of 5,2−6,3 $Bq \cdot l^{-1}$, respectively. These concentrations in the sediments belonging to the upper water layer varied in the range of 160-310 $Bq \cdot kg^{-1}$ and of 6,7−12,9 $Bq \cdot l^{-1}$, respectively. As a rule, radiocesium activity concentrations in the bottom

stagnant water sediments reached their maximum values in the surface layer of 9 cm thickness. These concentrations in the sediments of the upper water layer peaked in the 8-20 cm sediment depth interval.

6. Bottom feeding sources in Lake Tapeliai substantially reduce the radiocesium load in the respective sediments as well as the sedimentation rate.
7. Radiocesium activity concentrations in water of the closed humic meromictic lake are about 7-fold larger than those of the neighboring ones. For this reason, closed meromictic lakes must be treated as the radiological critical objects.
8. Sedimentation in the permanently stagnant water layer of Lake Lydekinis is mainly due to flock, which is formed in the bottom zone of the upper water layer during processes of the gravitation mixing in autumn.

Recommendations

1. Estimating the super warm Lithuanian lakes as a potentially secondary source of radioactive pollution for water systems, it is suggested the 4 °C isotherm depth in winter to consider as one of the most important parameter. It allows evaluating an amount of lake water with elevated mineralization enhanced by buoyancy of the sediment interstitial fluids.
2. Meromictic water bodies must be revealed among the other super warm lakes as critical radioecological objects.
3. In carrying out engineering works in lakes, a comprehensive study must be performed to prevent them to turn into the meromictic water bodies. The possible risk must be evaluated.

List of published works on the topic of the dissertation
In the reviews scientific journals

Moisejenkova, A., Tarasiuk, N., Koviazina, E., Maceika, E., and Girgždys, A. 2012. 137Cs in lake Tapeliai (Lithuania), *Lithuanian Journal of Physics* 52(3): 238–252. ISSN 1648-8504. (ISI Web of Science)

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Moisejenkova, A.; Girgždys, A.; Tarasiuk, N. 2011b. Termodinaminių procesų įtaka radiocezio elgsenai Tapelių ežere [Peculiarities of the Thermal Regime of the Surface Sediments in Lake Tapeliai], iš *Mokslas – Lietuvos ateitis: Fizika ir fizinė kompiuterija* [elektroninis išteklius] [*Science – Future of Lithuania: Physics and physical computing* [CD]] 3(6), 95–100. Vilnius: Technika. ISSN 2029-2341. (Index Copernicus)

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RADIOCEZIO IR HIDROFIZINIŲ PARAMETRŲ DINAMIKOS TYRIMAI LIETUVOS EŽERUOSE

Problemos formulavimas

Viena svarbiausių medžiagų Žemėje yra vanduo. Žinoma, kad jis paplitęs atmosferoje bei sudaro paviršinę ir požeminę Žemės hidrosferą.

Nuo pirmųjų radioaktyviųjų atliekų susidarymo momento pradėta nagrinėti radionuklidų migraciją požeminiuose vandenyse. Visi gamtinio vandens objektai yra savotiškai unikalūs, tačiau didžiausią dėmesį reikėtų skirti ežerams. Ežerai yra sudedamoji gamtos išteklių dalis, kuria žmonės linkę naudotis.

Kilkus (2000) ežerų terminėje klasifikacijoje išskiria keturis ežerų tipus pagal jų priedugninio vandens temperatūrą (T) žiemos laikotarpiu: 1) ypač šilti ežerai, kurių T > 4 ºC; 2) šilti ežerai, T = 4 ºC; 3) vidutiniškai šilti ežerai, 3 < T < 4 ºC; 4) šalti ežerai, T < 3 ºC.

Ypač šiltų ežerų atsiradimas, kai priedugninio vandens temperatūros žiemos laikotarpiu viršija 4 ºC temperatūrą, atitinkančią maksimalius vandens tankius gėlavandenėje sistemoje, kelia didžiausią mokslinį susidomėjimą, nes atsiranda daug klausimų. Visų pirma, termohalininis stabilumas priedugninio vandens lygyje reikalauja, kad, esant aukštesnėms nei 4 ºC temperatūroms, priedugninis vanduo ypač šiltuose ežeruose turi būti papildomai mineralizuotas, kad jo tankis būtų didesnis nei aukščiau esančių termiškai stratifikuotų vandens sluoksnių. Antra, neaišku, kas sukelia padidintą vandens mineralizaciją. Be to, ypač šilti ežerai, esant padidintai priedugninio vandens mineralizacijai, turėtų veikti kaip galingi ištekančių upelių antrinio užterštumo šaltiniai.

Tyrimų objektas

Radiocezio dinamikos ir hidrofizinių vandens parametrų sąryšis ypač šiltuose Lietuvos ežeruose.

Darbo tikslas

Įvertinti radiocezio dinamikos ypatumus ypač šiltų ežerų vandenyje, dugno nuosėdose ir vanduo – dugno nuosėdos sistemos sandūroje (nustatant galimą radiocezio patekimo mechanizmą į priedugninį vandenį), juos siejant su eksperimentiškai gautais standartinių vandens parametrų sezoniniais pokyčiais. Įvertinti radiocezio dinamikos ypatumus nežinomo terminio režimo Tapelių ežere.

Darbo uždaviniai

1. Atlikti kompleksinę ypač šilto dimiktinio ežero vertikaliosios struktūros sezoninių pokyčių bei radiocezio aktyvumo koncentracijų vertikaliųjų profilių vandenyje analizę ir nustatyti priedugninio vandens praturtinimo radioceziu mechanizmą rudens ir žiemos laikotarpiais.
2. Nustatyti vidutinio gylio dimiktinio ežero terminį režimą ir jo atitikimą ežerų terminei klasifikacijai.

3. Įvertinti nežinomo terminio režimo ežero užtaršos radioceziu ypatumus ir jų sąryšį su ežero terminės struktūros pokyčiais.
4. Ištirti radiocezio elgsenos ypatumus ypač šiltame spalvoto vandens ežere.
5. Patobulinti eksperimentinę vandens hidrofizinių parametrų nustatymo ir vandens ir dugno nuosėdų mėginių paėmimo metodikas.

Tyrimų metodika

Darbe taikomi jonų mainų metodika hidrofizinių vandens parametrų nustatymui ir gama spektrometrijos metodas radiocezio pasiskirstymo dugno nuosėdose ir vandenyje nustatymui.

Darbo mokslinis naujumas

Rengiant disertaciją buvo gauti šie mokslui nauji rezultatai: atlikti kompleksiniai radiocezio dinamikos ir hidrofizinių vandens parametrų sezoninių pokyčių ypač šiltuose ežeruose tyrimai. Nustatyta, kad ypač šiltų ežerų priedugninio vandens praturtinimo radioceziu mechanizmas žiemos laikotarpiu yra termodinaminio pobūdžio ir susijęs su dugno nuosėdų skysčių plūdrumo efektais. Tai reikšminga tuo, kad ypač šiltų ežerų savivalos nuo teršalų mechanizmai yra susiję su terminiu režimu ežere. Parodyta, kad šis mechanizmas pradeda veikti esant aerobinėms sąlygoms ir tampa vieną iš priežasčių priedugninės anaerobinės zonos formavimosi pradžios. Nustatyta, kad ežero dugniniai šaltiniai mažina sedimentacijos greitį ir atitinkamai mažina šios zonos dugno nuosėdų užterštumą radioceziu.

Darbo rezultatų praktinė reikšmė

Atskleistas ypač šiltų ežerų dugno nuosėdų savivalos nuo radiocezio termodinaminio pobūdžio mechanizmas leidžia giliau suprasti šių ežerų egzistavimo fenomeną, susietą su galingomis aukštai mineralizuoto vandens anaerobinėmis zonomis. Šių zonų formavimosi pasėkoje, ypač šilti ežerai, laikui bėgant labai greit po užterštų atmosferinių iškritų, tampa galingais radionuklidų ir pramoninių teršalų šaltiniais ištekantiems upeliams. Tyrimai parodė, kad ypač šiltuose uždaruose meromiktinio tipo ežeruose apatiniame stagnaciniame vandens sluoksnyje formuojasi dešimteriopai didesnės teršalų koncentracijos negu dimiktinių ežerų vandenyse. Todėl šio tipo ežerai turi būti laikomi padidintos ekologinės rizikos objektais.

Darbo rezultatai pagilina sampratą apie ežerų savivalos nuo teršalų procesus ir leidžia prognozuoti radiocezio taršos remediacijos procesus ypač šiltuose ežeruose.

Ginamieji teiginiai

1. Ypač šiltų ežerų priedugninio vandens praturtinimo radioceziu žiemą mechanizmas yra termodinaminio pobūdžio ir yra susijęs su dugno nuosėdų poringo skysčio plūdrumo procesais. Todėl uždari meromiktiniai ežerai branduolinių objektų avarijos metu turi būti laikomi kritiniais radioekologiniais objektais.
2. Termodinaminiai šilumos apykaitos dugno nuosėdų poringo skysčio praturtinto radioceziu plūdrumo procesai prasideda dar esant aerobinėms sąlygoms ir yra viena iš priežasčių priedugninio vandens anaerobinės zonos formavimuisi. Jie lemia paviršinių dugno nuosėdų maišymosi procesą ir temperatūrinio gradiento pradžios nuosmukį gilyn į dugno nuosėdas.
3. Esama ežerų terminė klasifikacija pagal priedugninę temperatūrą žiemą turi būti patobulinta, nes yra ežerų, kurie keičia savo terminę būklę priklausomai nuo šilumos kiekio, sukaupto giliausios ežero dalies dugno nuosėdose.

Darbo apimtis. Disertaciją sudaro įvadas, 5 skyriai, bendrosios išvados ir rekomendacijos, literatūros sąrašas, autoriaus mokslinių publikacijų disertacijos tema sąrašas. Disertacijos apimtis – 128 puslapiai, 27 formulės, 74 paveikslai ir 3 lentelės.

Pirmasis skyrius skirtas mokslinės literatūros analizei. Jame pateikti ežerų terminės struktūros pokyčiai žiemos laikotarpiu. Aprašytas radiocezio patekimas ir pasiskirstymas ežerų ekosistemoje, taip pat jo migracija vandens telkinio dugno nuosėdose. Pateikiami radiocezio pasiskirstymo Lietuvos ežeruose tyrimai. Skyriaus pabaigoje formuluojamos išvados ir tikslinami disertacijos uždaviniai. Antrajame skyriuje pateikti vandens ir dugno nuosėdų bandinių paėmimo metodika ir radiocezio aktyvumo nustatymas gama spektrometrijos metodu. Aprašomi tiriamieji ežerai. Trečiajame skyriuje pateikta radiocezio dinamikos ryšio su hidrofizinių vandens parametrų sezoniniais kitimais analizė. Ketvirtame skyriuje atliktas radionuklidų migracijos modeliavimas. Pateikti rezultatai ir jų analizė. Penktame skyriuje aprašomas matavimo metodikos tobulinimas.

Bendrosios išvados

1. Termodinaminis šilumos apykaitos dugno nuosėdų poringo skysčio plūdrumas įtakoja žiemą vykstančius ypač šiltų ežerų priedugninio vandens praturtinimo radioceziu procesus. Šis procesas vyksta dar esant

aerobinėms sąlygoms rudenį ir yra priedugninio vandens anaerobinės zonos formavimosi pirmtaku žiemą.

2. Termodinaminis dugno nuosėdų skysčio plūdrumas lemia paviršinių dugno nuosėdų maišymosi procesą ir temperatūrinio gradiento pradžios smukimą gilyn į dugno nuosėdas.
3. Ežero priedugninio vandens terminis režimas priklauso nuo šilumos kiekio, sukaupto per šiltąjį sezoną giliausios ežero dalies paviršinėse dugno nuosėdose. Giliausios ežero dalies dugno nuosėdų paviršiaus aukščiausios temperatūros pasiekiamos rudenį gravitacinio maišymosi metu ir priklauso nuo konkrečios meteorologinės situacijos. Dėl šios priežasties kai kuriais metais jo priedugninio vandens temperatūra žiemą gali būti aukštesnė arba žemesnė nei 4 ºC.
4. ^{137}Cs tūrinių aktyvumų vertikalus profilis išmatuotas ypač šiltame sekliame ežere esant aerobinėms sąlygoms, rodo kilimą nuo 1,5 iki 2,1 Bq·m^{-3}, kai vandens temperatūra kinta nuo 4,6 ºC 2,45−2,85 m gylio intervale.
5. Radiocezio savitojo ir tūrinio aktyvumų didžiausios vertės, išmatuotos tirtuose ežeruose, kito atitinkamai 110−190 Bq·kg^{-1} ir 4,6−8,7 Bq·l^{-1} intervaluose. Dugno nuosėdų sluoksnis su didžiausiomis radiocezio koncentracijomis išsidėsto 6−19,5 cm dugno nuosėdų gylyje.
6. Ežero dugniniai šaltiniai pastebimai įtakoja radiocezio pasiskirstymą dugno nuosėdose, dvigubai ir daugiau mažinant savo zonose dugno nuosėdų užterštumą ir sedimentacijos greitį.
7. Ypač šilto uždaro humusinio meromiktinio ežero tūrinis radiocezio aktyvumas yra apie 7 kartus didesnis, palyginus su kitais ypač šiltais ežerais. Šie meromiktiniai ežerai gali būti vertinami kaip padidintos radiologinės rizikos objektai.
8. Ypač šilto uždaro humusinio meromiktinio ežero giliausių vietų sedimentacija susieta su dribsnių, susidariusių dėl šių vandenų įtraukimo į viršutinio aerobinio vandens sluoksnio gravitacinio maišymosi procesą, nusėdimu. Vidutiniame ežero gylyje (2–4 m) sedimentacija susieta su pelkinės zonos apatinės dalies dumblių nykimu.

Rekomendacijos

1. Vertinant ypač šiltus Lietuvos ežerus kaip vandens sistemų potencialius antrinio užterštumo šaltinius, rekomenduojama 4 ºC izotermės gylį ežere žiemą laikyti vienu svarbiausiu parametru. Šis parametras leidžia įvertinti priedugninio vandens tūrį, kurio mineralizacija yra padidinta dėl dugno nuosėdų poringo skysčio plūdrumo.

2. Vertinant visus Lietuvos ežerus, reikia nustatyti tarp ypač šiltų ežerų meromiktinius vandens telkinius, kaip padidintos radiologinės rizikos objektus.
3. Atliekant inžinerinius veiksmus su hidrologiniais objektais, reikalingi išsamūs hidrofizinių vandens parametrų ir teršalų sklaidos tyrimai, nustatant riziką hidrologiniams objektams tapti meromiktiniais vandens telkiniais.

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RESEARCH OF DYNAMICS OF RADIOCESIUM AND HYDROPHYSICAL PARAMETERS IN THE LITHUANIAN LAKES

Summary of Doctoral Dissertation
Technological Sciences, Environmental Engineering (04T)

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RADIOCEZIO IR HIDROFIZINIŲ PARAMETRŲ DINAMIKOS TYRIMAI LIETUVOS EŽERUOSE

Daktaro disertacijos santrauka
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