

## Environmental changes induced by human activities in the Northern Curonian Lagoon (Eastern Baltic): diatoms and stable isotope data

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**Abstract.** A sediment core collected from the northwestern part of the Curonian Lagoon, which was deposited approximately during 1800–2002, was analysed for several proxy records. Changes in diatom assemblages and carbon, nitrogen and oxygen stable isotopes ( $\delta^{13}\text{C}$ ,  $\delta^{15}\text{N}$  and  $\delta^{18}\text{O}$ ) revealed two periods, which are characterized by differences in the sedimentation rate, sediment type and trophic state of the northern part of the Curonian Lagoon. Low  $\delta^{15}\text{N}$  values in organics and prevailing fresh-brackish benthic diatoms indicate low enrichment in the shallow, freshwater lagoon during the period 1800–1955. The eutrophic conditions in this shallow lagoon are reflected by a high abundance of planktonic diatoms common in nutrient-rich basins and increased  $\delta^{15}\text{N}$  values in organics of the sediments since 1955. Starting approximately in the 1960s, decreased freshwater run-off and increased brackish-water inflow into the lagoon were observed. These changes were likely caused by the construction of the hydropower station (and a reservoir) near the Nemunas River and the artificial deepening of the Klaipėda Strait during 1960–1962 and later, also by the rising sea level in the SE Baltic. The changed river run-off and the artificially deepened strait significantly influenced the fresh-brackish water circulation and environmental conditions in the northern part of the Curonian Lagoon in the last decades.

**Key words:** diatoms,  $\delta^{13}\text{C}$ ,  $\delta^{15}\text{N}$ ,  $\delta^{18}\text{O}$ , eutrophication, Curonian Lagoon.

### INTRODUCTION

Environmental conditions, various natural forcing factors and human economic activities affecting the vulnerable European coasts and particularly the semi-enclosed coastal systems of the Baltic Sea have been discussed in recent studies (Andrén et al. 1999; Struck et al. 2000; Weckström 2006; Tuovinen et al. 2010; Voss et al. 2011). A comprehensive overview of the European lagoons and problems related to their ecological conditions and assessment is presented in the paper by Newton et al. (2014). The Baltic Sea is a semi-enclosed inland sea surrounded by nine countries and their economic activities greatly influence the ecological balance of the sea, especially transitional coastal areas. The Curonian Lagoon is one of these semi-enclosed coastal lagoons in the Baltic Sea, which are sensitive to environmental changes. Various methods, including the methods we have chosen, namely

the analysis of diatom assemblages and stable isotope analysis, can be used to assess the recent and past environmental changes of coastal lagoons and embayments.

Diatoms are a highly abundant group of phytoplankton algae in freshwater and marine environments, deep-water and littoral zone of the basin (Round et al. 1990). Their assemblage composition strongly depends on their environment (e.g. depth, salinity, pH, trophic status) and each diatom species has its optimum and tolerance limits for different environmental variables (Van Dam et al. 1994). Diatom valves generally preserve well in the sediment layer because of their resistant silicon dioxide valves. Changes in coastal diatom species assemblages from sediment sequences have been successfully used to study, e.g., the eutrophication and changes in turbidity, salinity and freshwater input, and the possible reasons for the observed palaeoenvironmental changes (Battarbee 1986; Cooper et al. 2010).

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Stable isotope ratios of light isotopes such as carbon ( $\delta^{13}\text{C}$ ), oxygen ( $\delta^{18}\text{O}$ ) and nitrogen ( $\delta^{15}\text{N}$ ) are frequently used to investigate environmental conditions and changes at present and in the past. The use of stable isotope ratios is an indirect method elucidating changes in the hydrological regime as it is commonly related to changes in source materials (Voss et al. 2000; Lesutienė et al. 2014). The method has been widely applied to both marine (Stein 1990; Lujanienė et al. 2015) and lacustrine (Leng & Marshall 2004) sediments because stable carbon and oxygen isotope ratios of carbonates are often linked to palaeotemperature and palaeoenvironmental changes (Broecker 1982), whereas carbon and nitrogen isotope ratios in organics are related to the trophic status and organic matter sources in ecosystems (Rolff 2000; Voss et al. 2000; Lesutienė et al. 2014). Since eutrophication is a severe and wide-spread problem, it is important to understand the possible natural and anthropogenic sources of nutrient supply (Savage et al. 2010; Voss et al. 2011).

Shallow lagoons are complicated environments with both fresh- and brackish-water input and periodic resuspension of bottom sediments (Trobajo & Sullivan 2010). The Curonian Lagoon is a good example of such a complicated environment. The northern part of the lagoon is influenced by many natural processes: fresh- and brackish-water exchange via the narrow strait, resuspension of bottom sediments because of waves and currents, seasonal changes in freshwater inflow from rivers, frequent brackish-water intrusions during autumn–winter because of the prevailing western–northwestern wind and changes in the Baltic Sea level (Dubra 1978; Žaromskis 1996; Dailidienė et al. 2006; Dailidienė & Davulienė 2008; Jakimavičius & Kovalenkoviėnė 2010; Zemlys et al. 2013). The environment of this lagoon is also greatly influenced by human activities: rivers (the largest Deyma, Nemunas and Minija) drain urban areas, the sea port is located in the northern part of the lagoon and the strait was artificially deepened twice compared to its natural depth two centuries ago (Gailiušis et al. 2005; Jakimavičius & Kriaučiūnienė 2011; Zemlys et al. 2013; Suzdalev et al. 2015). All these natural and anthropogenic factors create specific, variable environmental conditions in the Curonian Lagoon, especially in its northern part.

Detailed studies of the Curonian Lagoon environmental conditions by applying analysis of stable isotopes and algae in many cases include only several seasons, years (e.g. Gasiūnaitė et al. 2005; Aleksandrov & Dmitrieva 2006; Karosienė & Paškauskas 2012; Lesutienė et al. 2014; Remeikaitė-Nikienė et al. 2016) or decades (Voss et al. 2000). The aim of this study is to describe environmental changes (natural and influenced by human activities) in the northern part of the Curonian Lagoon

since ca 1800, using diatom assemblage analysis together with stable isotope analysis on a dated sediment core.

## STUDY AREA

The Baltic Sea is one of the largest brackish seas in the world, situated between about 10–30° E and 54–66° N. The Baltic Sea consists of several basins of different depth that are connected by sills. The deepest basins are Eastern (depth 250 m) and Western Gotland (460 m); the depth of other basins is less (for example, Arkona – 45 m, Bornholm – 100 m, Bothnian Sea – 120 m). The Baltic Sea is connected to the North Sea via the narrow Danish Straits and the Kattegat. The salinity of the upper layer of the brackish water in the central Baltic Sea (Baltic Proper) is about 6–8 PSU and the salinity of the deep-water layer is about 10–14 PSU (Lass & Matthäus 2008).

The Curonian Lagoon is located in the central–eastern part of the Baltic Sea and is separated from the Baltic Proper by the narrow (width 0.4–3.8 km) sandy Curonian Spit (Fig. 1). The total area of the Curonian Lagoon is 1584 km<sup>2</sup> (Dubra 1978). The sediment core for this study was obtained from the northern part of the lagoon (Lithuanian territorial waters), which makes up 413 km<sup>2</sup> (Dubra 1978). The depth of the lagoon in this area is only 1–2 m in the eastern part and reaches 3–4 m along the Curonian Spit in the western part (Žaromskis 1996). The recent bottom sediments are composed of fine sand, which is brought mostly by the Nemunas River in the central and eastern parts of the lagoon, whereas the coarser (medium) sand accumulates mainly in the western, nearshore part as a result of aeolian activity. Sedimentation of fine silty mud can be observed in the southern part of Lithuanian territorial waters at a depth of 3–4.5 m (Trimonis et al. 2003). Because of a large freshwater inflow from rivers (the Nemunas and Minija) and irregular brackish-water intrusions from the sea, salinity varies from 0 to 7 PSU in the northern part of the lagoon (Gasiūnaitė et al. 2008).

The Curonian Lagoon is connected with the Baltic Sea via the narrow Klaipėda Strait. The width of the strait changes from 0.4 km at the port gate to 1.2 km in the southern part and the length is about 11 km. The strait is the only way for freshwater outflow and brackish-water intrusions. According to long-term (1960–2007) water balance calculations, the inflow of brackish water from the Baltic Sea to the Curonian Lagoon is 6.1 km<sup>3</sup>/year and the freshwater outflow from the Curonian Lagoon to the sea is 27.6 km<sup>3</sup>/year (Jakimavičius & Kovalenkoviėnė 2010). The average depth of the strait was 5–6 m until the 19th century and 7–8 m until the middle of the 20th century.

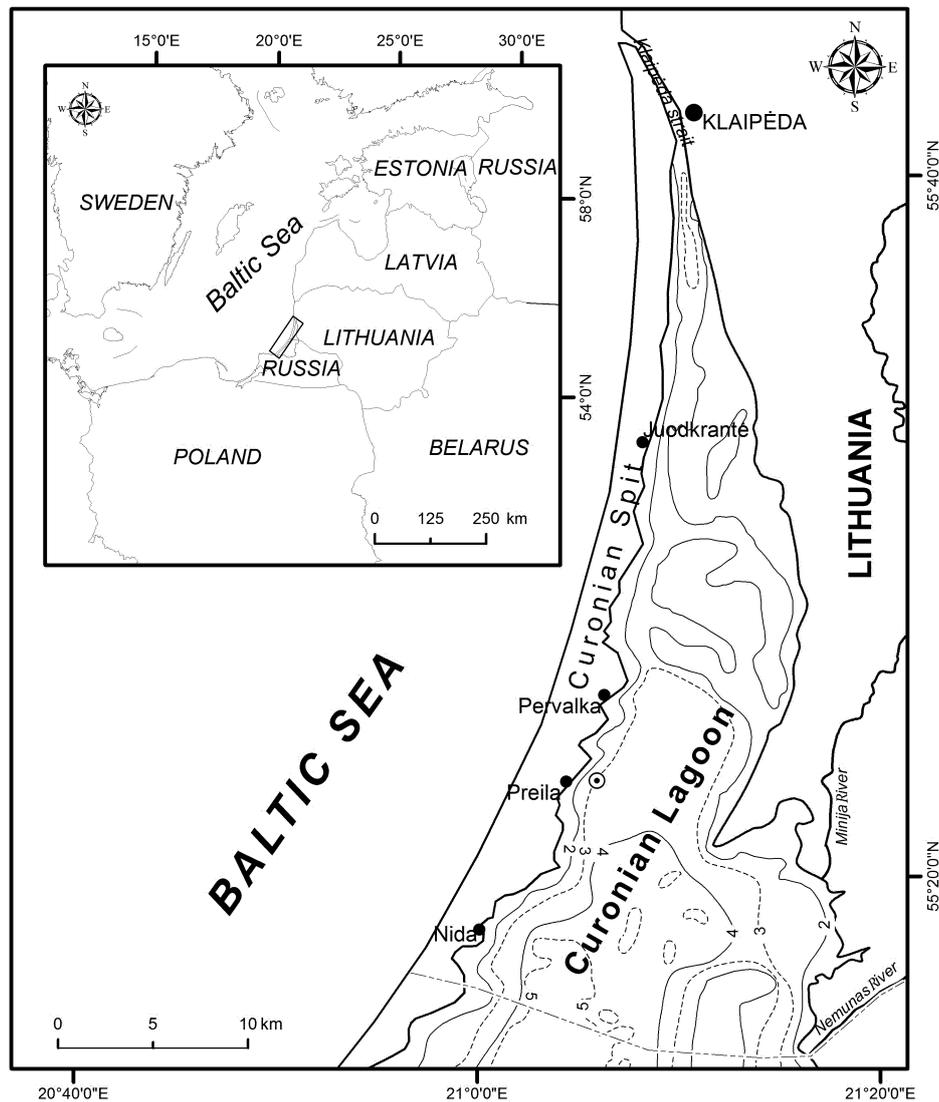


Fig. 1. Location of the investigated sediment core.

The strait was dredged to 11 m in 1960–1962 and later to 12 m in 1982–1983 and the water flow in the strait increased by 10–15% (Gailiusis et al. 2005; Jakimavičius & Kriauciūnienė 2011). The growth of the economic activity of the Port of Klaipėda required the reconstruction of the port entrance and the northern part of the strait reached a depth of 14 m until 2003.

## MATERIAL AND METHODS

### Sediments

The sediment core of 60 cm length was taken with the Niemistö gravity corer from a boat in the northwestern part of the Curonian Lagoon (N 55°22'12'' and

E 21°05'11'') in 2003. The measured water depth at the coring site is 3.0 m (Fig. 1). The northwestern part (along the Curonian Spit) of the lagoon is deeper and allows the accumulation of fine-grained sediments. In general, fine-grained sediments from the deeper parts of a coastal embayment/lagoon contain complete and undisturbed sediment sequences, necessary for detailed proxy analyses.

The sediment core was subdivided into 1 cm samples. In order to identify the content of organic matter, calcium carbonate and terrigenous input in the sediments, loss-on-ignition (LOI) was analysed (Bengtsson & Enell 1986). Firstly, the samples were slowly pre-dried at 40–50 °C, homogenized in the mortar and finally dried at 105 °C until the weight remained constant. The samples

were then combusted at 500 °C for 4 h to burn all organic matter. Finally, the samples were burned for 4 h at 950 °C to remove carbonates. Organic matter, carbonate and mineral proportions were determined. The water content and the dry bulk density of sediments were estimated by weighing standard volume samples dried at 105 °C.

### Analysis of $^{210}\text{Pb}$ and other radionuclides by gamma spectrometry

Dried samples were analysed for  $^{210}\text{Pb}$ ,  $^{214}\text{Pb}$  and  $^{137}\text{Cs}$  using a gamma-ray spectrometer with a HPGe GWL-series detector at the Nature Research Centre, Vilnius, as described by Gudelis et al. (2000). The detector characteristics are as follows: detector diameter – 54.7 mm, detector length – 67.8 mm, active well depth – 40 mm, well inside diameter – 15.5 mm, total active volume of the detector – 126 cm<sup>3</sup>, absorbing layers of high-purity aluminium – 0.5 mm, inactive germanium – 0.3 µm, resolution, FWHM, at 1.33 MeV ( $^{60}\text{Co}$ ) – 2.25 keV, and at 122 keV ( $^{57}\text{Co}$ ) – 1.2 keV. The samples were placed inside the well in small beakers of 3 mL volume for acquisition times up to 300 000 s in order to get good statistics of gamma spectra for low-activity samples.

The gamma transmissions used for activity calculations were 46.5 keV for  $^{210}\text{Pb}$ , 661.7 keV for  $^{137}\text{Cs}$  and 351.9 keV for  $^{214}\text{Pb}$  as a short-lived granddaughter of  $^{226}\text{Ra}$ . For the activity calculation and the quality assurance procedure the gamma spectra processing software GammaVision-32 was used. The gamma spectrometric system was calibrated for counting efficiency using commercially available multi-radionuclide standard sources and a reference solution for different densities and filing heights with the selected matrix as described in detail by Marčiulionienė et al. (2015). The detection limit for the counting time of 200 000 s was about 0.014 Bq for  $^{137}\text{Cs}$ , 0.065 Bq for  $^{210}\text{Pb}$  and 0.021 Bq for  $^{214}\text{Pb}$ , while measurement errors did not exceed 8%, 15% and 20% for  $^{137}\text{Cs}$ ,  $^{210}\text{Pb}$  and  $^{214}\text{Pb}$ , respectively. The normal precision of gamma spectrometric measurements of our laboratory was validated during the national comparison exercise of 2013, organized by the Lithuanian Metrology Inspectorate and Center for Physical Sciences and Technology, and the intercomparison of 2014 organized by STUK – Finnish Radiation and Nuclear Safety Authority.

### Diatom analysis

The laboratory preparation of sediment samples for diatom analysis followed Battarbee (1986). Diatom species were identified using a ‘Nikon Eclipse E200’ microscope (magnification  $\times 1000$ ). Identification was mainly based

on the following literature: Krammer & Lange-Bertalot (1986–1991), Snoeijs (1993), Snoeijs & Vilbaste (1994), Snoeijs & Potapova (1995), Snoeijs & Kasperovičienė (1996), Snoeijs & Balashova (1998). The number of counted valves is 500–1000 per slide. Some diatoms were identified only to genus level due to the poor condition of valves (broken) or due to the valve being positioned in girdle view. Quite a large number of valves from the Genus *Fragilaria* lie in girdle view on the diatom slides. Only three taxa of this genus were identified. *Fragilaria heidenii* was the most abundant (up to 75% of the total assemblage), while *Fragilaria brevistriata* Grunow and *Fragilaria construens* (Ehrenberg) Grunow comprise up to 2% of the total diatom assemblage. The valves of these species which appear in girdle view on the diatom slides were grouped together and named *Fragilaria* spp. in the diagram. The *Fragilaria* spp. is marked as ‘fresh-brackish’ because all three species are periphytic and characteristic of a ‘fresh-brackish’ environment. The species *Actinocyclus normanii* is attributed to the ‘brackish-fresh’ group according to the ecological classification in Van Dam et al. (1994). The most frequent and ecologically important diatom species are presented as percentages of the total diatom sum. The species are grouped according to their salinity requirement (Van Dam et al. 1994; Loseva 2000; Barinova et al. 2006, pp. 230–231): (1) brackish – salinity is 1.8–9.0‰, (2) brackish-fresh – salinity is 0.9–1.8‰, (3) fresh-brackish – salinity is less than 0.9‰ and (4) fresh – salinity is less than 0.2‰. Diatoms are also grouped according to their habitat into planktonic and periphytic (including benthic and epiphytic).

For the calculation and presentation of diatom data the programs TILIA and TILIA–GRAPH (Grimm 1992) were used. The diagram was subdivided into local diatom assemblage zones (LDAZ) using characteristic taxa and sum-of-squares cluster analysis (Grimm 1987).

### Stable isotope analysis

Before the  $\delta^{13}\text{C}$  analysis, the sediment samples were pretreated with acid (1N HCl) to remove carbonates. This was done in 13  $\times$  100 mm test tubes covered with vented closures in an ultrasonic bath at 70 °C. Samples (up to 0.5–1 g) were treated with aliquots of  $\sim 10$  ml of 1N HCl for 60 min. When no more bubbling was observed, the samples were dried using a dry bath under a hood (this can be sped up by flushing with filtered air or nitrogen) and oven-dried 12 h at 50 °C (Balesdent & Mariotti 1996).

After the pretreatment, all samples of organics (at least two per sediment sample) were weighed and wrapped in tin capsules after removing carbonates. The prepared samples were combusted with the elemental analyser

(FlashEA1112) connected to the isotope ratio mass spectrometer (Thermo Finnigan Delta Plus Advantage). Stable carbon and oxygen isotope ratios were measured in  $\text{CO}_2$ , while the stable nitrogen isotope ratio was measured in  $\text{N}_2$ . Stable isotope data are expressed as delta values according to the formula  $\delta X = [R_{\text{sample}}/R_{\text{standard}} - 1] \times 10^3$ , where the numerator is the isotope ratio of the sample ( $^{13}\text{C}/^{12}\text{C}$ ,  $^{18}\text{O}/^{16}\text{O}$  or  $^{15}\text{N}/^{14}\text{N}$ ) and the denominator is the isotope ratio of the standard ( $^{13}\text{C}/^{12}\text{C}$ ,  $^{18}\text{O}/^{16}\text{O}$  and  $^{15}\text{N}/^{14}\text{N}$ , respectively). The international standards (IAEA-600 and IAEA-N-1) from the International Atomic Energy Agency (Vienna) were used for the calibration of the reference gases ( $\text{CO}_2$  and  $\text{N}_2$ ). The detailed methodology of stable isotope analysis is described in Garbaras et al. (2008) and Ceburnis et al. (2011). Repeated analysis of homogeneous material gave a standard deviation of delta values less than 0.08‰ for carbon and 0.2‰ for nitrogen.

The measurements of carbonates for  $\delta^{13}\text{C}$  and  $\delta^{18}\text{O}$  values were performed with Gas Bench II (Thermo Fisher Scientific) connected to the isotope ratio mass spectrometer (Delta V Advantage). Stable isotope analysis was done by analysing carbon dioxide produced during the reaction of carbonate with 100%  $\text{H}_3\text{PO}_4$  at 70 °C for 2 h. The reproducibility was  $\pm 0.1$ ‰. All stable carbon and oxygen isotope values are reported relative to the V-PDB scale. The detailed methodology is given in Paul & Skrzypek (2007).

## RESULTS

### Sediment composition

The sediment core is composed of silty mud. According to visual inspection, the lower (60–40 cm) part of the sequence consists of coarse silty mud. The topmost (40–0 cm) part of the sediment column is composed of fine silty mud. The analysed sediment core has accumulated approximately during the last two centuries, from 1800 to 2002. The calculated linear sedimentation rate is approximately 0.3 cm/year for the whole core. However, the sedimentation rates of the two sections of coarse and fine silty mud are different:  $\sim 0.1$  cm/year and  $\sim 0.6$  cm/year, respectively.

The percentage content of clastic, carbonate and organic matter based on LOI is presented in Fig. 2. Two intervals can be distinguished according to varying components of the sedimentary matrix.

The lowest part of the core (58–40 cm) contains coarse silty mud with less than 26%  $\text{CaCO}_3$ . The amount of clastic matter is highest in the sequence and reaches 85% at a depth of 43 cm, 79–85% at a depth of 53–58 cm. The content of organic matter varies between 6% and

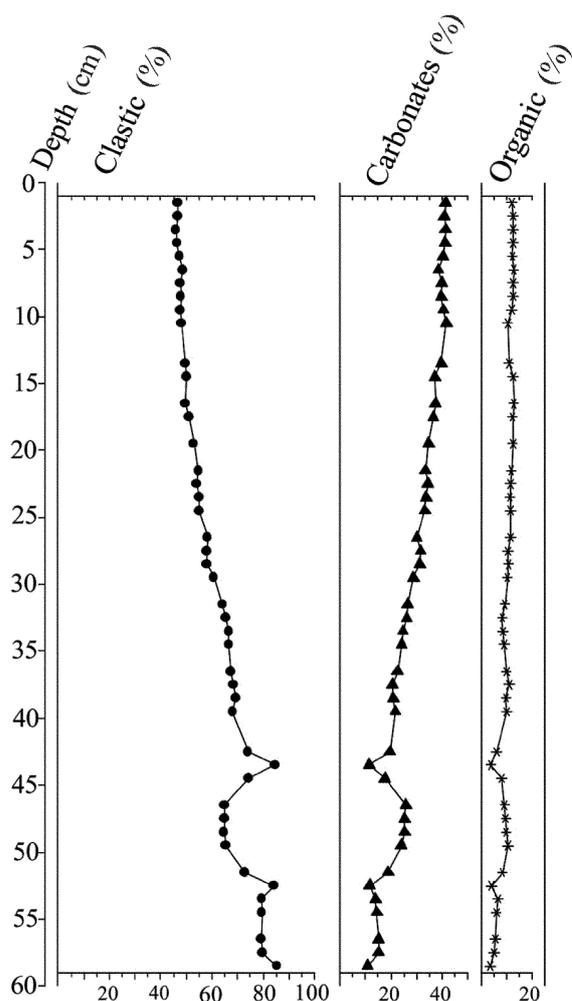


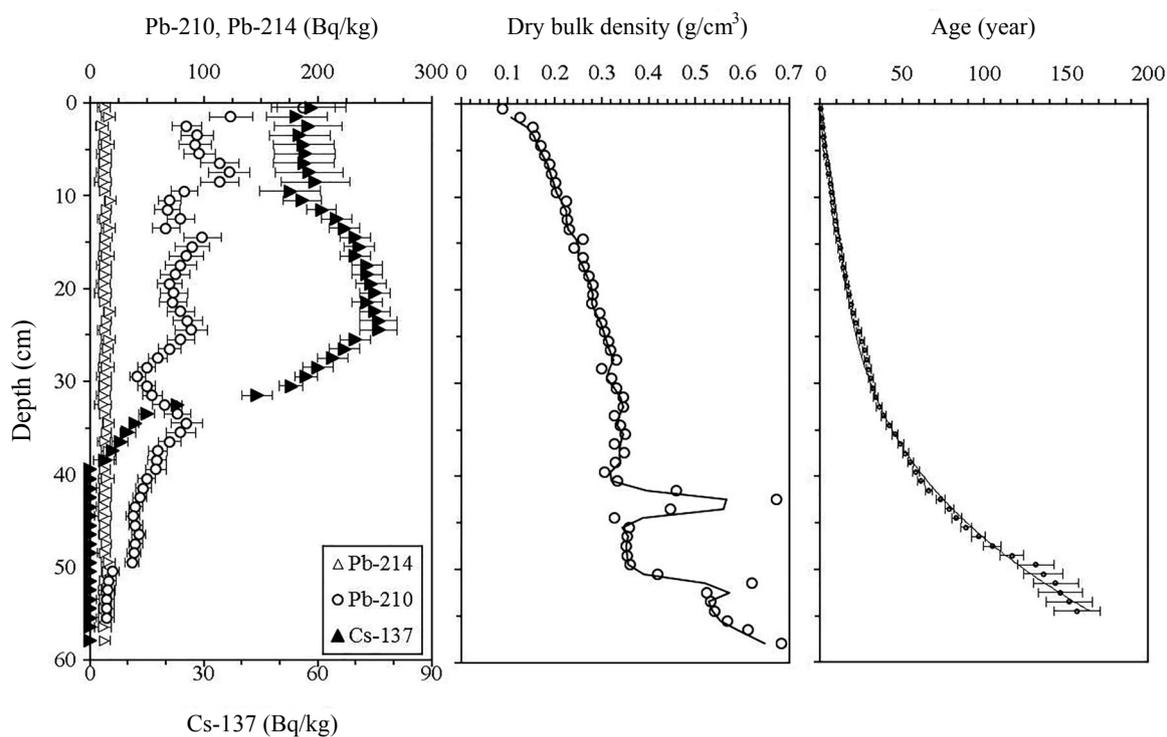
Fig. 2. The percentages of clastic, carbonate and organic matter according to LOI results in the sediment core.

11%. The percentage of clastic matter decreases from 85% to 65% at a depth of 46–49 cm. However, the curve of carbonate content rises from 11% to 26% and organic matter reaches 9–11% at the same interval.

Sediments of the upper part of the core (40–0 cm) become carbonaceous. The content of  $\text{CaCO}_3$  gradually increases from 26% to 40%. The content of clastic matter decreases from 74% to 47%, but the percentage of organic matter increases only slightly and varies between 8 and 13%. The topmost sediments (10–0.5 cm) can be characterized by almost constant values of clastic, carbonate and organic matter varying between 46% and 48%, 38% and 42%, and 12% and 13%, respectively.

### Radionuclide distribution and $^{210}\text{Pb}$ dating

The initial data on the activity concentration of  $^{210}\text{Pb}$  (total),  $^{214}\text{Pb}$  and  $^{137}\text{Cs}$  in the sediment core taken from the Curonian Lagoon are given in Fig. 3. The specific activity



**Fig. 3.** The activity concentration of  $^{210}\text{Pb}$ ,  $^{214}\text{Pb}$  and  $^{137}\text{Cs}$  versus depth (1.0 cm thick slices) in the sediment core, the sediment dry bulk density versus depth (with a moving average trend line) and the sediment age–depth model based on the constant rate of  $^{210}\text{Pb}$  supply (CRS) approach (the line represents a polynomial function with  $R^2 = 0.9958$ ).

of  $^{210}\text{Pb}$  in the core decreases from 180–190 Bq/kg at the top of the core to 15–25 Bq/kg at a depth of 51 cm, however, unevenly with lower (3, 12 and 30 cm depth) and higher (8, 15, 25 and 35 cm depth)  $^{210}\text{Pb}$  values observed. These changes can be dependent on sediment mixing due to wind activity, variable sorption capacity of particulate matter, variable sedimentation and consequently pronounced  $^{210}\text{Pb}$  dilution for intervals with a higher sedimentation rate and the  $^{210}\text{Pb}$  concentration effect for intervals with a lower sedimentation rate. At a depth of 51 cm the specific activity of  $^{210}\text{Pb}$  is within the limits of uncertainties close to the level of  $^{214}\text{Pb}$  in sediments, which corresponds to the supported  $^{210}\text{Pb}$  level. Deeper in the core, between 51 and 59 cm, the specific activity of  $^{210}\text{Pb}$  is constant around 15–25 Bq/kg.

Using the dry bulk density (Fig. 3) and unsupported  $^{210}\text{Pb}$  activity data, a constant rate of  $^{210}\text{Pb}$  supply (CRS) model (Appleby & Oldfield 1978; Binford 1990; Appleby 2001; Mažeika 2006; Piotrowska et al. 2010) was applied to calculate the sediment mean mass accumulation rate and to construct the sediment chronology for the past (up to) 150–200 years of sedimentation. The unsupported  $^{210}\text{Pb}$  activity was calculated by subtracting the supported activity, assuming to be equal to  $^{214}\text{Pb}$ , from the total

$^{210}\text{Pb}$  activity. The sediment age–depth model is presented in Fig. 3. The dating uncertainties derived from the  $^{210}\text{Pb}$  CRS approach were typically 1–4 years for the top part of the core; however, they reached 10–20 years, approaching the limits of the method, for the bottom part of the core.

The sediment mass accumulation rates for the past 185 years are in the range of  $0.25 \pm 0.06$  to  $3.8 \pm 0.1$  ( $\text{kg}/\text{m}^2$ )/year with a mean sediment mass accumulation rate of  $1.8 \pm 0.09$  ( $\text{kg}/\text{m}^2$ )/year for the whole core.

The  $^{137}\text{Cs}$  values are highest between ca 10 and 30 cm. No distinct peaks are observed, attributable to the nuclear weapon tests in the atmosphere in the early 1960s and the Chernobyl NPP accident in 1986 (Fig. 3).

For the 0–10 cm depth interval the  $^{137}\text{Cs}$  activity is close to 55 Bq/kg. Below the 10 cm depth and down to 25 cm the  $^{137}\text{Cs}$  activity in sediments is slightly increasing, reaching a maximum value ( $75 \pm 5$  Bq/kg) at a depth of 25 cm where a not very well defined  $^{137}\text{Cs}$  peak is determined. Below 25 cm the  $^{137}\text{Cs}$  activity is steeply decreasing and at 40 cm depth it reaches the level lower than minimum detectable activity. The profile of the  $^{137}\text{Cs}$  activity agrees relatively well with the  $^{210}\text{Pb}$  CRS

dates, despite the  $^{137}\text{Cs}$  from the Chernobyl accident apparently masking the weapons fallout record, so it was not possible to distinguish the 1963 depth due to  $^{137}\text{Cs}$  mobility in the sediments.

### Diatoms

A relatively small number of diatom species characterizes the analysed sediment core. Altogether 49 diatom species (25 genera) were identified during the analysis of 38 sediment samples. Diatoms were abundant both in coarse and fine silty mud. Based on cluster analysis and characteristic species, two local diatom assemblage zones (LDAZ) could be distinguished (Fig. 4).

LDAZ CL-1 encompasses the depths 58–35 cm. These sediments accumulated approximately during the period 1800–1960. The zone is characterized by a prevalence of fresh-brackish diatoms up to 99% (mainly *Aulacoseira granulata*, *Aulacoseira islandica*, *Fragilaria heidenii*, *Martyana martyi*, *Stephanodiscus rotula*, *Thalassiosira lacustris*). The most abundant species is periphytic *Fragilaria heidenii* and it reaches 30–75% of the total diatom sum. Marine-brackish diatoms make up only 2% of the total diatom sum (*Cyclotella striata* and *Cocconeis scutellum* are most common), as do brackish-fresh diatoms. At a depth of 38 cm this group starts to gradually increase and reaches 20% at the top of the zone (Fig. 4). The brackish-fresh diatoms are represented by low abundances of periphytic *Cocconeis pediculus*, *Diploneis domblittensis* and planktonic *Cyclotella meneghiniana* through most of the zone. The increase around 38 cm in the brackish-fresh diatoms group is mainly caused by a clear increase in the abundance of planktonic *Actinocyclus normanii*.

LDAZ CL-2 encompasses the depths 35–2 cm. These sediments were deposited approximately during 1960–2002. Brackish-fresh diatom species increase from 20% to 49% in the middle of the zone. *Actinocyclus normanii* dominates this ecological group and comprises as much as 49% of the total diatom assemblage in some samples. The brackish-fresh group decreases to 22–25% at the top of the zone. Marine-brackish diatoms make up to 1.5% of the assemblages and are mostly represented by marine planktonic *Coscinodiscus radiatus*. Diatoms of this group almost disappear at the top of the zone and make up to 0.5%. The abundance of the fresh-brackish diatom group decreases and makes up 50–85% of the total diatom sum. The amount of the species *Fragilaria heidenii* decreases and makes up 13–49% of the total diatom assemblages, while planktonic *Aulacoseira islandica* increases to 16–45%. The percentage of *Cavinula scutelloides*, *Martyana martyi* and *Stephanodiscus rotula* is low and remains stable throughout this zone.

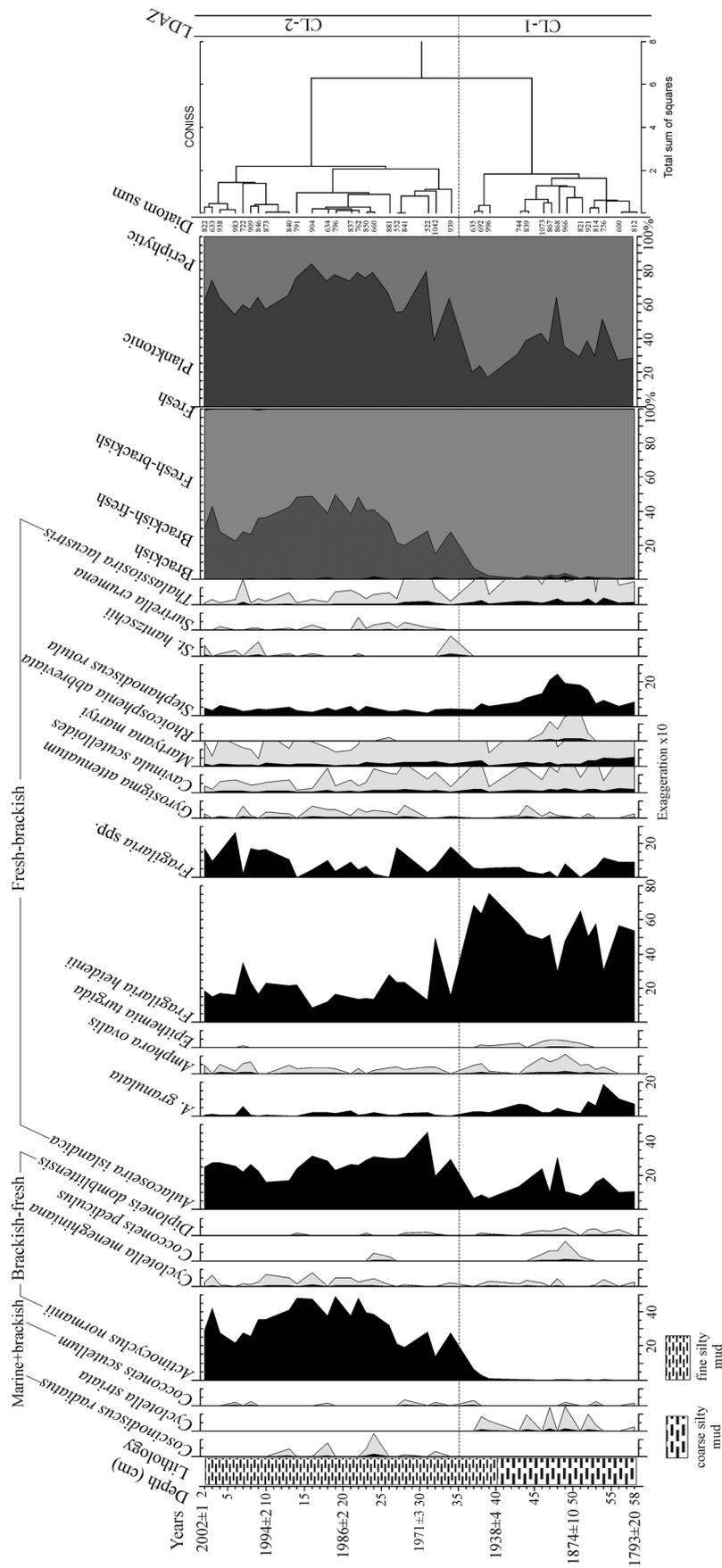
### Stable isotopes

Stable isotope data were tested for normality (Shapiro-Wilk normality test). A significance level of 0.05 was chosen. The values of  $\delta^{13}\text{C}$  in organics and total  $\delta^{15}\text{N}$  have a normal distribution with  $p$ -values of 0.81 and 0.09, respectively. Stable carbon and oxygen isotope values in carbonates do not show a normal distribution with  $p$ -values of 0.001 and 0.004. Stable isotope ratios were correlated independently in organics and in carbonates. The Pearson's correlation was applied to normally distributed data and the Spearman's correlation to not normally distributed data. The values of  $\delta^{13}\text{C}$  in organics and total  $\delta^{15}\text{N}$  show a low linear negative correlation, however, this correlation is not statistically significant ( $r = -0.28$ ,  $p = 0.06$ ), whereas a high statistically significant positive correlation ( $r = 0.77$ ,  $p < 0.001$ ) between  $\delta^{13}\text{C}$  and  $\delta^{18}\text{O}$  values in carbonates was determined.

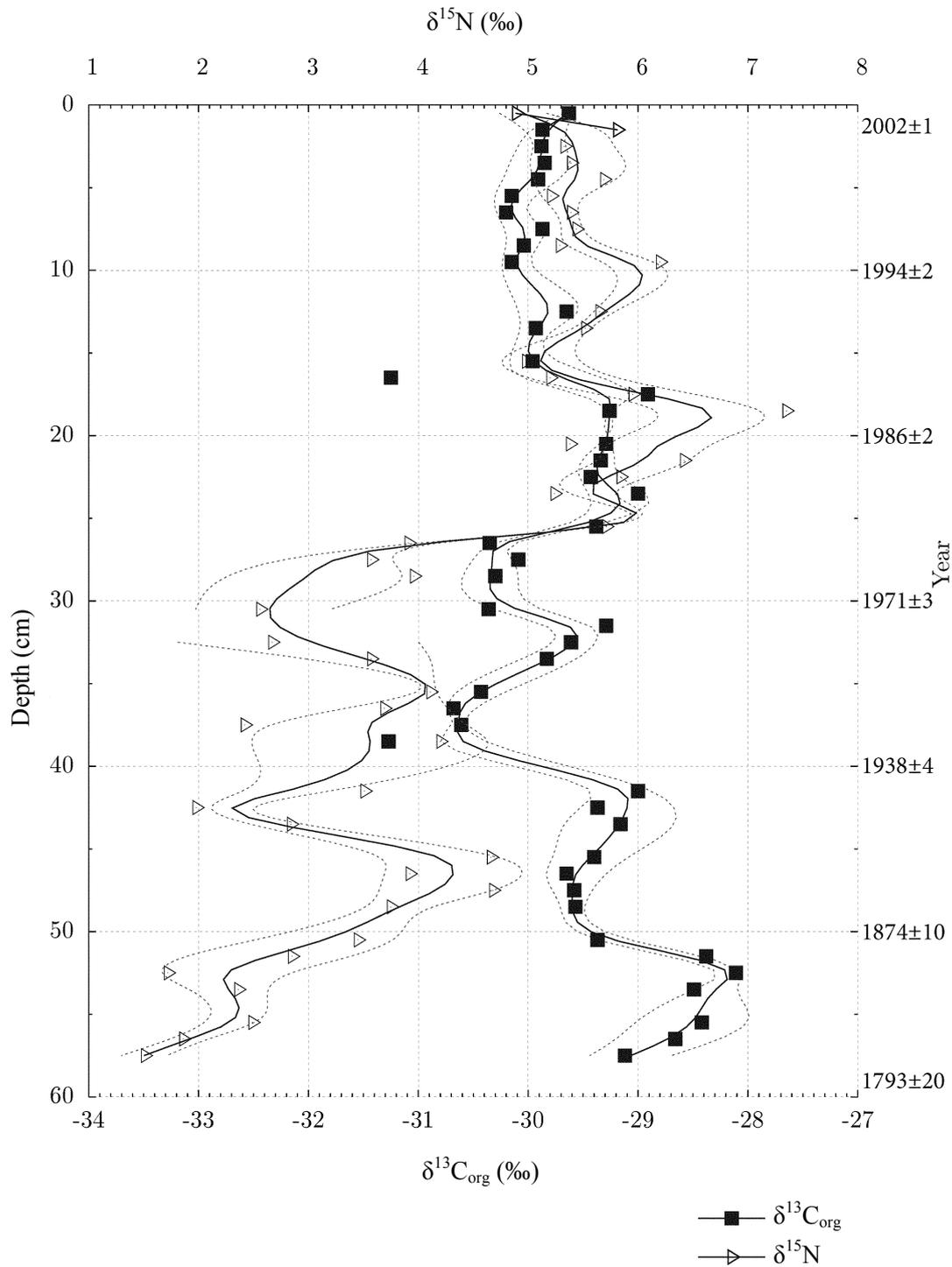
The values of  $\delta^{13}\text{C}$  in organics and total  $\delta^{15}\text{N}$  are presented in Fig. 5 and  $\delta^{13}\text{C}$  and  $\delta^{18}\text{O}$  values in carbonates in Fig. 6. The approximation of data points was done using a weighted cubic spline interpolation algorithm with weights  $\omega_i = 1/\sigma_i$  (here  $\sigma_i$  is standard deviation). The upper and lower curves represent the interpolation of the standard deviation (in the case of measurements in organics) from an average of a set of three measurements for each sample.

The  $\delta^{13}\text{C}$  values in organic fraction of the sediments vary from  $-31.3\text{‰}$  to  $-28.1\text{‰}$ , with the average value of  $-29.7\text{‰}$ ; the range is  $3.2\text{‰}$ . The curve of  $\delta^{15}\text{N}$  has the lowest value of  $1.5\text{‰}$  and the highest value of  $7.4\text{‰}$ ; the average value is  $4.3\text{‰}$  and the range is  $5.9\text{‰}$  (Fig. 5). The most  $^{13}\text{C}$ -enriched values are found at the bottom of the core, while upwards values decrease by about 1‰ and reach the lowest value at a depth of 30 cm. The most  $^{15}\text{N}$ -depleted values are at a depth of 58 cm but upwards the stable isotope values fluctuate and gradually increase to more positive approximately at a depth of 30 cm. The lower part of the core also has the higher standard deviations in both carbon and nitrogen values in comparison with the upper part of the core. Stable isotope ratios show smaller fluctuation upwards from a depth of 30 cm and the topmost part of the sequence (15–1 cm) is characterized by quite constant  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  values.

Carbon and oxygen stable isotope ratios in carbonates were analysed together as they showed a positive correlation coefficient. The  $\delta^{13}\text{C}$  values vary from  $-4.2\text{‰}$  to  $-2.2\text{‰}$ , with an average value of  $-3.3\text{‰}$ . The  $\delta^{18}\text{O}$  values fluctuate from  $-9.5\text{‰}$  to  $-6.8\text{‰}$ , with an average value of  $-8.3\text{‰}$  (Fig. 6). The stable isotope ratios show high fluctuations in the lowest part of the core (58–30 cm), while upwards from a depth of



**Fig. 4.** Age, sediment types, the most frequent and ecologically important taxa and diatom salinity, habitat groups in the sediment sequence. Exaggeration of the species percentage (grey colour) is  $\times 10$ .



**Fig. 5.** Variation in  $\delta^{13}\text{C}$  in organic fraction and total  $\delta^{15}\text{N}$  values in the sediment core.

30 cm both carbon and oxygen isotope values display a decreasing trend. Between the depths of 26 and 17 cm the decrease is less distinct and from 17 cm to the top of the sequence the isotope values remain

stable. The trends at the top are similar to the organics measurements, which also show little variance in the topmost part of the sediment sequence.

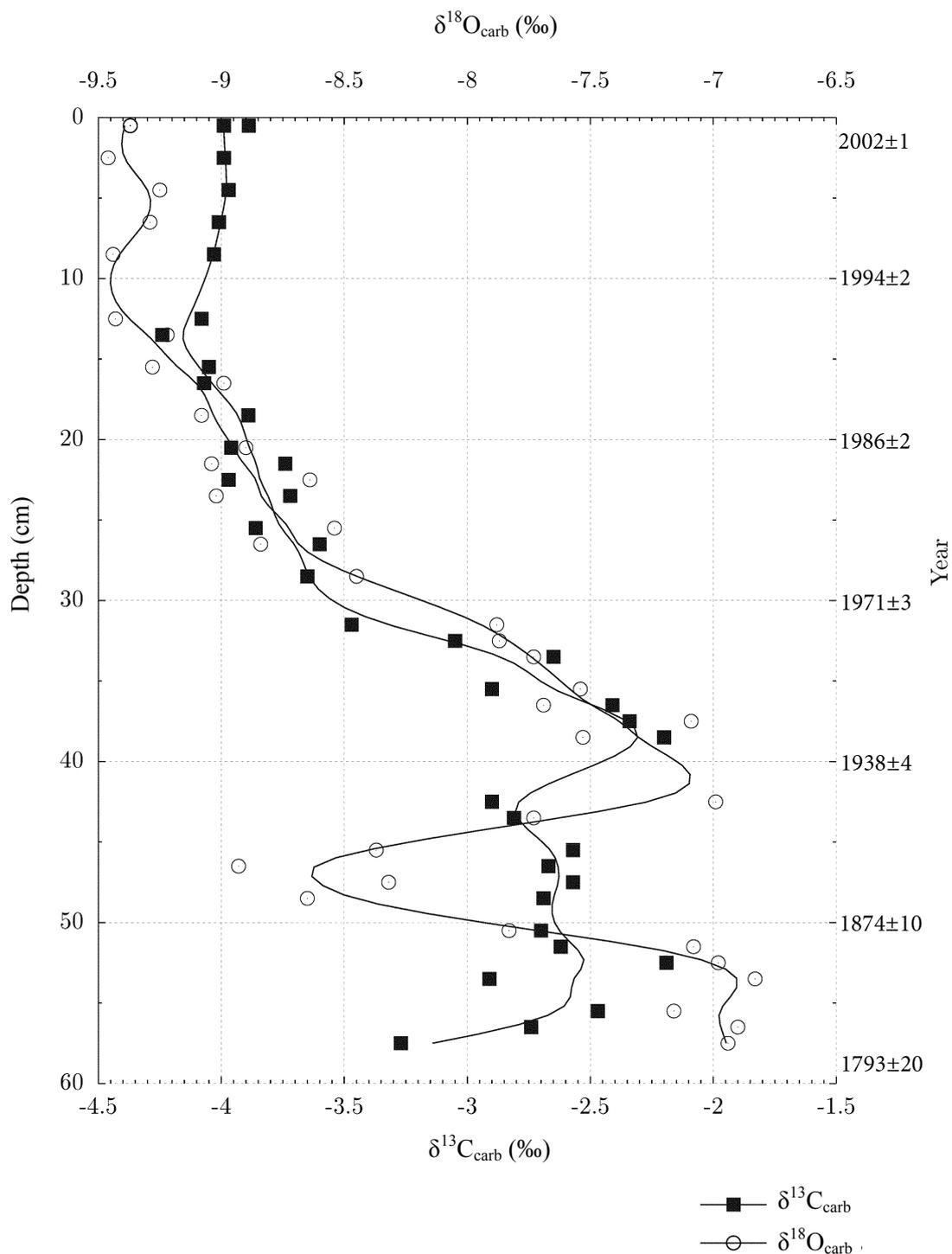


Fig. 6. Variation in  $\delta^{13}\text{C}$  and  $\delta^{18}\text{O}$  values in carbonate fraction of the sediment core.

**DISCUSSION**

Lithology, diatoms and stable isotope analysis data reflect clear changes in environmental conditions in the north-western part of the Curonian Lagoon during the last

ca 200 years. The investigated sediment sequence can be divided into two parts. Sediment composition, diatoms and stable isotope analysis show that significant changes in environmental conditions began approximately in the period 1940–1970 (sediment interval 40–30 cm).

The time of environmental changes reflected by different analysis is slightly different, therefore the sediments that accumulated approximately in 1800–1955 (58–35 cm) were named as Zone 1 and the sediments that accumulated in 1955–2002 (35–2 cm) – as Zone 2.

The deposition of sediments of Zone 1 took about 160 years according to the age model. The first signs of eutrophication in the Baltic Sea sediments are detected in the mid-1800s, although eutrophication strongly intensified only after the early to mid-1900s (Andrén et al. 1999; Savage et al. 2010; Tuovinen et al. 2010). The sediments that deposited in the period 1800–1955 are characterized by a prevalence of fresh-brackish diatoms (up to 99%). The species assemblages are strongly dominated by benthic *Fragilaria heidenii*. This species is common in the Curonian Lagoon and the life-form of the taxon in this particular environment is characterized as benthic, epipsammic (Snoeijs & Potapova 1995; Kasperovičienė & Vaikutienė 2007). A dominance of benthic diatoms is usually a characteristic feature of very shallow environment or of relatively low nutrient concentrations and turbidity (Andrén et al. 1999; Weckström 2006). However, planktonic fresh-brackish diatoms make up about 26% (17–63%) in LDAZ CL-1. The planktonic species *Aulacoseira granulata*, *Aulacoseira islandica* and *Stephanodiscus rotula*, usually characteristic of nutrient-rich, eutrophic waters (Van Dam et al. 1994; Aleksandrov & Dmitrieva 2006; Kiss et al. 2012), represent a large proportion in some samples of the zone. Based on diatoms, the northern part of the lagoon was relatively fresh, shallow and slightly eutrophied in the period 1800–1955. Variable values of  $\delta^{13}\text{C}$  in organics and  $\delta^{15}\text{N}$  as well as  $\delta^{13}\text{C}$  and  $\delta^{18}\text{O}$  values in carbonates in Zone 1 indicate an unstable environment. Low  $\delta^{13}\text{C}$  values in sediments resulted mainly from the input of freshwater phytoplankton and vegetation debris from the catchment (Peterson & Fry 1987), low ( $\sim 3.5\text{‰}$ )  $\delta^{15}\text{N}$  values in general are characteristic of terrestrial material (Middelburg & Nieuwenhuize 1998). The Nemunas River basin is the main source of fresh water, delivering 97.1% of its total run-off into the northern part of the lagoon (Jakimavičius & Kovalenkoviėnė 2010). The annual Nemunas River discharge increased during the period 1800–1935 and remained at the same level until 1960 (Gailiušis et al. 2011). The recent diatom and stable isotope data show that run-off of the Nemunas River was the main source of freshwater input (together with nutrients) into the lagoon until approximately 1955.

Small changes in the lithological composition of sediments can be noticed in the middle part of Zone 1 at a depth of 53–43 cm (approximately in the period 1860–1930): decreased percentage of clastic matter, increased carbonates and organics. At the same depth the curves of fresh-brackish planktonic *Aulacoseira*

*granulata*, *Aulacoseira islandica* and benthic *Fragilaria heidenii* fluctuate and slightly decrease but the percentage of fresh-brackish planktonic *Stephanodiscus rotula*, benthic *Rhoicosphenia abbreviata* and planktonic brackish *Cyclotella striata* increases. The  $\delta^{15}\text{N}$  values significantly increased but  $\delta^{18}\text{O}$  values decreased at a depth of 53–43 cm. Low values of  $\delta^{18}\text{O}$  usually are characteristic of freshwater inflow (Epstein & Mayeda 1953; Harwood et al. 2008). However, the fluctuation of the percentage of the mentioned diatom species (a small increase in brackish diatoms) and stable isotope values (higher values of  $\delta^{15}\text{N}$ ) can be related to changes in fresh- and brackish-water flows and temporally increased salinity in the investigated area of the lagoon. Declined mean annual discharge of the Nemunas River was observed around 1900 (Linkevičienė 2009). Brackish-water intrusions could be more frequent because of lower input of fresh water into the lagoon at that time. We can suppose that the described fluctuations of diatoms and stable isotope values in sediments that deposited during 1800–1955 can be related to natural changes in fresh- and brackish-water flows into the lagoon, because the strait retained the natural depth until 1960 (Gailiušis et al. 2005).

The eutrophication of European coastal areas due to nutrient loading from agricultural sources began in the 1950s after the Second World War (Newton et al. 2014). Various proxy data obtained from Baltic Sea sediments suggest a non-uniform timing of eutrophication in different areas of the sea. For example, the first signs of eutrophication in the southwestern part of the sea appeared approximately 130–140 years ago (Andrén et al. 1999), whereas in some parts of the Baltic Sea, changes in the trophic state only appeared after the 1980s (e.g. Archipelago Sea; Tuovinen et al. 2010). In the Gulf of Finland, the Oder estuary and the Arkona and Bornholm basins clear changes due to anthropogenic eutrophication were generally evident from the beginning of the 1940s (Struck et al. 2000; Weckström 2006). Our study in the northwestern part of the Curonian Lagoon suggests increasing eutrophication in the lagoon approximately from 1955, corresponding well to many other areas of the Baltic Sea. The accumulation rate of fine silty mud in the northwestern part of the Curonian Lagoon increased from the beginning of the 1940s. The decreased percentage values of fresh-brackish periphytic diatoms (mostly *Fragilaria heidenii*) in LDAZ CL-2 likely indicates lower transparency of the water column because of increased phytoplankton abundance (Karosienė & Paškauskas 2012). Abundantly were found in this zone planktonic brackish-fresh *Actinocyclus normanii* and fresh-brackish *Aulacoseira islandica* diatoms, which overall characterize increased eutrophication and turbidity (Weckström 2006; Wang et al. 2008; Tuovinen et al. 2010). Previous investigations on the Curonian Lagoon

phytoplankton communities revealed that the increase in phytoplankton growth and diatom biomass might be influenced by elevated phosphorus and dissolved inorganic nitrogen concentrations in the water, and increased salinity and turbidity (Gasiūnaitė et al. 2005; Karosienė & Paškauskas 2012). Gradually increasing and varying (approximately between 5‰ and 6‰)  $\delta^{15}\text{N}$  values within Zone 2 can be related to more frequent brackish-water inflows and a large allochthonous material input from the sea (Müller & Voss 1999; Yamamuro & Kanai 2005) as well as  $^{15}\text{N}$ -enriched sewage from the catchment (Voss et al. 2000; Remeikaitė-Nikienė et al. 2016). Meanwhile, carbon isotope values vary in a narrow range and remain low (about –30‰), indicating a stable freshwater phytoplankton input into the bulk biomass of sediments (Lesutienė et al. 2014; Remeikaitė-Nikienė et al. 2016). Carbon and oxygen stable isotope ratios in carbonates show a decreasing trend in the topmost part of the sediment core. The values of  $\delta^{18}\text{O}$  vary around –9‰ and are characteristic of freshwater inflow (Harwood et al. 2008). The stable isotope data show that freshwater discharge still was the main source of fresh water and nutrients in the lagoon after 1955. However, a couple of local coinciding anthropogenic activities could accelerate eutrophication and more frequent brackish-water intrusions in the northern part of the Curonian Lagoon and resulted in an increased accumulation of planktonic (especially brackish-fresh *Actinocyclus normanii*) diatoms in the sediments. Starting from 1960, the annual run-off of the Nemunas River decreased because of the installed big reservoir on the Nemunas River and the launching of the Kaunas Hydropower Station (Gailiūšis et al. 2011). These changes coincide with the Klaipėda Strait deepening in 1960–1962 (Gailiūšis et al. 2005). The water flow in the strait increased by about 10% and the annual brackish-water inflow increased by about 24% compared to inflow volumes before the dredging (Gailiūšis et al. 2005; Dailidienė & Davulienė 2008; Jakimavičius & Kriauciūnienė 2011). The rising sea level in the SE Baltic, which is especially noticeable since the 1970s (Dailidienė et al. 2006) also added to a more intensive brackish-water inflow to the lagoon. Such a setting has introduced conditions for more frequent flows of brackish water into the northern part of the lagoon, increased turbulence and resuspension, and more rapid accumulation of fine-grained sediments in the deep-water areas of the lagoon.

The location of the investigated sediment core is also important. It is in the western part of the lagoon, which is deeper and allows fine-grained sediments to accumulate there (Trimonis et al. 2003). According to salinity measurements, brackish-water currents from the Baltic Sea flow at the bottom along the western, deeper, part of the lagoon and fresh water of rivers flows in the eastern

part (Dailidienė & Davulienė 2008; Zemlys et al. 2013). Studies of the diatom community in the near-bottom water layer also demonstrate the salinity effect on the ecological conditions of the lagoon. Brackish diatoms made up to 30–40% of the total sum in July–October (1986–1987, 1991, 2003–2004), especially in the deep-water area of the lagoon (Kasperovičienė & Vaikutienė 2007). *Actinocyclus normanii* was dominant (up to 97%) in the phytoplankton in 2002 (Aleksandrov & Dmitrieva 2006) and 2003–2004 summer–autumn seasons (Kasperovičienė & Vaikutienė 2007). The phytoplankton composition is analysed in different time periods and seasons. It cannot be correlated directly with the diatom composition of sediments, because sediments accumulate diatoms of many seasons. However, data from phytoplankton analysis confirm a sporadic inflow of brackish water into the lagoon. *Actinocyclus normanii* is common in the Baltic Sea near freshwater discharge and abundant in plankton and sediments near the eastern coast of the Baltic Sea (Snoeijjs & Vilbaste 1994; Bubinas et al. 1998). It is considered that *Actinocyclus normanii* is also found in some freshwater basins due to increasing eutrophication (Kiss et al. 2012). Probably, this planktonic species abundantly gets into the northern part of the lagoon when favourable hydrometeorological conditions are formed and spreads there because of relatively high eutrophication of the lagoon. It has been established that higher sea level (compared to the lagoon) is formed and water flows from the sea via the narrow strait during periods of prevailing northwestern/western wind direction in autumn (Dailidienė & Davulienė 2007; Ferrarin et al. 2008; Jakimavičius & Kovalenkoviėnė 2010; Dailidienė et al. 2011). Data of sediment traps (located in the strait) revealed a high sediment flux in autumn–winter seasons, when the prevailing wind direction is from the West and Northwest. A large amount of *Actinocyclus normanii* brought from the sea with water and re-suspended sediments was found in the material from sediment traps, collected in autumn and winter of the 1997–2008 period (Trimonis et al. 2010). It can be assumed that two coincident local events had a significant influence on the environmental changes in the northern part of the Curonian Lagoon in 1960 and later. The hydropower station construction near the Nemunas River resulted in lower freshwater run-off into the lagoon and the deepening of the strait allowed more frequent brackish-water intrusions, resuspension and a large amount of sediment flux at the same time. Along with the overall increasing eutrophication of the Baltic Sea, related to industries developed in the densely populated catchment area (Voss et al. 2011), the two mentioned local reconstructions influenced the hydrological regime and more rapid eutrophication processes in the northern part of the Curonian Lagoon in the last decades.

## CONCLUSIONS

The recent study of the sediment core has revealed obvious changes in environmental conditions in the northern part of the Curonian Lagoon during the last two centuries. A low sedimentation rate of coarse silty mud, low percentage of organic matter, small  $\delta^{15}\text{N}$  values and the prevalence of fresh-brackish benthic diatoms in the sediments indicate little enrichment of the freshwater, shallow lagoon in the period of 1800–1955.

A noticeable increase in eutrophication is observed in the sediments that deposited approximately in 1955 and later, as in many parts of the Baltic Sea. Higher values of  $\delta^{15}\text{N}$ , abundant planktonic brackish-fresh and fresh-brackish diatoms characteristic of eutrophic waters indicate increased eutrophication in the lagoon during 1955–2002. Two simultaneous local circumstances (decreased freshwater discharge and the deepened strait) in 1960 and later could cause significant environmental changes in the lagoon. Stable isotope values fluctuate in a small range and mostly indicate prevailing freshwater discharge. However, abundant planktonic brackish-fresh *Actinocyclus normanii* can be related to increased eutrophication and more frequent brackish-water intrusions in the northern part of the lagoon. After the Second World War the eutrophication of the Curonian Lagoon was influenced by a general increase in eutrophication because of the urbanization of the coastal area like in the other parts of the Baltic Sea. A further increase in eutrophication was significantly influenced by local artificial constructions and activities, which resulted in changes in the hydrological and ecological conditions of the northern part of the lagoon.

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## **Inimtekkeline keskkonnamõju Läänemere Kura laguuni põhjaosas ränivetikate ja isotoopandmete valguses**

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Kura laguuni loodeosast võetud ajaliselt viimast kahtsadat aastat katvat settepuursüdamikku uuriti mitmete meetoditega, välja selgitamaks inimtekkelist keskkonnamõju ränivetikakoosluste, C, N ja O stabiilsete isotoopide ( $\delta^{13}\text{C}$ ,  $\delta^{15}\text{N}$  ning  $\delta^{18}\text{O}$ ) põhjal. Analüüs tuvastas kaks perioodi laguuni arengus, mis erinevad teineteisest settimiskiiruse, settetüübi ja veekogu troofsusastme poolest. Perioodi 1800–1955 iseloomustavad madalad  $\delta^{15}\text{N}$  väärtused ja põhiliselt riimageveeline ränivetikafloora. Alates 1960. aastast on märgata riimveelise keskkonna tugevnemist, mis on arvatavasti tingitud rajatistest Nemunase jõel ja Kura väina süvendamisest, aga ka Läänemere üldisest veetaseme tõusust.