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THE DISTRIBUTION OF CARBON STABLE ISOTOPES AS AN INDICATOR OF TEMPORAL AND SPATIAL DYNAMIC, AND DISTRIBUTION OF AUTOCHTONIC AND ALLOCHTONIC ORGANIC MATTER IN A SHALLOW ESTUARINE LAGOON

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Abstract. Physical and chemical parameters were measured in a mostly freshwater estuarine lagoon in the SE Baltic. Present paper demonstrates an attempt to trace the sources and analyse the seasonal and spatial patterns of distribution of POC, DIC and DOC in the Curonian lagoon mostly by the isotopic content in different forms of carbon. Samples were collected in 2012-2014 in 9 stations in the Curonian lagoon including riverine and marine input/output stations. Riverine inputs and summarizing outflow to the Baltic sea locations (Nemunas river delta and Klaipeda channel stations were sampled monthly, while POC, DIC and DOC samples in other stations were collected on a seasonal basis. The observed results allow easily differentiate between estuarine and riverine POM samples, while the differences in DOC δ13C content between sampling stations were found to be not statistically reliable. The high biological productivity of the Nemunas river along with the minor contribution of the Baltic Sea inflows to the overall hydrodynamics of the lagoon explain similarity of content between riverine and estuarine material in the spring and autumn. However, the δ13C content of DIC and DOC could serve as indicator of external inputs only in connection with seasonal water residence variations.

Keywords: particulate matter, nutrients, eutrophication, Baltic Sea, river discharge

1. Introduction

Studies of hydrologic mixing in estuarine environments using tracer elements are important in differentiating natural and anthropogenic contributions to pollution and eutrophication. The salinity, as a conservative tracer, is often used to construct both the mass-balance models and trace spatial mixing patterns in estuaries. However, in oligohaline coastal systems, dominated by the riverine inputs, the application of salinity could be not so effective. Stable isotopes are another useful tool for characterizing water dynamics within the watershed and estuarine systems, especially when constraining residence time and delineating water movement within the watershed. Likewise the stable isotopes could help in discriminating between different potential inputs (external inflow, precipitation and groundwater) of water to a system, what happens to the water within the system. Stable isotopes could help in determining mixing and flow paths of water within a system, making it possible to confirm, reject, or constrain models including mathematical circulation models. However, there are many constraints in stable isotope use, such non-conservative isotopes as c δ18C and δ12N could provide valuable information not only on the variability of external sources but also on seasonal and spatial patterns of biological processes in estuarine systems. Considering the variability in carbon input sources the autotrophy/heterotrophy characteristics of ecosystem could be tackled as well. Stable isotope analysis has been widely used to study organic matter sources supporting estuarine food webs (Pasquaud et al., 2007; Bouillon et al., 2011; Bristow et al., 2013) and even, quite recently, in the Curonian lagoon (Lesutiene et al., 2014). However in most cases the analysis included δ15N and δ13C in POM and living organism tissues rather than pools of DIC and DOC.

In this paper we aimed to assess (i) the seasonal variation of external input sources in isotopic contents of particulate organic matter entering and leaving the Curonian lagoon in 2012 (data collected before the start of the project) and 2013 -2014 (data collected within the project framework). Secondly (ii) we were aimed to analyze the spatial variation in δ15C isotopic content in POM, DOC and DIC samples collected seasonally in the water column at 9 stations in 2013. And consequently, by comparing the results to the annual hydrological cycle and biological process dynamic to analyze the differences in spatial and seasonal distribution of δ15C isotopic content in POM, DOC and DIC (iii).

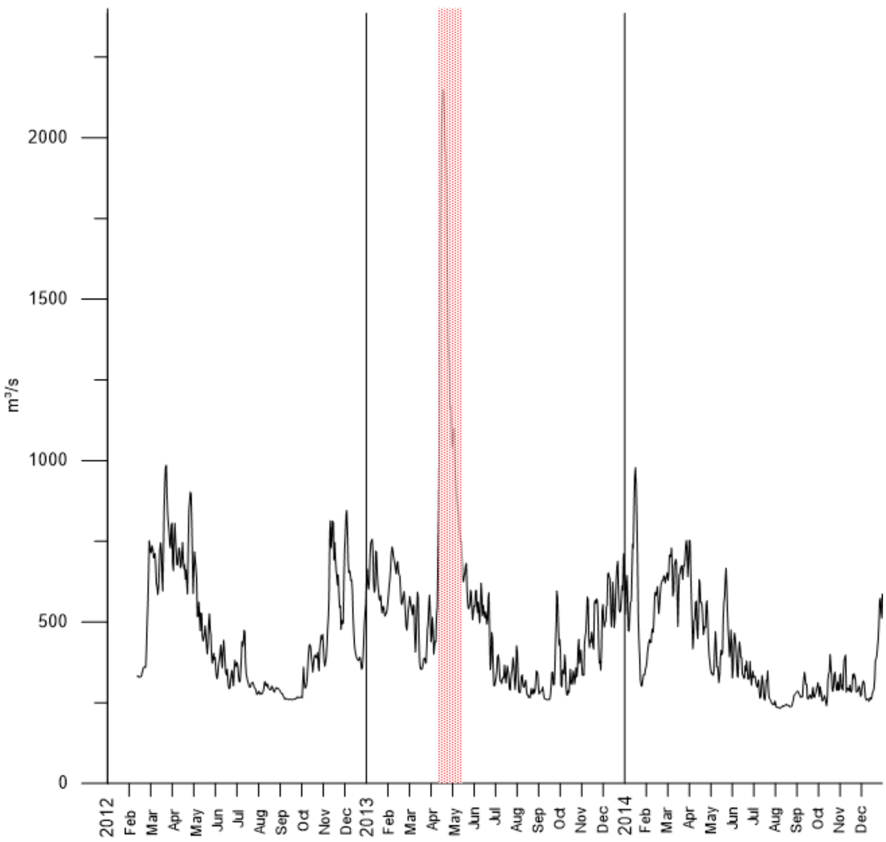

Fig. 3. **Seasonal dynamics of the Nemunas River discharge (m^3/s) in 2012-2014) (Lithuanian Hydrometeorological Service data). Extreme spring flood period in 2013 (> 700 m^3/s) marked.**

2. Materials and methods

2.1. Study Site and Hydrologic Conditions

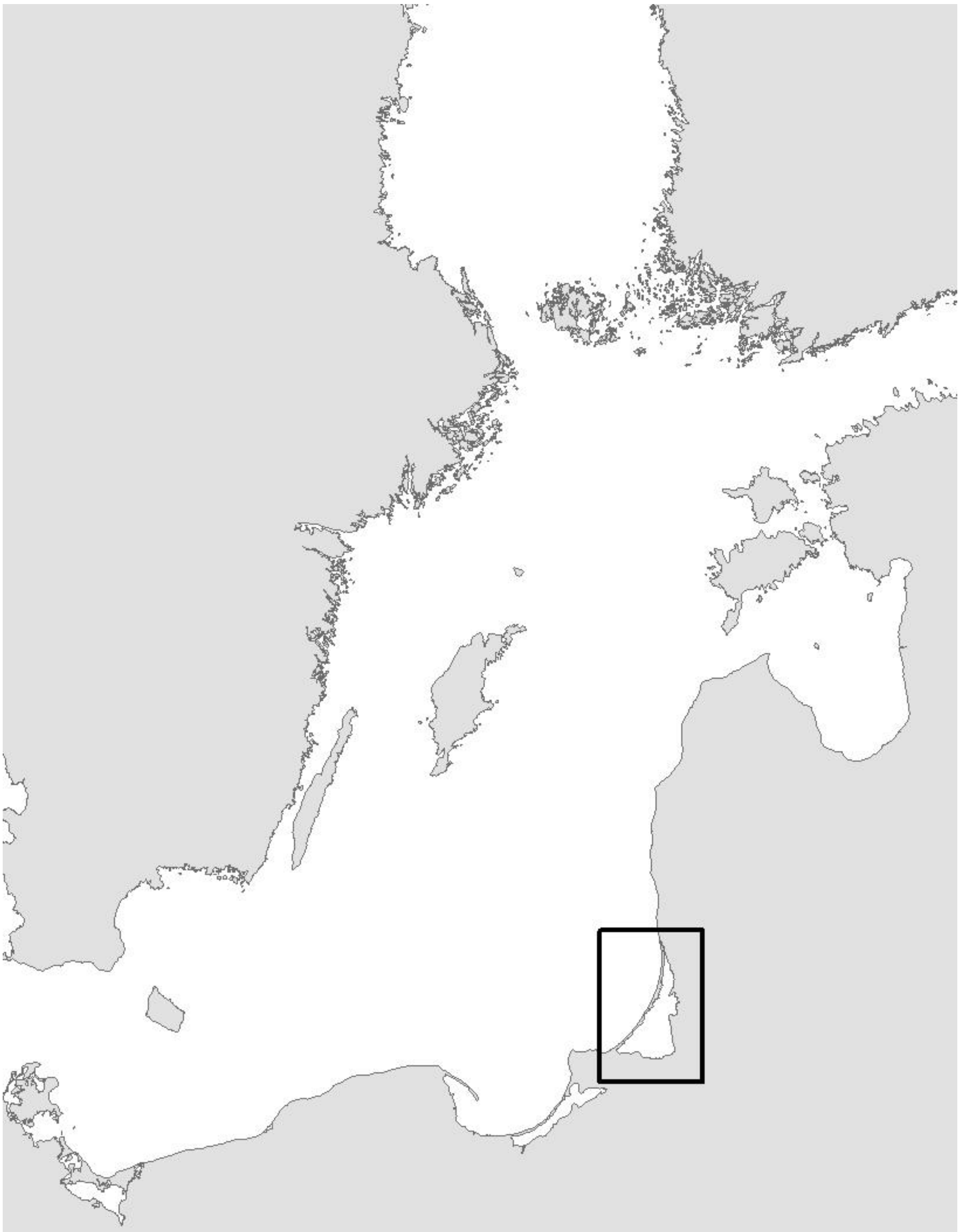

Fig. 1. **Location of the Curonian lagoon in the Baltic Sea .**

The Curonian Lagoon is a large, shallow water body (total area 1584 km^2, mean depth 3.8 m) located along the southeastern coast of the Baltic Sea (Figure 1). Curonian lagoon is an estuarine coastal lagoon, dominated by the Nemunas river discharges, which make up to 90 % of the total runoff (Razinkovas et al. 2008). The Lagoon is considered eutrophic or hyper-eutrophic due to cyanobacteria blooms which develop in summer, particularly during calm conditions, and include the presence of toxin-producing forms (Paldavičienė et al., 2009). The hydrology of the lagoon varies seasonally with changes in river discharge. Water residence time is typically 10 to 40 d during periods of elevated river discharge and increases to 60-100 d during low discharge (Ferrarin et al., 2008). During elevated discharge, the northern part of the lagoon is a mixing zone of marine, lagoon and riverine waters, while in the summer; there is little input to the lagoon from either source (Ferrarin et al., 2008). The Curonian Lagoon receives ~20% of its anual POM inputs from the Nemunas River – the fourth largest river entering the Baltic Sea (Jakimavičius and Kovalenkovienė, 2010). Tidal influence in the lagoon is minimal throughout the year but occasional wind-driven events push water from the Baltic Sea into the lagoon through the narrow Klaipeda Strait. The mixing of fresh and brackish water masses creates spatially and temporally unstable gradients with salinities ranging from 0 to 8 (values >7 are typical for the Baltic Sea) at a distance up to 20 km from the sea entrance to the lagoon (Dailidienė and Davulienė, 2008). The results of longterm water balance calculation in the Curonian Lagoon show that cumulative river inflow (Nemunas discharge near Smalininkai; Nemunas tributaries down from Smalininkai: Šešupė, Jūra, Šešuvis and Minija; tributaries of the Curonian Lagoon: Akmena-Danė and Deimena) is 21.847 km^3/year, precipitation – 1.194 km^3/year, and evaporation 1.006 km^3/year. Long–term brackish and fresh water exchange via the Klaipėda Strait is the following: inflow of brackish water from the Baltic Sea to the Curonian Lagoon – 6.125 km^3/year and fresh water runoff from the Curonian Lagoon to the Baltic Sea – 27.655 km^3/year (Fig 2). During the period of study (2012-2014), river discharge was highest in late April 2013, (> 1000 m3 s-1), which was quite uncommon (Dailidienė and Davulienė, 2008, Jakimavičius and Kovalenkovienė, 2010), whereas low discharge was observed during mid-late summer both in 2012 and 2013-2014 (< 300 m3 s-1; Fig. 3). Regarding the sources of organic carbon the autochthonous phytoplankton production is condireded to the dominating source (approximately 60% to the annual budget of POM) in this system (Galkus and Jokšas, 1997). Marine inputs entering the lagoon normally have a 3-6 fold lower concentration of particulate matter relative to inputs from the Nemunas River and therefore their contribution to the POM budget is small (Jokšas et al., 2005).

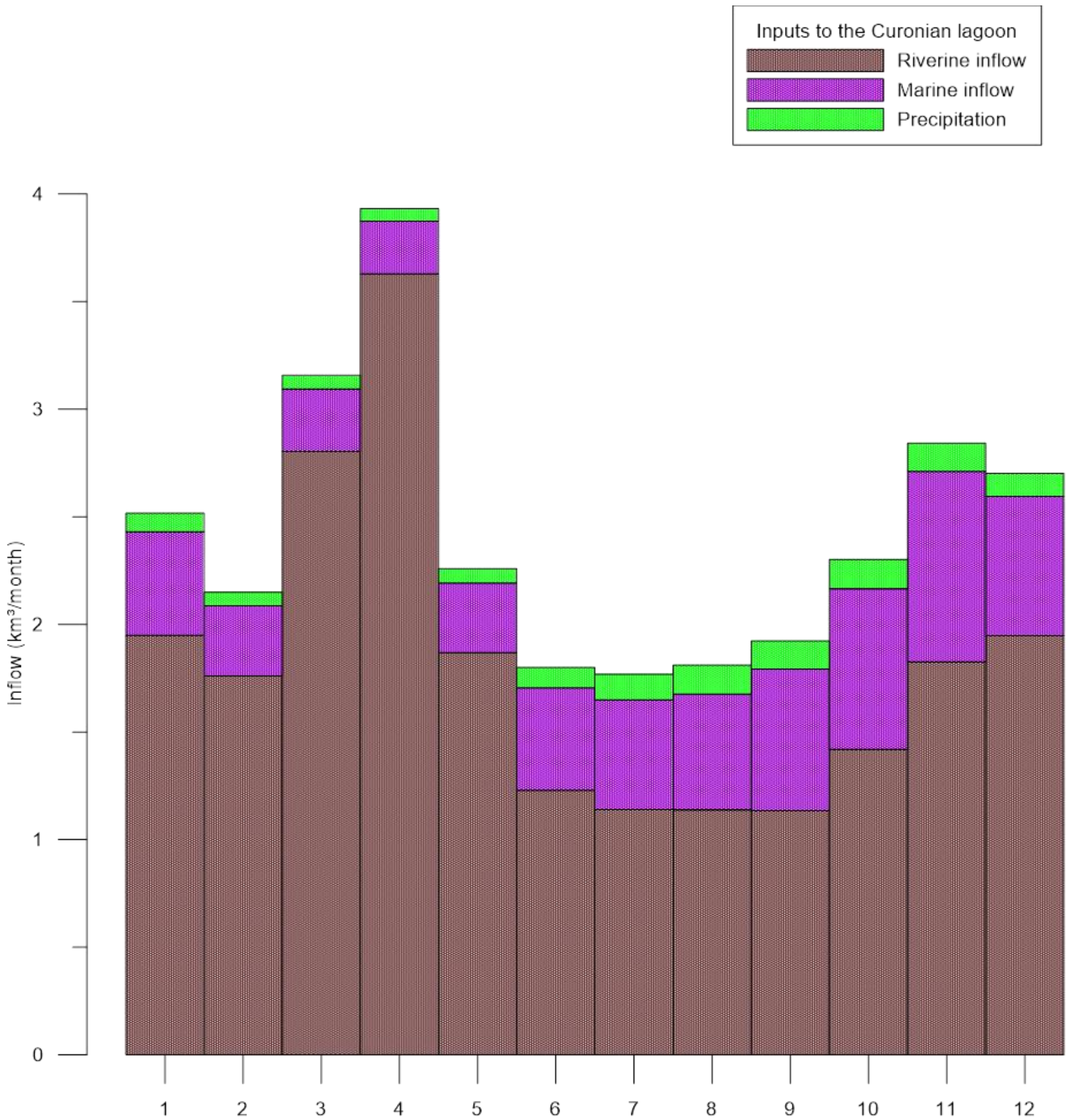

Fig. 2. **Monthly structure of the water inputs to the Curoninan lagoon in 1960-2007 (data from Jakimavičius and Kovalenkovienė, 2010) .**

2.2. Sampling and analysis

Sampling of riverine particulate organic matter (RPOM) and estuarine particulate organic matter (EPOM) at locations characterizing inflow (river mouth) and outflow (Klaipeda strait) was performed at monthly intervals in both 2012 and 2013. To assess the spatial variation in isotopic EPOM contents in different parts of the lagoon in February April and July 2013 samples were collected at 9 stations distributed across the lagoon (Fig. 1). Three replicate water samples were collected using a 10-L sampling bottle and transported to the laboratory in acid pre-washed plastic containers. EPOM samples were taken at 1 m depth in calm weather to avoid periods of heavy resuspension. Temperature, salinity and other chemical parameters were measured on each sampling occasion.

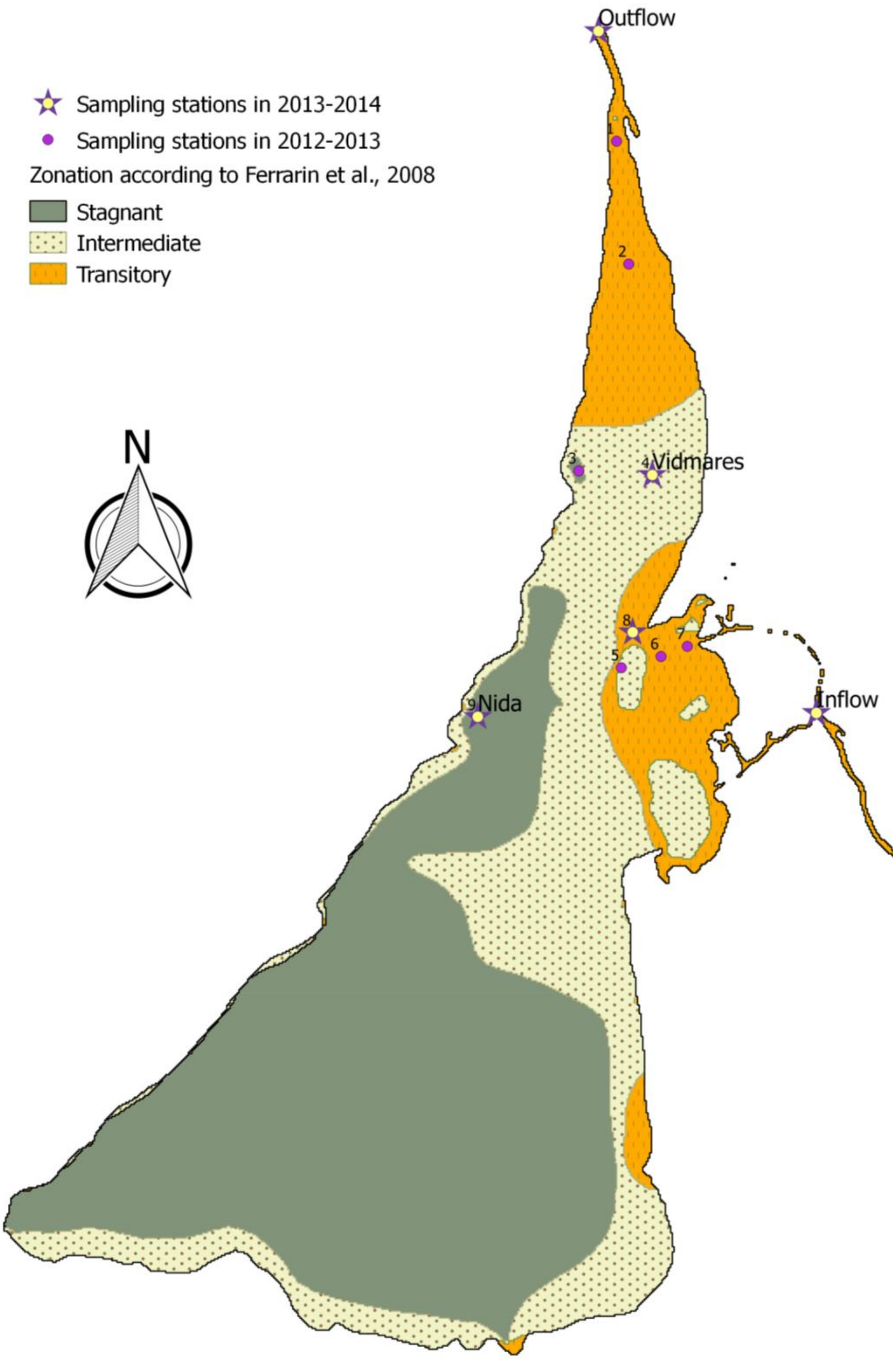

Fig. 3. **Pelagic sampling sites distribution plotted against the hydraulic regime based zonation map**

POM samples were prefiltered through 70 µm mesh to remove zooplankton and retained onto glass fibre filters (Whatman, GF/F, 0.7µm pore size). POM concentrations (< 70 µm fraction, mg DW L-1) and chlorophyll a content (µg L-1) were determined for each sample. Filters were dried for 48 h at 60 ˚C and weighed to determine total suspended solids. From each filter, three subsamples were prepared for SIA by cutting a 4-8 mm diameter circle using a puncher, packed into tin capsules, and used as analytical replicates. For each filter, 10 to 20% of the filtering area was analysed. Carbonates have near zero δ13C, therefore must be removed by acidification (Cloern et al., 2002). In this study no carbonate removal from POM filter samples was chosen as a compromise between accuracies for δ13C and δ15N estimates, because neutralization of carbonates by acid wash decrease δ15N values in the organic matter (Lorrain et al., 2003, Jacob et al., 2005).

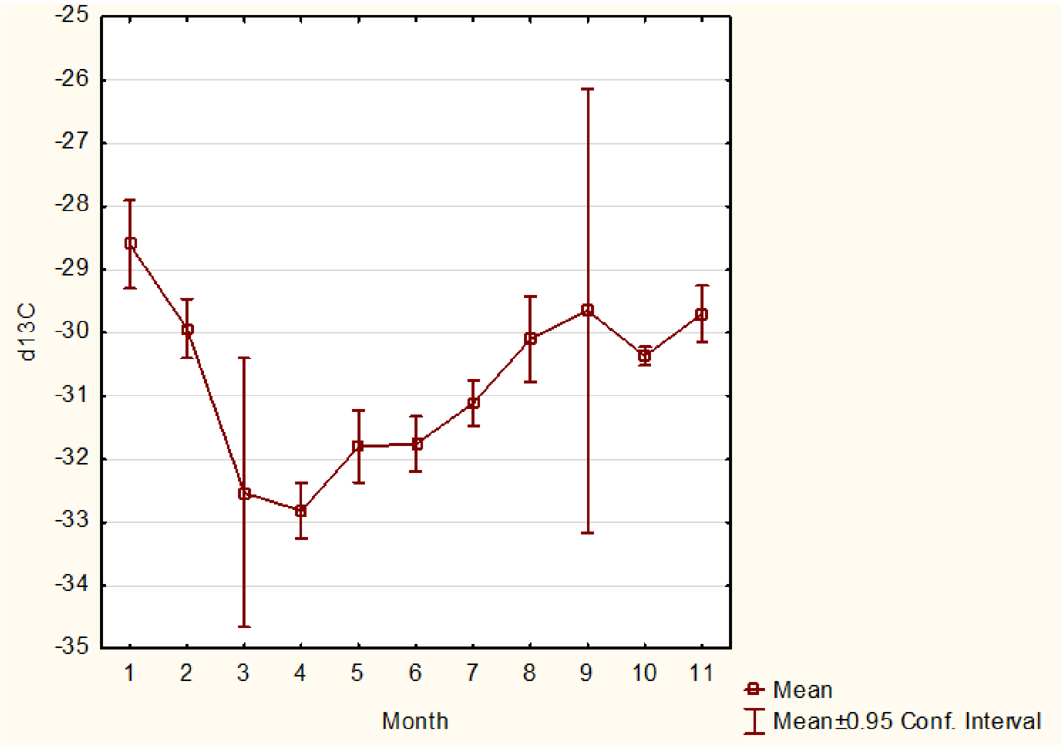

Fig. 5. **Seasonal variation in δ13C values determined in suspended particulate organic matter samples collected in the Curonian lagoon in 2013 - 2014.**

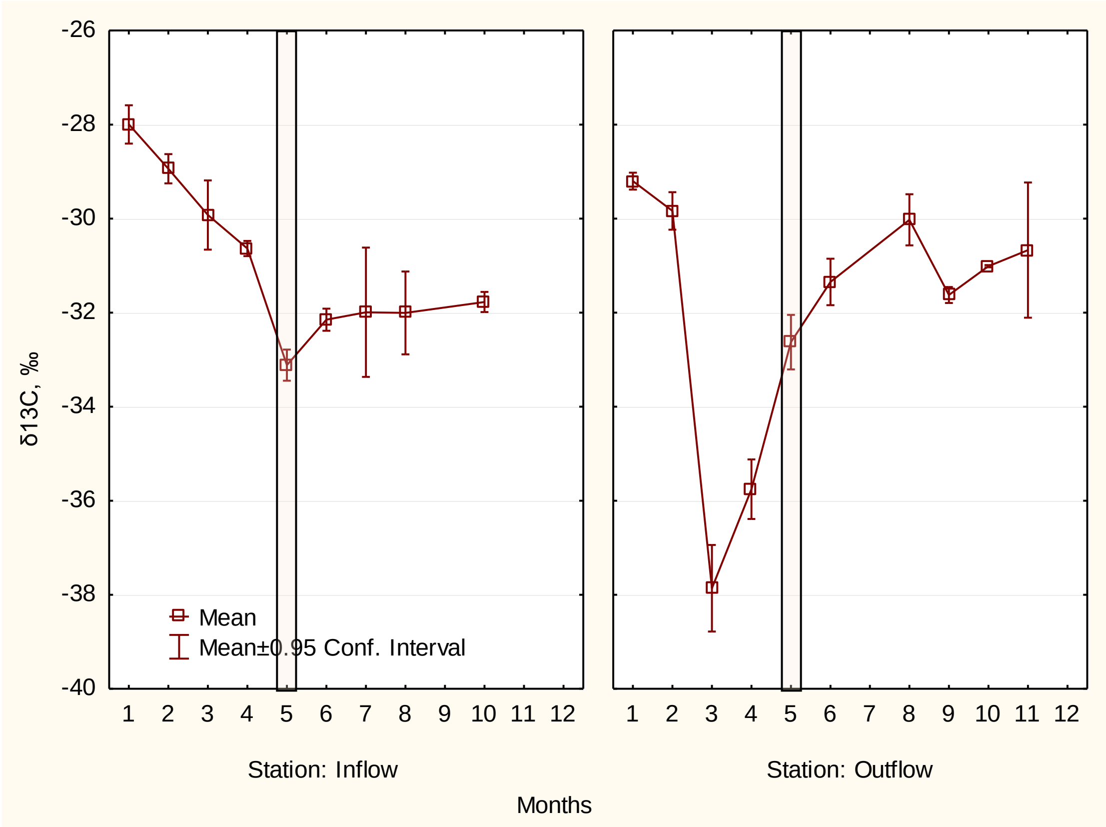

Fig. 6. **Seasonal variation in δ13C values determined in suspended particulate organic matter (POC) samples collected at the stations representing inputs and outputs from the Curonian lagoon in 2013. Extreme flood period in 2013 marked.**

2.3. Statistical methods and calculations

To assess the differences in δ13C content in POM, DIC and DOC samples collected at 9 stations in the Curonian lagoon combination of classification trees and ANOVA methods were applied. The computation was carried out with the statistical software R v3.0.2. To compute classification trees the R package for Recursive Partitioning and Regression Trees (rpart v4.1-8) was used. Rpart package implements the Classification and Regression Tree (CART) algorithm, first introduced by Breiman et al. (1984). A decision tree (a flow-chart-like structure, where each internal (non-leaf) node denotes a test on an attribute, each branch represents the outcome of a test, and each leaf (or terminal) node holds a class label) was constructed. The topmost node in a tree is the root node (Fig. 18). For the case presented in this paper, the Information gain was selected as a measure. It is based on the concept of entropy that comes from the information theory. Information gain is a statistical feature and it tells how good the given attribute separates examples from the training set in respect to their classification. While entropy, defines homogeneity of examples, the Information gain estimates probable reduce of entropy.

3. Results

The simple analysis of lumped of POM samples revealed quite uniform seasonal pattern of both values and variation in δ13C. The winter values are decreasing rapidly reaching minimum in March-April period (Fig. 5), characterized by the maximum variation, which also coincides with the maximum discharges entering the lagoon from the Nemunas river (Fig. 3). However it is critically low values of δ13C during the anomalous discharge level in 2013 that responsible for such variation (Fig. 6). The gradual increase in δ13C values along the summer is stabilized in September, when the variation again reaches the maximum values (Fig.5).

As the the Nemunas river provide the most important source of allochtonous carbon sampling situated in the Nemunas river delta was representing the riverine POM, DOC and DIC, while the station in Klaipeda channel representing outflow of estuarine POM, DOC and DIC (Fg 7).

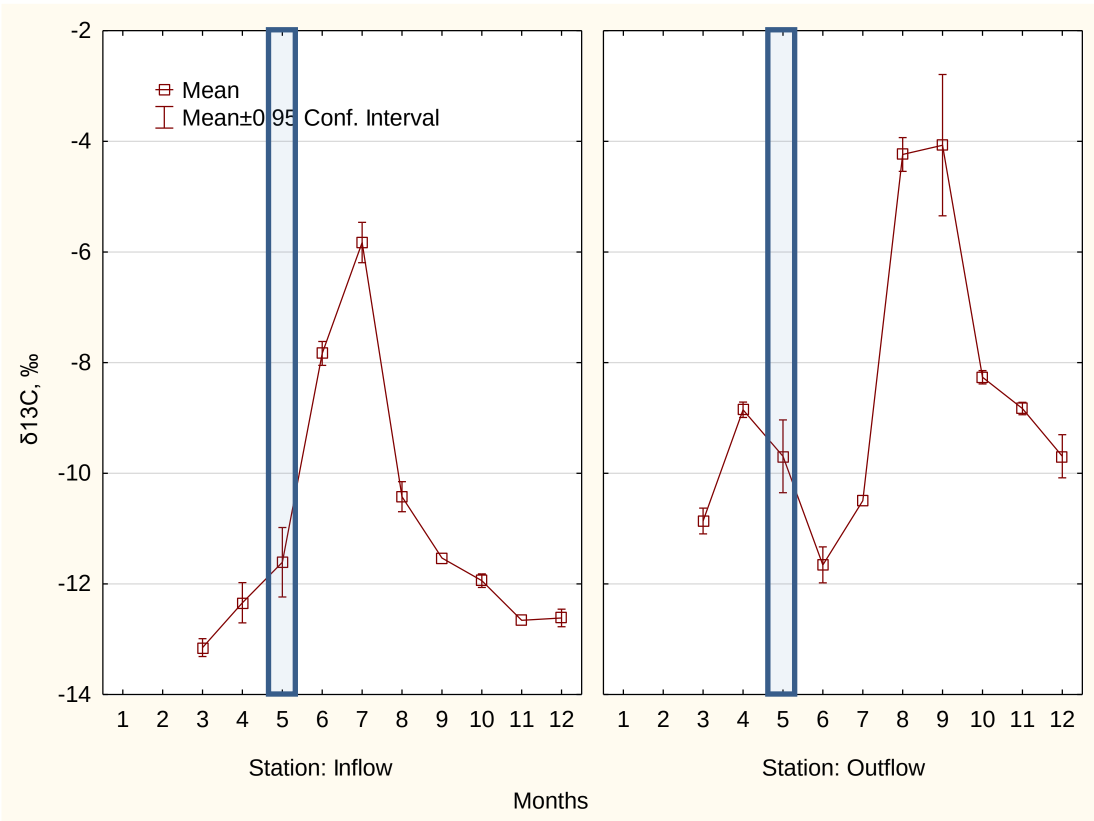

Fig. 7. **Seasonal variation in δ13C values determined in dissolved inorganic carbon (DIC) samples collected at the stations representing inputs and outputs from the Curonian lagoon in 2013. Extreme flood period in 2013 marked.**

The dissolved inorganic carbon (DIC) isotopic content featured clear seasonal patterns both in the riverine lagoon inputs and in the water masses leaving the Curonian lagoon. However there were clear differences in temporal dynamics. The riverine DIC feature the single peak during the midsummer season, while the highest δ13C values in waters leaving the Curonian lagoon were observed in August-September period (Fig.8). It could be stated that in general DIC δ13C values in riverine waters were lower than estuarine ones.

DOC isotopic content seasonal patterns were similar in both riverine and estuarine waters featuring gradual decrease in spring-autumn period (Fig. 9).

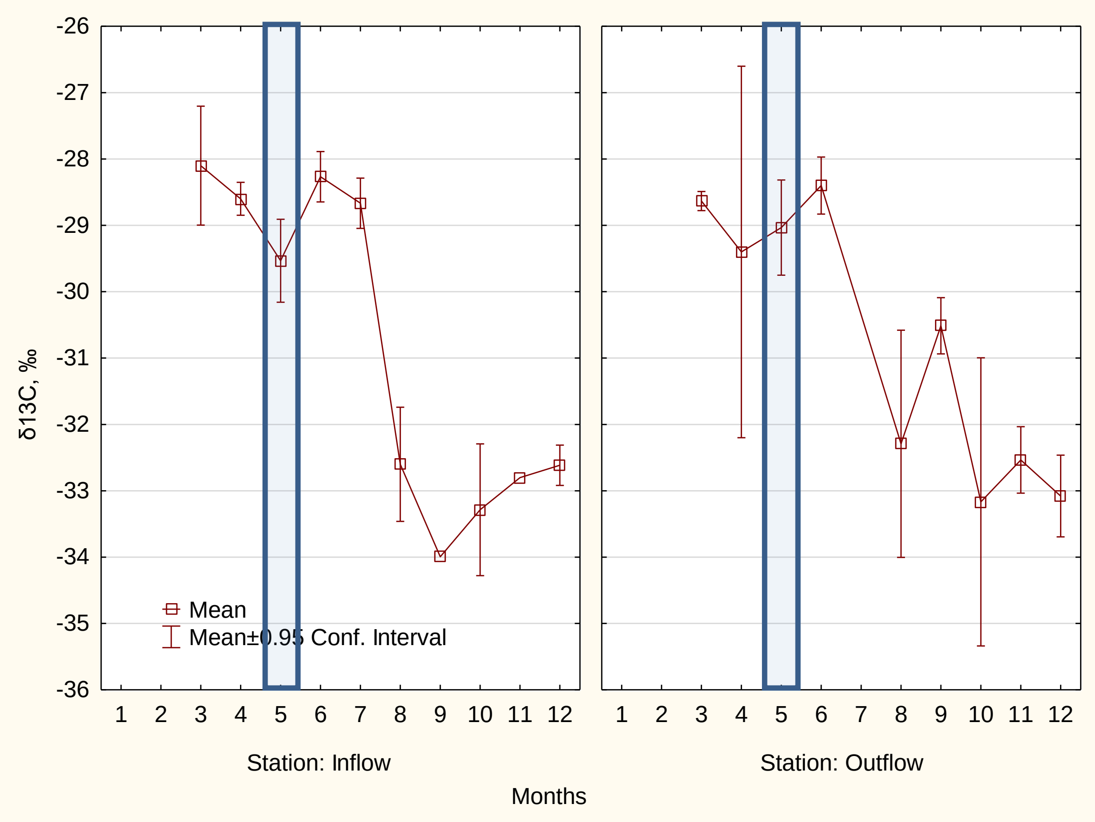

Fig. 9. **Seasonal variation in δ13C values determined in dissolved organic carbon (DOC) samples collected at the stations representing inputs and outputs from the Curonian lagoon in 2013. Extreme flood period in 2013 marked.**

As both the DOC and DIC are considered to be related to biological processes we used Chl A content in the water column as a proxy of biological activity. However there was reliable statistical relationship only between Chlorophyll A and DIC and POM values showing the opposite character correlations (Fig. 8-11 and 14).

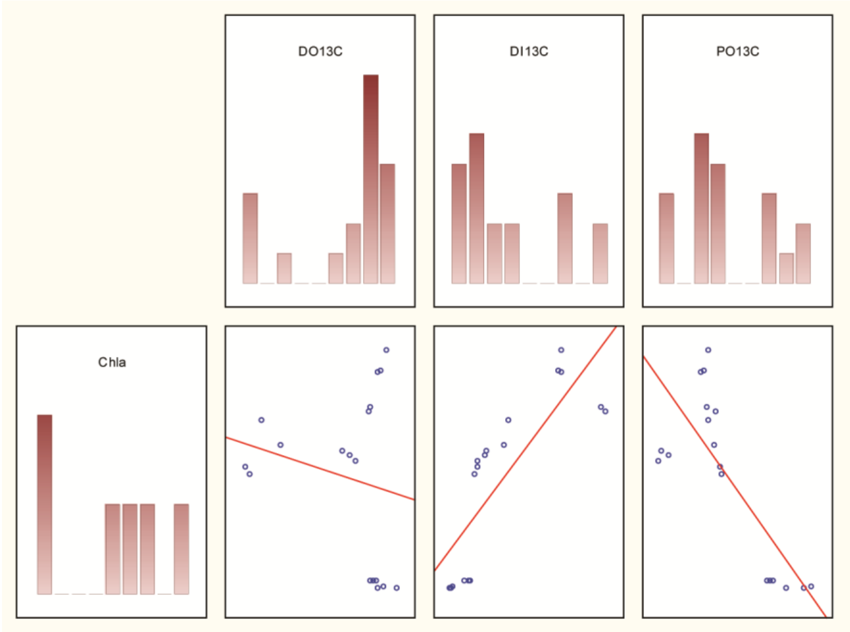

Fig. 8. **Relationship between concentration of Chlorophyll A and δ13C values determined at the station representing riverine inputs to the Curonian lagoon in 2013 (correlations were significant between Chl A and both DIC and POM).**

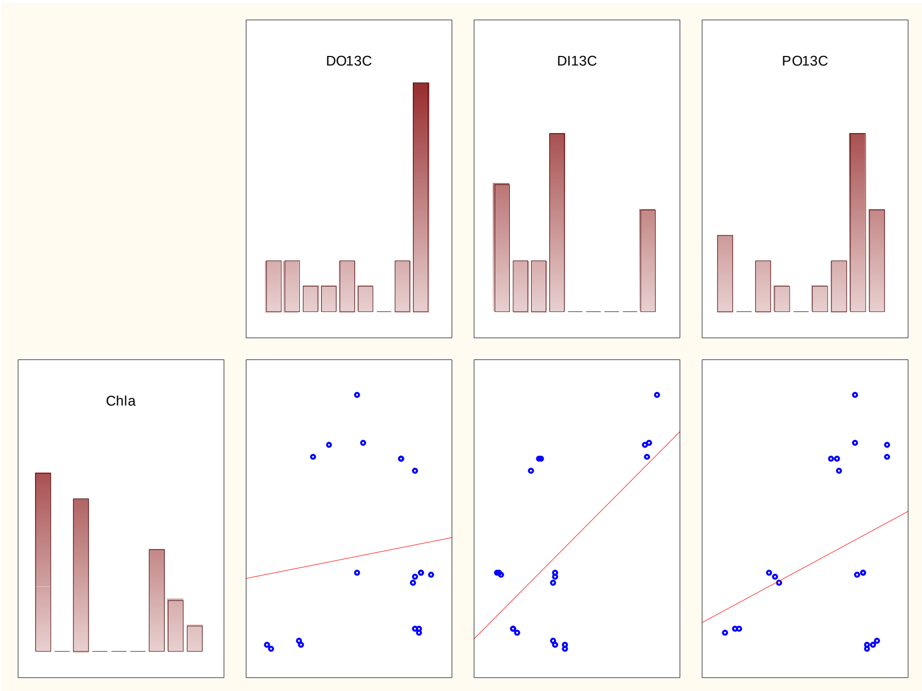

Fig. 9. **Relationship between concentration of Chlorophyll A and δ13C values determined at the station representing outflow from the Curonian lagoon in 2013 (correlations were significant between Chl A and DIC).**

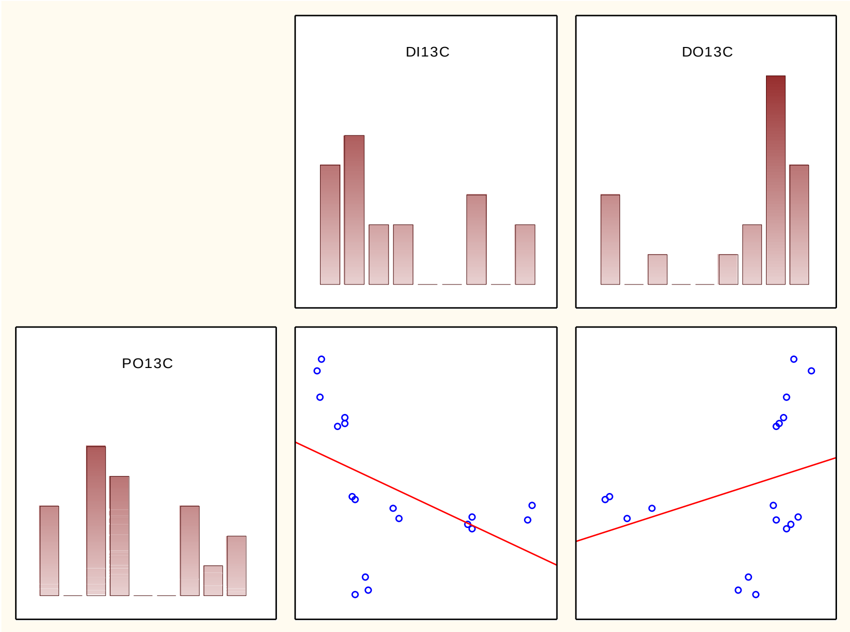

Fig. 10. **Relationship between concentration of POM and DIC and DOC δ13C values at the station representing inflow to the Curonian lagoon in 2013 (significant correlations between POM and DOC).**

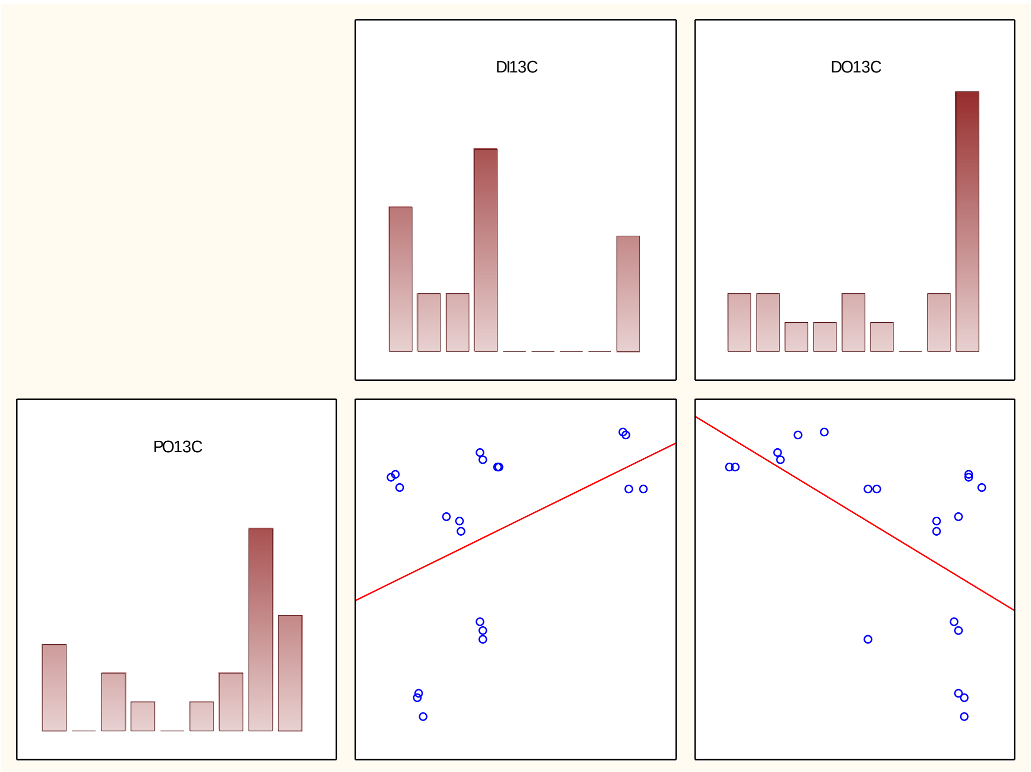

Fig. 11. **Relationship between concentration of POM and DIC and DOC δ13C values determined at the station representing ouflow from the Curonian lagoon in 2013 (correlations were significant between POM and DOC).**

One of the sampling points (No. 4) according to the hydraulic zonation scheme was representing stagnant part of the lagoon characterized by long residence time and least influence from the riverine or outflow or estuarine intrusions of the brackish waters from the Baltic sea. We used it as a proxy for large southern portion of the Curonian lagoon. It also should serve as indicator of allochtonous carbon mostly produced by internal biological activity.

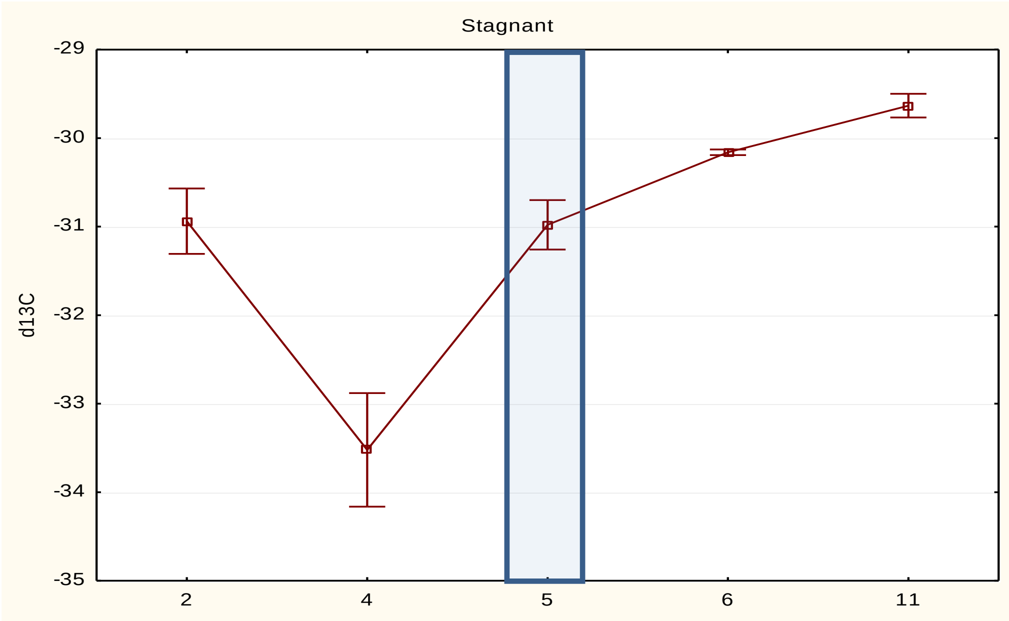

Fig. 12. **Seasonal variation in δ13C values determined in suspended particulate organic matter samples collected at the water sampling point 4 (representing stagnant part of the lagoon). Extreme flood period in 2013 marked.**

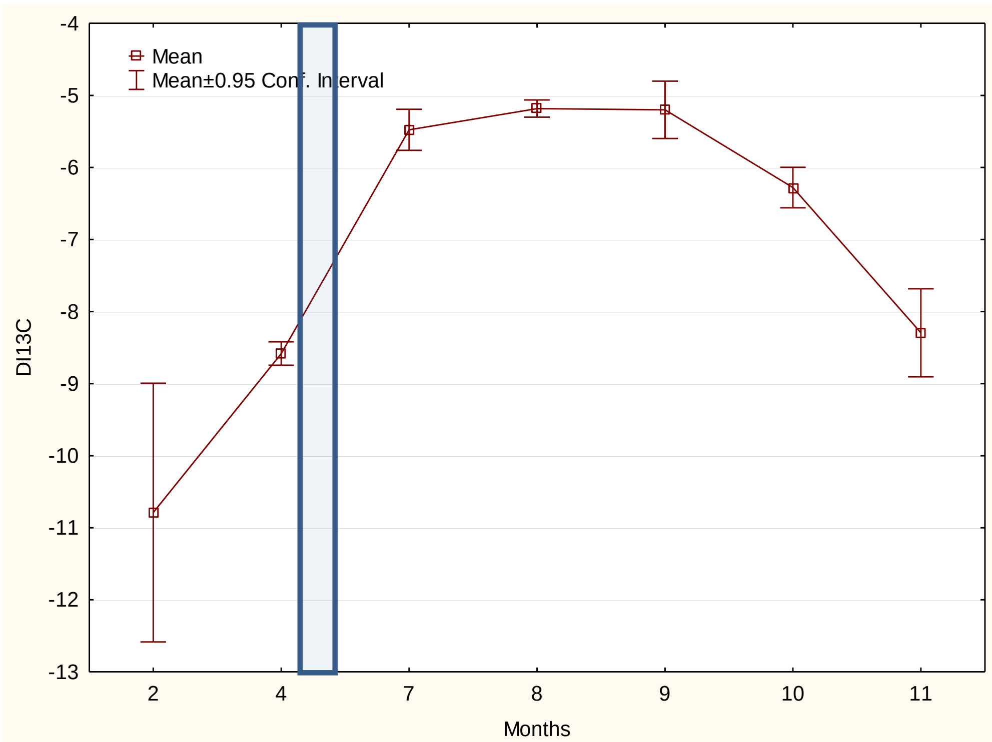

Fig. 13. **Seasonal variation in δ13C values determined in DIC samples collected at the water sampling point 4 (representing stagnant part of the lagoon). Extreme flood period in 2013 marked.**

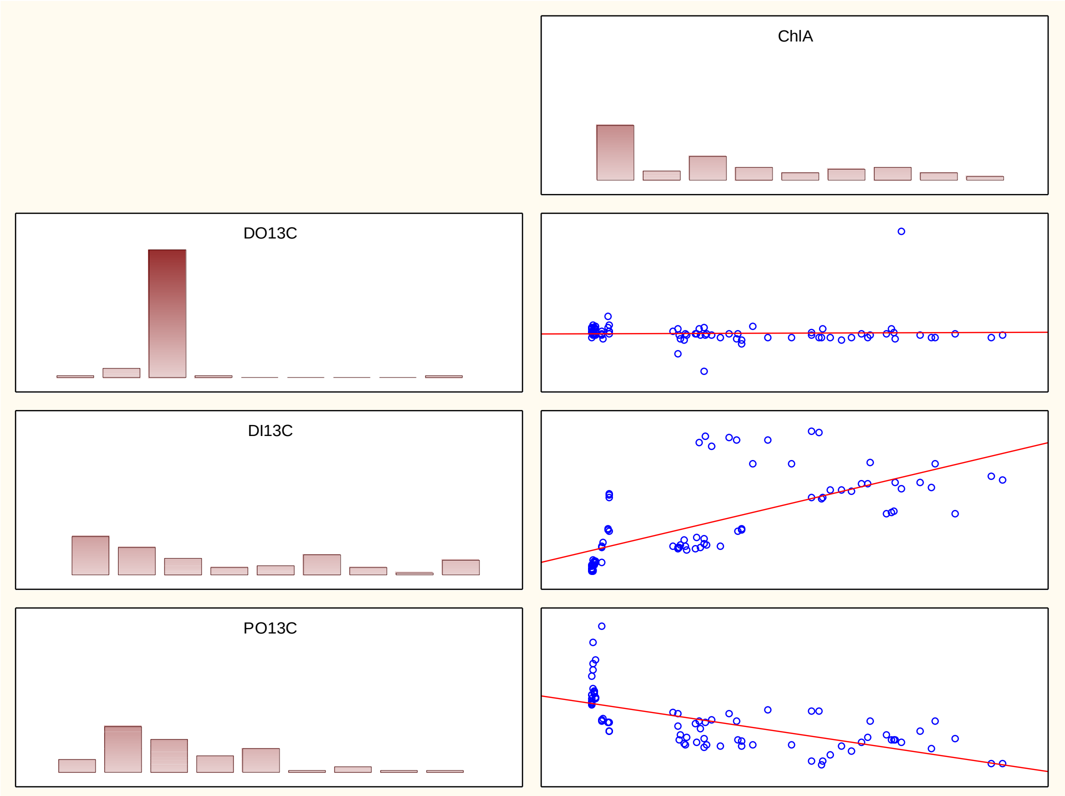

Fig. 14. **Relationship between concentration of POM and DIC and Chlorophyll A values determined at the stations in the lagoon (correlations were significant between Chl A and both DIC and POC).**

Discussion

Our data indicate that autochthonous organic matter contributes significantly to the total suspended matter and that the suspended organic matter composition cannot be explained in terms of conservative mixing of riverine and terrestrial sources on the one hand and marine sources on the other hand. As it was already mentioned by Lesutiene et al (2014) POM from the Nemunas River and Curonian Lagoon were well differentiated by their δ13C signatures in summer but not spring and autumn. Not surprisingly the dynamics of POM "stagnant" part of the lagoon is different from riverine POM and POM leaving the Curonian lagoon. The observed similarity between riverine and lagoon POM during spring and autumn in part to hydrologic influences whereby higher river discharge during these periods results in greater proportional contribution of POM from the river – that could be clearly seen when comparing the period of very late extreme flood in 2013. However the Nemunas itself is a lowland river which, like other low-gradient systems, can develop appreciable phytoplankton communities due to their low water velocity and long transit times (Lucas et al. 2009; Bukaveckas et al. 2011). So far only a 3 sampling sites were analyzed as representing very clearly the hydraulic zonation of the lagoon. Further research should be done by integrating the hydraulic and carbon cycling model currently in preparation. However we analyzed statistically the 9 stations representing the whole Central and Northern portions of the lagoon.

ANOVA analysis proved that differences in δ13C between collected DOC samples were not statistically significant (Fig. 17), while the isotopic content in POM and DIC samples was useful for differentiating.

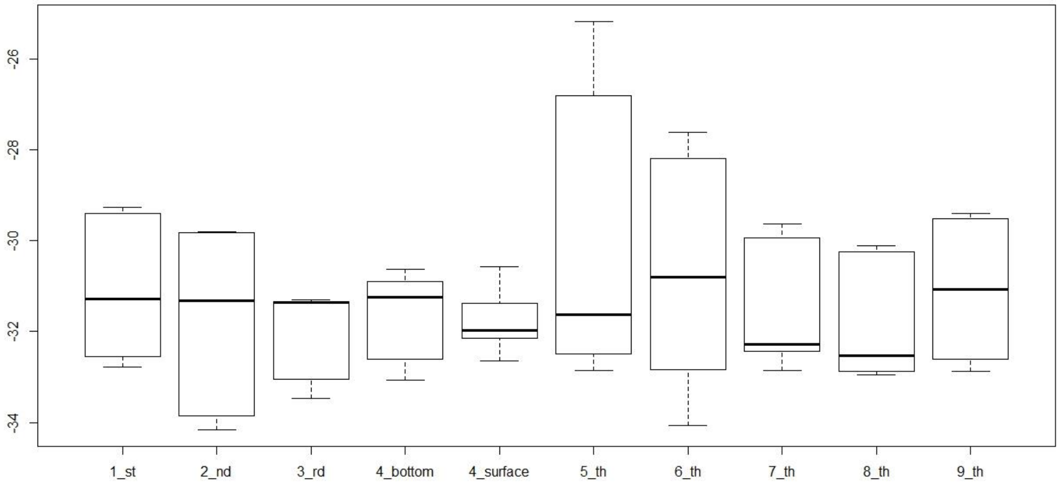

Fig. 15. **Variability of δ13C values determined in POM samples collected at different stations (Fig. 4) in the Curonian lagoon.**

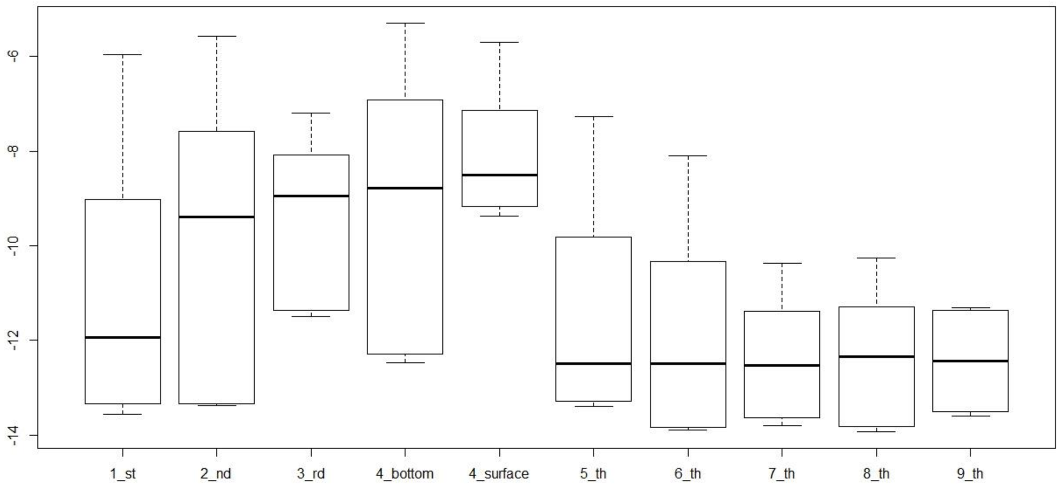

Fig. 16. **Variability of δ13C values determined in DIC samples collected at different stations (Fig. 4) in the Curonian lagoon.**

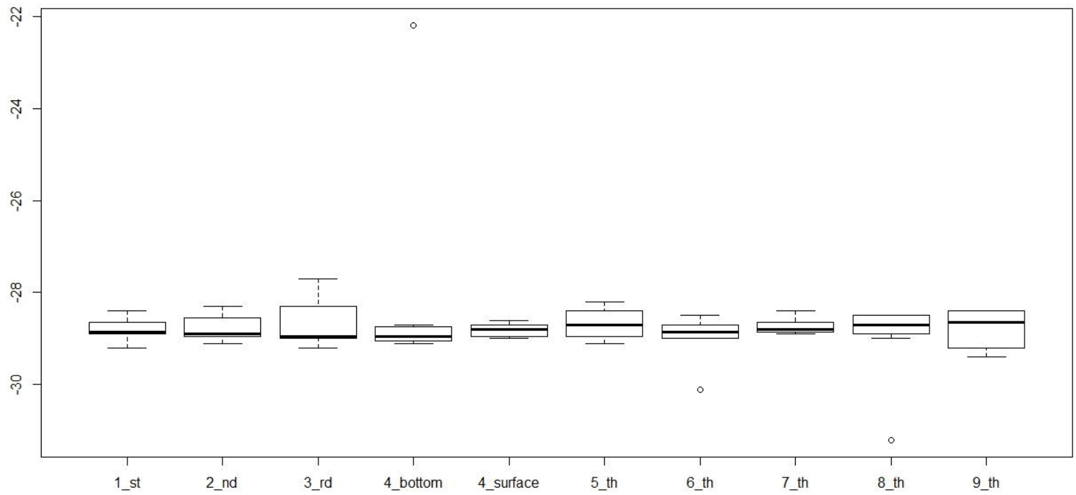

Fig. 17. **Variability of δ13C values determined in DOC samples collected at different stations (Fig. 4) in the Curonian lagoon.**

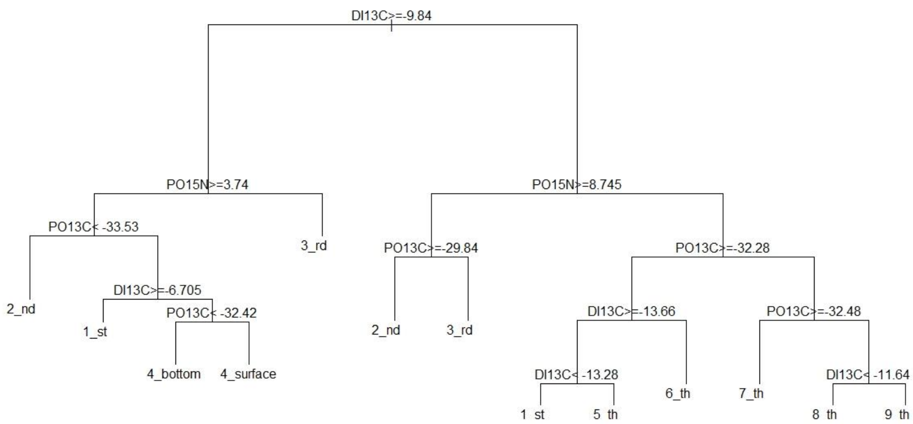

Fig. 18. **Classification tree produced using CART algorithm.**

According the results which based on computation of information gain DIC δ13C content appeared to be the best suitable attribute in describing differences between stations (Fig. 18).

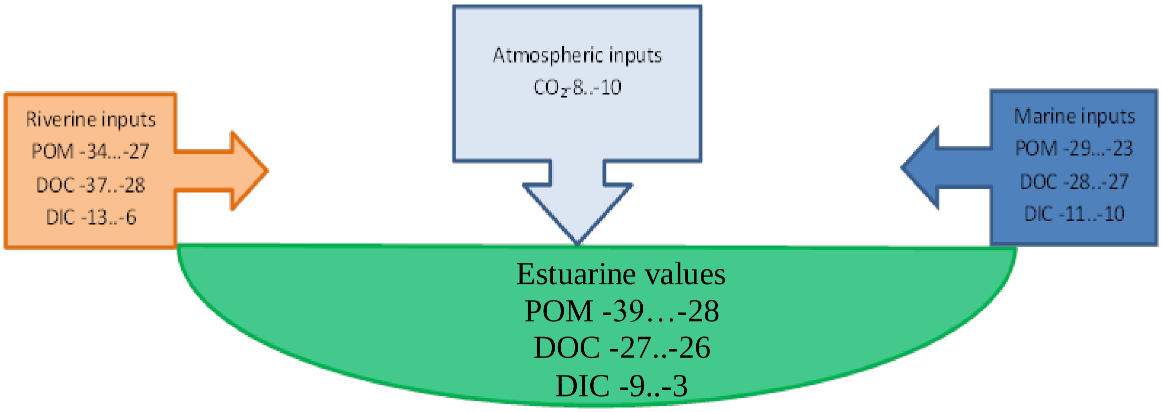

Fig. 19 **Schematic representation of carbon form and their isotopic content contributing to the organic matter in the Curonian lagoon**

Similarly to the findings of Chanton & Lewis (1999) we could state that the usefulness of both DOC and DIC δ13C data for characterizing the trophic processes within the estuary was dependent upon the residence time of water within the season.

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ANGLIES STABILIŲJŲ IZOTOPŲ PASISKIRSTYMAS KAIP AUTOCHTONINĖS IR ALOCHTONINĖS ORGANINĖS MEDŽIAGOS PASISKIRTSTYMO IR SEZONINĖS DINAMIKOS INDIKATORIUS SEKLIOJE ESTUARINĖJE LAGŪNOJE

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Santrauka

Anglies stabiliojo izotopo δ13C koncentracijos buvo nustatytos dalelinės suspenduotos organinės medžiagos (POM), neorganinės ištirpusios anglies (DIC) bei ištirpusios organinės anglies (DOC) mėginiuose Kuršių mariose. Mėginiai buvo surinkti 2012-2013 metais 9 stotyse centrinėje ir šiaurinėje Kuršių marių dalyse bei Nemuno deltoje ir Klaipėdos kanale. Stotys Nemuno deltoje ir Klaipėdos kanale yra sistemos įvesties ir išvesties taškai. Juose mėginiai buvo rinkti kiekvieną mėnesį, kitose stotyse medžiaga buvo renkama sezoniškai. Rezultatai parodė skirtumus tarp upinės ir estuarinės kilmės POM mėginių, tuo tarpu DOC mėginių skirtumai tarp atskirų stočių nebuvo reikšmingi. Didelį panašumą tarp upinės ir estuarinės kilmės mėginių ypatingai pavasarį ir rudenį galima paaiškinti aukštu Nemuno kaip lygumos upės biologiniu produktyvumu, tuo tarpu Baltijos jūros įtaka nėra reikšminga dėl palyginus nedidelio poveikio hidrologiniams procesams Kuršių mariose. DIC ir DOC izotopinės sudėties svarba apibūdinant trofinius procesus Kuršių mariose priklauso nuo sezoninės sistemos vandens apykaitos laiko kaitos.

Pagrindiniai žodžiai: Kuršių marios, stabilieji izotopai, δ13C, sezoninė dinamika