

**SOURCES AND GENESIS OF BURIED ORGANIC MATTER IN  
HOLOCENE SECTIONS OF SMALL LAKE SAPROPELS  
(SOUTHERN WEST SIBERIA AND EASTERN BAIKAL AREA)**

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**Abstract.** We consider methodological approaches to the identification of the sources and genesis of buried organic matter in lacustrine sediments by a number of organogeochemical indicators: data of a biological analysis (biostratification based on layer-by-layer determination of organic relics in bottom sediment sections); hydrocarbon biomarkers (molecular composition of normal aliphatic hydrocarbons (*n*-alkanes), nitrogen compounds of a protein complex, etc.); and C/N ratio reflecting a difference in the biochemical compositions of bioproducers. The results of biological analysis (biostratification) show that planktonogenic sapropel (phyto- and zooplankton, the autochthonous source of organic matter) in Lake Ochki formed for 10,760 years, and sphagnum and hypnum moss were supplied from the bogged shores (allochthonous source of organic matter). In Lake Minzelinskoe, peaty sapropel formed at the stage of a flooded lowland bog (5905 years ago); since 3980 years ago and till the present, macrophytogenic sapropel has formed.

**Keywords:** lake sapropel, genesis of organic matter, bioproducers, hydrocarbon biomarkers, biostratification, autochthonous source of organic matter

**Methodical approach to revealing sources and genesis of buried organic matter in the stratified Holocene sequences of lacustrine sapropels**

*Complex biological analysis (biostratification).* Revealing sources and genesis of lacustrine sapropels of organic matter (OM) within the stratified Holocene sequences is best performed by *direct method of quantitative layer-by-layer counting of remains of organisms (complex biological analysis – biostratification)* according to the methodical approach which was put forward by N.V. Korde, who is the well known expert in biostratification and typology of sapropels of small lakes in the European part of Russia and Eastern Baikal Region [6]. The data of the integrated biological analysis allow us to judge not only about sources of organic matter, but achieve a deciphering of sedimentation conditions in the geological past (Holocene and Late Glacial). In this regard, the direct method of biostratification should be recognized as prioritized and most reliable for the purposes of revealing sources and genesis of organic matter

of lacustrine sapropel sequences buried in the Holocene. In conjunction with this method, generally accepted organic-geochemical indicators may be used: hydrocarbon biomarkers (normal aliphatic hydrocarbons such as *n*-alkanes), components of protein-carbohydrate conjugate, hydrogen (HI) indices, and the ratio  $C_{org}/N_{org}$  [8].

**Hydrocarbon biomarkers.** The traditional method for study of molecular organic matter composition of oil source rocks is pyrolysis in the version of Rock-Eval (RE-pyrolysis) and the method of Chromato-Mass-Spectrometry (Pyr-GC MS), which, due to its simplicity and reliability began to be widely used for study of immature organic matter in samples of soils and recent lacustrine sediments [2]. This is based on the fact that pyrolysis of organic matter sample of recent sediment may be represented as a complex process comprised of a series of consecutive stages of decomposition of some organic matter components differing in nature and thermal stability and, consequently, in temperature intervals of their decomposition. As a result, a pyrogram of sample may be approximated by the sum of “simple” pyrograms of separate organic matter components. The method of deconvolution (factorization) of some experimental organic matter pyrograms into pyrograms of separate organic matter components proposed in [9] allows the contribution of each organic matter component in recent sediments to be estimated quantitatively.

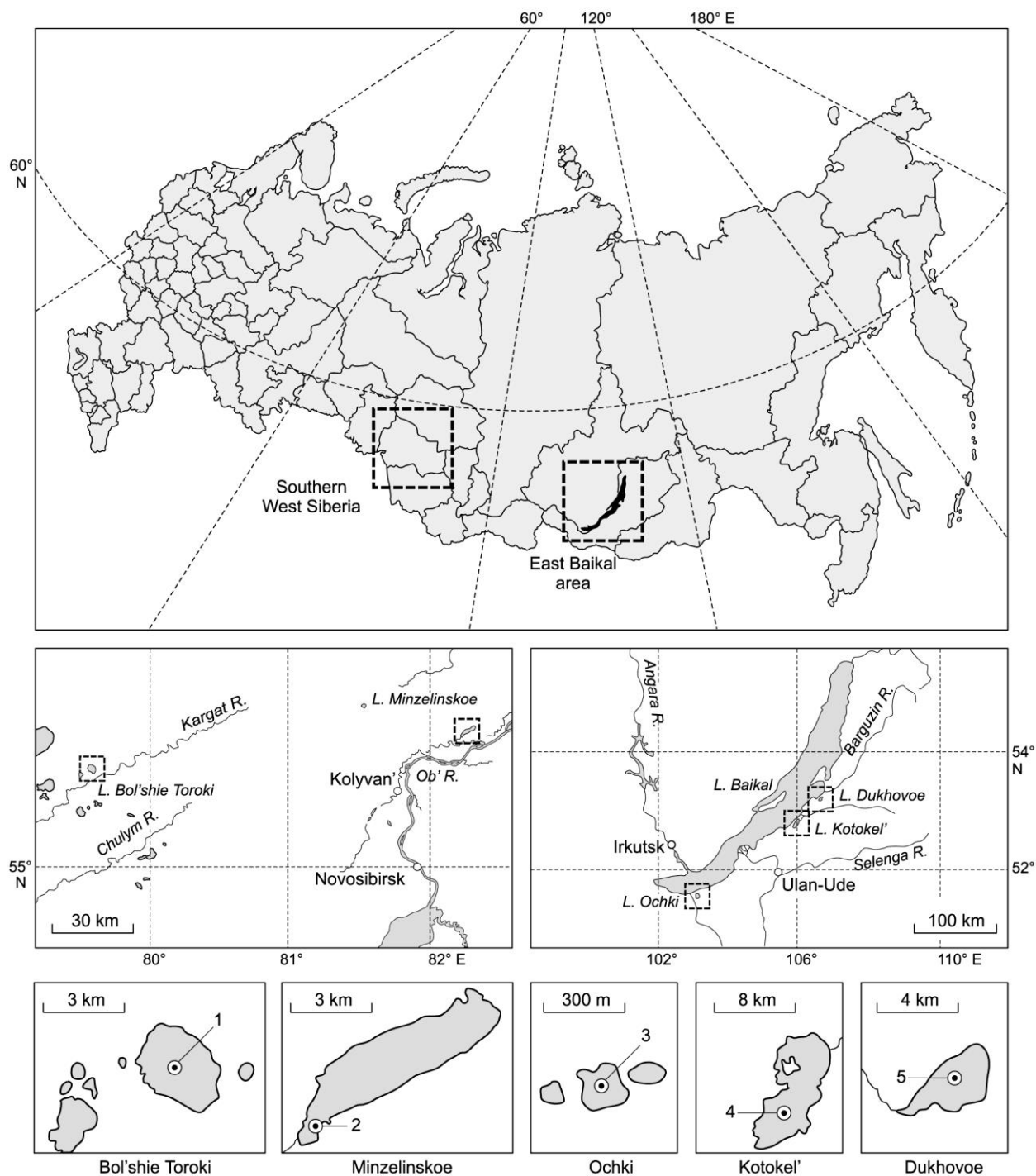
One of the most important relict hydrocarbons retaining their biochemical structure are *hydrocarbons of the hopane series*. In immature organic matter of recent sediments, biohopanes (products of diagenetic chain biosynthesis of hopanoid of bacteriohopanepolyol transformation) predominate. In immature organic matter of recent sediments, biohopanes (products of diagenetic transformation chain) predominate. Biosynthesis of hopanoids occurs in both aerobic and anaerobic bacteria as well as in some types of sulfate reducers and methanophores [1]. *Steranes*, saturated tetracyclic hydrocarbons  $C_{27}-C_{35}$ , along with hopanes, are also the most important relict biologically marking hydrocarbons [11].

One of main factors of OM transformation at the early stages of diagenesis is its fermentative processing by microorganisms. From biochemical OM components, proteins are the least stable. Further, in order of increasing stability, are hydrocarbons, lignin, and lipids. The preservation of individual chemical components is defined by their structure. Thus, usually the long-chain and isoprenoid *n*-alkanes are transformed to a lesser extent than oxygen and nitrogen-bearing functional groups or unsaturated carbon-carbon bonds [10].

**The ratio of organic carbon to organic nitrogen C/N** reflects differences in the biochemical composition of organisms and gives a possibility of approximately (due to uncertainty of the degree of diagenetic OM transformation) judging the genesis of organic matter of bottom sediments [14]. It is known that higher terrestrial plants are depleted in nitrogen and

have a high C/N (20–40) value [12]. This ratio for diatomic plankton is 5.5–7.0 [4] and for Baikal phytoplankton (*Melosira baicalensis*) it is close to 10 [15]. The lowest C/N value (4.0–4.5) (Vinogradov, 1938) is typical of zooplankton; even lower value is C/N (2.8–3.4) [13].

**Subjects of research.** For our research, we selected typical small lakes with organic-mineral sapropel types in the south of Western Siberia (Bolshie Toroki and Minzelinskoe) and typical small lakes in the Eastern Baikal Region with organic sapropel type (Kotokel', Dukhovoe, and Ochki) (Fig. 1).

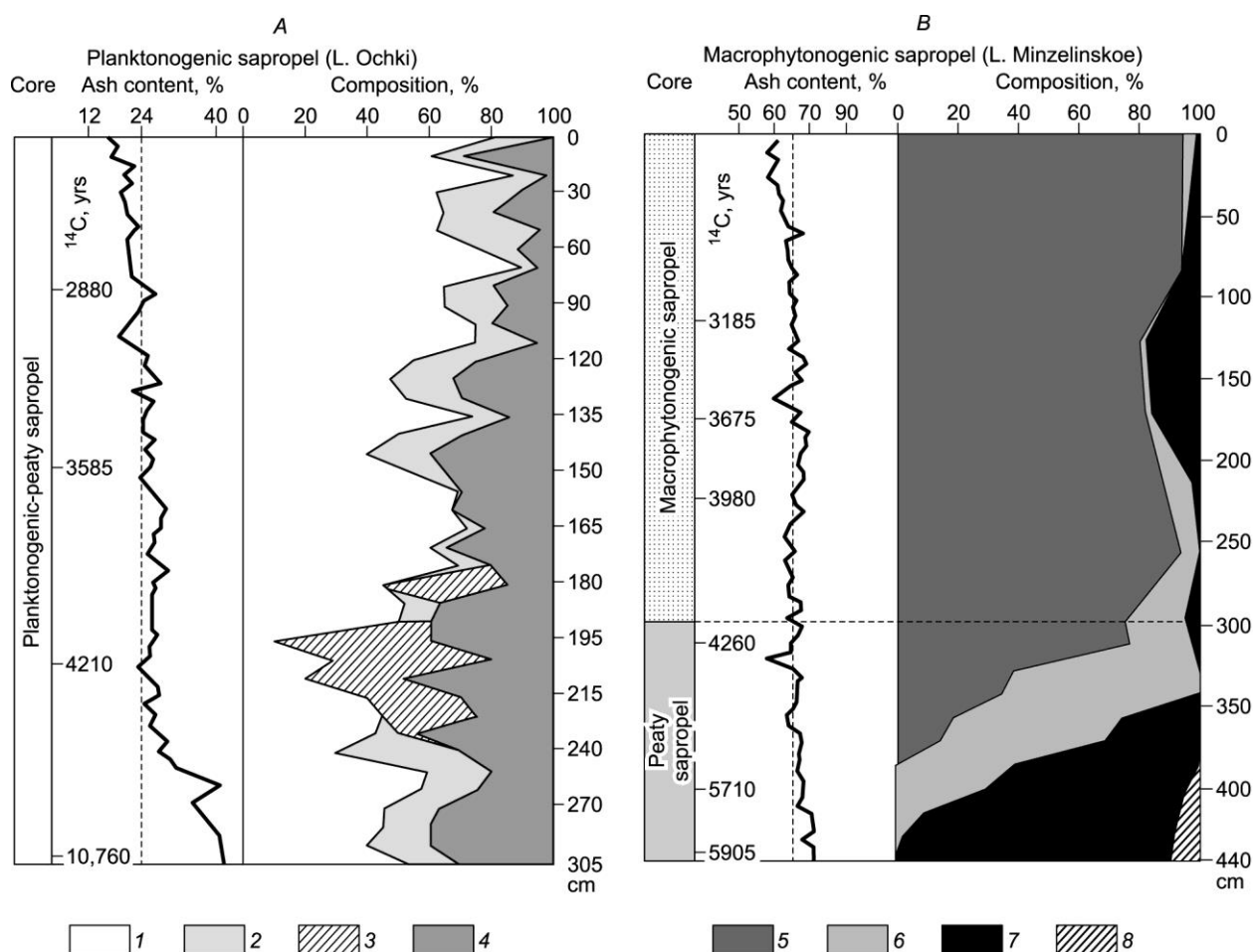


**Fig. 1.** Schematic location map of lakes of the south of Western Siberia (Bol'shie Toroki, Minzelinskoe) and lakes in the East Baikal Region (Kotokel', Dukhovoe, and Ochki). Locations of boreholes are shown below.

For sapropel lakes, high rates of sediment accumulation, organogenic deposits reflect well the biogeochemical processes; they contain a lot of paleontological material for purposes of biostratification, in addition dated by the radiocarbon method. This study is based on the material collected by authors during field seasons of 2011–2013. Long cores of lacustrine sediments with undisturbed stratification were obtained as a result of vibratory drilling of the lake bottom to the underlying rocks: Bol'shie Toroki (1.8 m), Minzelinskoe (5 m), Ochki (4.5 m), Dukhovoe (7 m), and Kotokel' (14 m). Drilling of lacustrine sediments was carried out by standard methods, but with the help of equipment created in Sobolev Institute of Geology and Mineralogy, SB RAS on the initiative of Dr. A.K. Krivonogov. This equipment has been repeatedly tested in many lakes. The drilling rig is compact, sectional, motor-borne, consists of a pneumatic pontoon with displacement tonnage about 5 tons, derricks with elevated mechanisms, drill rod set of total length 30 m. The vibration principle is applied: the modified Livingston piston drill allowing columns of the undisturbed sediment 2 m in length and 7.5 cm in diameter to be taken. The applied vibration drilling technology makes it possible to uncover the entire lacustrine sediments and to penetrate underlying rocks by 15–20 m [7]. Study of the entire Holocene sequences of deposits gives a better insight into conditions of a lacustrine sedimentogenesis throughout the whole period of the Holocene and Late glaciation, as well as the probable change of organic matter sources of organic matter inputs, and, consequently, different organic matter genesis in stratified layers of sedimentary strata.

***Biostratification of sapropel deposits.*** For study in detail of supply sources and genesis of buried (fossilized) organic matter in detail throughout the depth of stratified sequences, the following lakes have been selected: Lake Ochki in the Baikal Region and West Siberian Lake Minzelinskoe. The direct method of quantitative layer-by-layer calculation of preserved remains of organisms has been selected as a priority method according to the Korde approach (Korde, 1960, 1968), and based on the obtained results of biological analysis a biostratification of deposits of these lakes has been given.

***Lake Ochki.*** The biostratification of the sapropel Holocene sequence of Lake Ochki (Fig. 2A) revealed heterogenic origin of organic matter: remains of phyto- and zooplankton as autochthonous source of organic matter, and humus flakes, remains of green algae (*Drepanocladus*), and Sphagnum as allochthonous organic matter source. The amount of plankton residues in the upper sequence part (0–190 cm) varies within 90–40% with the tendency of their increase in depth. In the layer 195 cm, their minimum (10%) is marked. In the lower part of deposits (200–305 cm) there is a gradual increase in the content of plankton residues to 40–60%.



**Fig. 2.** Biostratification of Holocene sapropel sequences of Lakes Ochki (A) and Minzelinskoe (B) according to the data of complex biological analysis performed by T.A. Kopoteva (Institute of Water Problems, FEB RAS). 1, plankton; 2, green mosses; 3, sphagnum mosses; 4, humus flakes; 5, macrophytes submerged; 6, hypnum mosses; 7, macrophytes semisubmerged (helophytes); 8, mollusk shells.

Of three components attributed to allochthonous organic matter, the most representative are humus flakes, the content of which tends to increase in the sequence depth. So, if the amount of humus flakes in the upper one-meter sediments rarely exceeds 20%, then it reaches 40–50% in horizons of the lower part of the sequence. The humus substance is rough due to its enrichment with residues of tissues of vascular peatforming plants (cotton grass and sedges). Remains of mossy vegetation almost always take part in formation of lacustrine sediments. Nevertheless, the attention is drawn to an interval of bottom sediments (180–230 cm), in which, unlike the overlying and underlying layers, remains of not green, but Sphagnum moss are found in significant quantity (up to 50%). Pollen of coniferous trees is sporadically met throughout the whole core of sediment. Thus, during 10,760 years, planktogenous sapropel (autochthonous source of organic matter) was predominantly formed in Lake Ochki, and from marshy shores, residues of tissues of green mosses, Sphagnum, and humus substances were supplied into lacustrine sediments (allochthonous organic matter source).

**Lake Minzelinskoe.** The biostratification of the Holocene sequence in Lake Minzelinskoe (Fig. 2B) also points to the heterogeneous origin of organic matter: remains of submerged

(hydrilla, water soldier) and semisubmerged (typha, bulrush) macrophytes (autochthonous organic matter source), remains of green mosses (allochthonous organic matter source). The upper strata of sediments up to the horizon 300 cm are represented by macrophytogeneus sapropel, which under a microscope looks like structureless of amorphous gray mass. Taken shape remains of submerged macrophytes (so-called “soft” aquatic vegetation: hydrilla and water soldier, are not preserved in the sapropel sequence and these are represented by amorphous detritus (to 80%). On the contrary, remains of “hard” aquatic vegetation (semisubmerged macrophytes: (typha, bulrush)) are preserved and vary within 2–7%. Aggregates of benthonic diatoms such as *Pinnularia* are present.

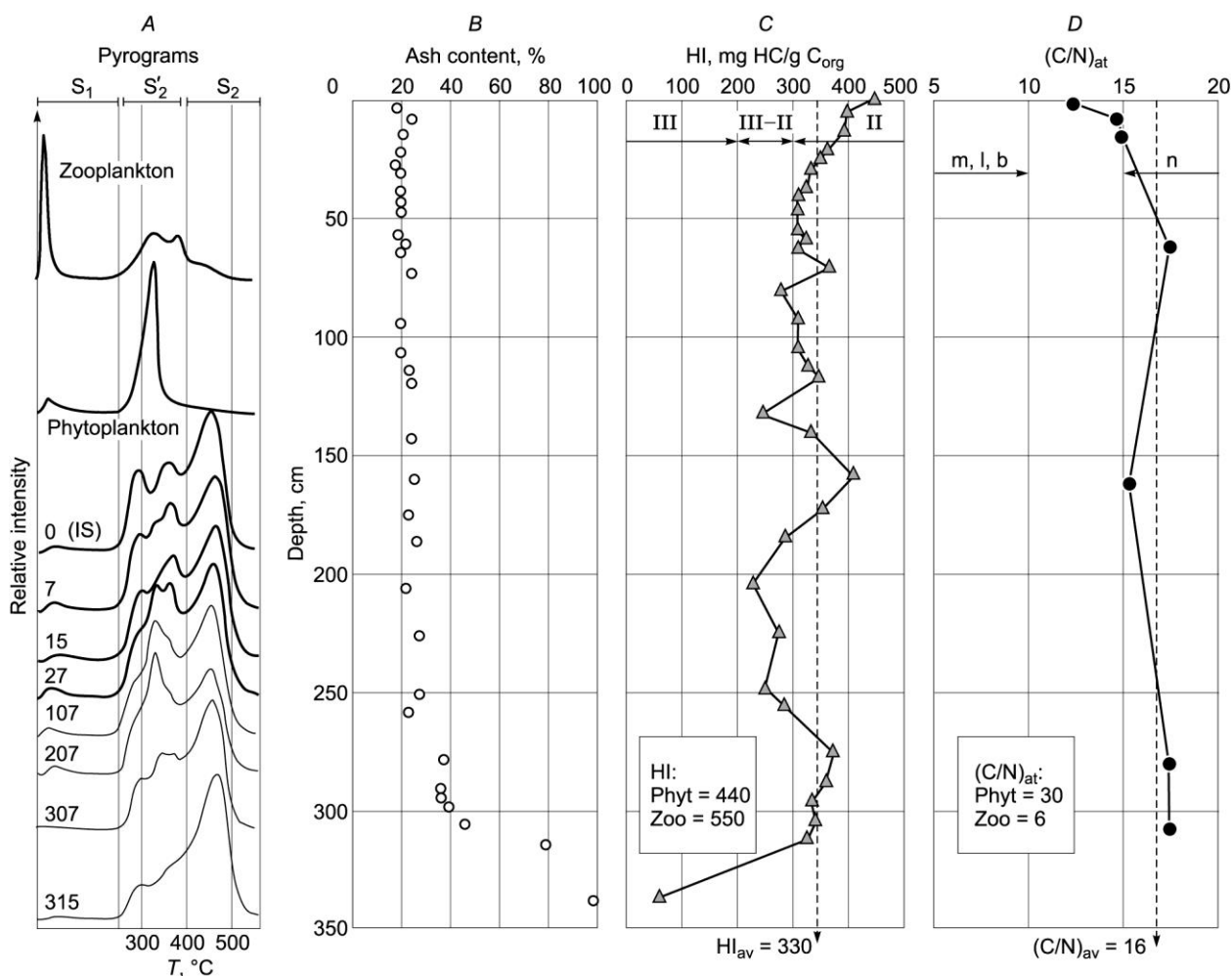
From the horizon 300 cm, macrophytogeneus sapropel is underlain by peaty (hypnaceous) sapropel of chocolate brown color of the high decomposition degree (to 50% and more). The fraction of remains of hypnum (green) mosses (*Drepanocladus aduncus*) is gradually increases from 15% in the horizon 300 cm to 50–60% in the horizon 335 cm. Within the core interval 335–360 cm, the peaty sapropel is almost black and has a high degree of decomposition (about 70%). Macrophyte remains (thipa, water soldier) are found in amount 60–70%, and hypnum mosses are found in amount 40–30%. In the core interval 360–366 cm, the peaty sapropel has a very high decomposition degree (85–90%).

There are many fragments of mollusk shells of the Planorbidae from 0.8–1.0 mm to 2.5–3.0 mm in diameter, as well shells of small bivalve mollusks (*Bivalvia*). The fraction of hypnum moss remains is 30%. Within the core interval from 396 to 440 cm, the decomposition degree of sapropel varies from 90 to 99%. Their composition is dominated by remains of semisubmerged macrophytes such as *Typha* and more rarely bulrush, remains of hypnum mosses are singular, and shell fragments are abundant.

It is assumed that swamping began in the time period dated at 5905 years, in the southwestern part of Lake Minzelinskoe, which is currently a shallow bay with flooded shores. The high-ash mass of sapropel rich in humus (interval 440–420 cm) and saturated with mollusk shells indicates that, seemingly, it was a shallow water body heavily overgrown with macrophytes. Then, formation of heavily watered lowland bog began along with accumulation of peaty hypnum sapropel with predominant *Drepanocladus aduncus*. In the subsequent time periods (3980 and 3185 years), due to rise of water level in the lake, a lowland march was flooded and the shallow bay formed with predominance in the capacity of the main sources of autochthonous organic matter of submerged macrophytes and, to a lesser degree phyto- and zooplankton. The process of formation of macrophytogeneus sapropel in the lake continues also in the present time.

Thus, according to the data of complex biological analysis and based on the biostratification of the Holocene sequences of Lakes Ochki and Minzelinskoe, sources of input and genesis of buried organic matter of two biological types of sapropels (planktonogenic and macrophytogeneous) have been revealed, which are confirmed by the data of material organic matter composition obtained with using scanning electron microscope (SEM).

**Data of pyrolytic analysis of organic matter.** In pyrograms of sediments (Fig. 3A), beginning from incoherent sediment (IS) the predominance of a high-temperature “kerogen” peak is clearly seen at  $T_{\text{peak}} \approx 460^\circ\text{C}$  to the depth of 307 cm. The fraction of hydrocarbons (HC), released at this peak relative to their total content remains practically constant to the depth of 279 cm and then it increases downward. This growth corresponds to decrease of labile, less stable compounds in the organic matter composition and, consequently, to increase of the degree of its diagenetic transformation.



**Fig. 3.** Pyrograms of zooplankton, phytoplankton, and samples of sediment from Lake Ochki (A). Figures in the pyrograms correspond to the depths (cm) of analyzed sediment samples. IS, incoherent sediment. Along the y-axis the intensity of a signal of flame-ionization detector normalized at temperature of the maximum release rate of hydrocarbons (HC). Given in graphs are: the values of ash contents (B), hydrogen index (HI) (C), and atomic ratio  $(C/N)_{\text{at}}$  for phytoplankton and zooplankton (D). HI was calculated by the formula  $HI = \Sigma HC \times 100 / C_{\text{org}}$ , where  $\Sigma HC$  is the total amount of pyrolytic hydrocarbons ( $S_1 + S'_2 + S_2$ ) (mg HC/g of sediment);  $(C/N)_{\text{at}}$  has been calculated from the data of element analysis for HC of marine (m), lacustrine (l), and bacterial (b) genesis and land plants (p), respectively.

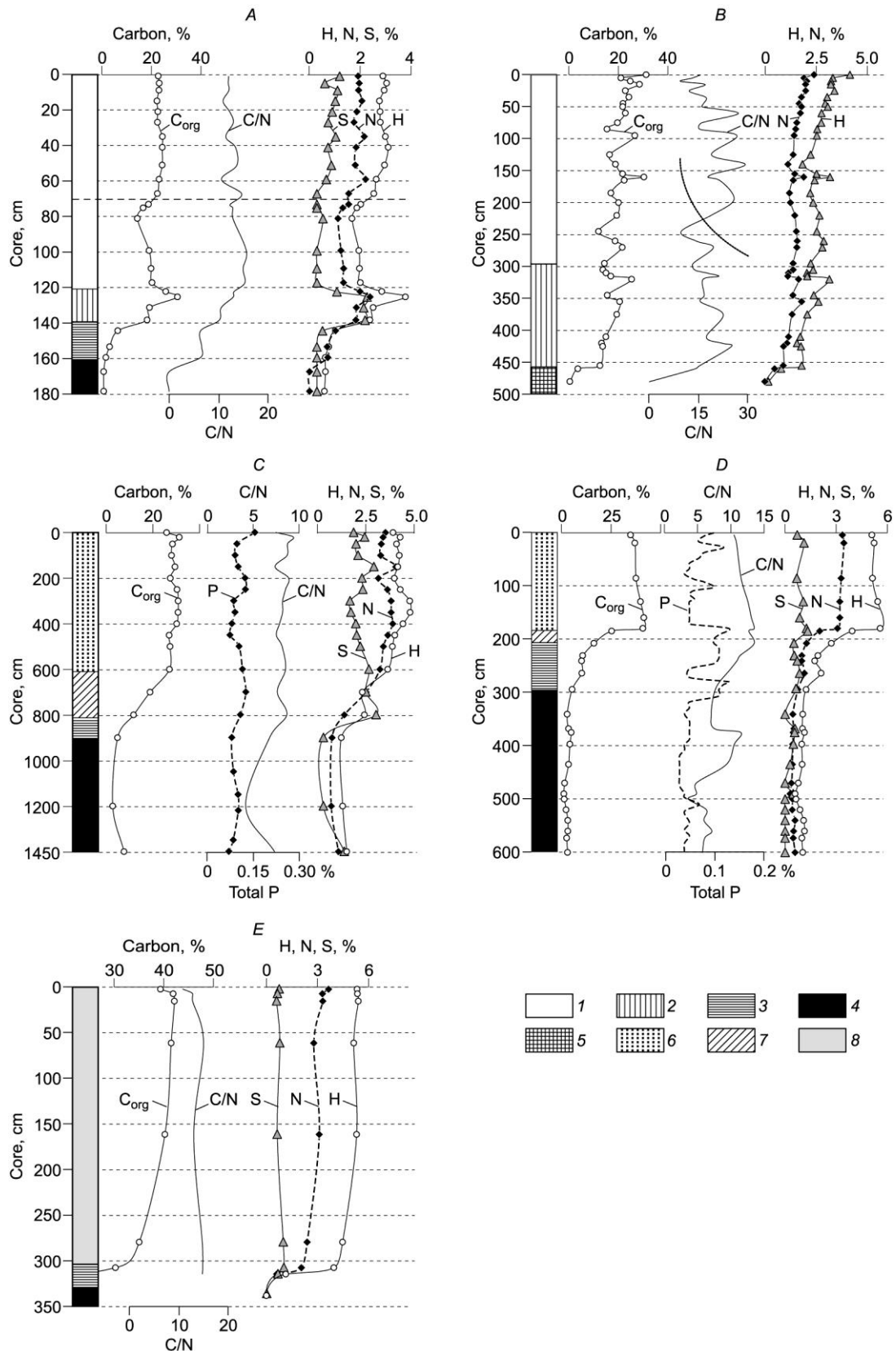
The peculiarity of pyrolysis of bioproducts and organic matter of sediments of Lake Ochki (Fig. 3A) is in the sharp dissimilarity of pyrograms of bioproducts and organic matter from pyrograms of bottom sediments: if the thermal decomposition for the former occurs mainly to 400°C (peaks S<sub>1</sub> and S'<sub>2</sub>), then the temperature of the maximum decomposition rate for the latter is shifted to higher temperatures. This indicates that it is already at the initial stage of diagenesis in the incoherent sediment during microbial treatment of biomacromolecules and they are transformed into geomacromolecules of kerogen. The content of organic matter is rather constant within the interval of sediments 0–250 cm and is about 80, the ash content is, correspondingly, 20% (Fig. 3B), however, the data of pyrolysis and elemental analysis demonstrate qualitative changes of organic matter (Fig. 3C, D). It is known (Tissot and Welte, 1978), that the value of the hydrogen index (HI) of organic matter are defined by the content of hydrogen in its composition: the more hydrogen, and, consequently, the higher the H/C ratio is, and the higher is the HI value, characterizing, in such a way, oxidation/reduction of organic matter. The HI and (C/N) at values for phyto- and zooplankton, as well as their mean values for the sediment are given (in Fig. 3C and 3D). The most oxidized organic matter is observed at the base of the sapropel strata (339 cm). This may be due to both the oxidative sedimentation conditions and the oxidation processes during early diagenesis, for example, owing to contacts with aerated groundwaters [9].

**The C/N ratio as a marker of sources of autochthonous and allochthonous organic matter.** According to the data [3] with the  $C/N \leq 12$  ratio, organic matter of the autochthonous origin predominate in bottom sediments of small lakes over allochthonous organic matter in its proportion from 12 to 40–47. In other publications [5], the authors place emphasis on the fact that in small lakes, the influence of the input factor of allochthonous organic matter into common organic matter has an impact on the whole bottom area as a consequence of this C/N value is higher than 10 in sediments of small lakes. The distribution of C/N values within stratified sapropel sequences of typical macrophyte Lakes Bol'shie Toroki and Minzelinskoe (Fig. 4A, B) as compared with typical phytoplankton Lakes Kotokel', Dukhovoe, and Ochki (Fig. 4B, D, E) are significantly different.

In Lake Bol'shie Toroki and Minzelinskoe, the C/N values vary in the sequences of macrophytogeneus sapropel from 10 to 15. In layers of peaty sapropel, the C/N values are slightly greater, and they vary within the range 15–18. In general, for the whole sapropel strata of Lakes Bol'shie Toroki and Minzelinskoe, the C/N values fall into the range of C/N values for organic matter of just as aquatic (submerged and semisubmerged macrophytes), so of terrestrial vegetation (sedges, cotton grass, and mosses). As a whole, the C/N values in the sapropel sequence of Lake Minzelinskoe are higher than those in Lake Bol'shie Toroki. This can be



explained by heterogeneity of organic matter of sapropel from Lake Minzelinskoe in which, according to the data of biological analysis, the share of allochthonous organic matter (remains of hypnum moss) is rather high.



**Fig. 4.** Vertical distribution profiles of organic carbon ( $C_{org}$ ), H, N, S, P, and C/N in the Holocene sediments of Lakes Bol'shie Toroki (A), Minzelinskoe (B), Kotokel' (C), Dukhovoe (D), and Ochki (E). 1, Macrophytogenic

sapropel; 2, peaty sapropel; 3, organic-mineral sediment; 4, underlying clays; 5, underlying sands; 6, planktonogenic sapropel; 7, high ash planktonogenic sapropel; 8, sapropel of mixed genesis (planktonogenic-peaty)

As a whole, the C/N values in the sapropel sequence of Lake Minzelinskoe are higher than those in Lake Bol'shie Toroki. This can be explained by heterogeneity of organic matter of sapropel from Lake Minzelinskoe in which, according to the data of biological analysis, the share of allochthonous organic matter (remains of hypnum moss) is rather high. The shores of this lake are heavily swamped in contrast to shores of Lake Bol'shie Toroki. Lake Kotokel' is characterized by the lowest C/N values, which vary in depth of the 8-m sapropel sequence from 7.4 to 9.3 (average over the sequence is 8.2), that falls into the range of C/N values for marine and lacustrine plankton. In Lake Dukhovoe, the C/N values vary in the 180 cm sequence of planktonogenic sapropel from 5.7 to 8.6, falling into the range of C/N values for marine and lacustrine plankton. Thus, it can be concluded that the main source of sapropel organic matter of Lakes Kotokel' and Dukhovoe is phytoplankton. In Lake Ochki, the C/N values in the depth of the sapropel sequence (0–310 cm) vary within 0.8–14.9, on the average 13.6. Quite high C/N values are explained by the material composition of sapropel: mixed (along autochthonous organic matter formed mainly by phytoplankton, the share of allochthonous organic matter, represented by remains of moss is also high). Thereby, in studied typical macrophyte lakes in the south of the West Siberia (Bol'shie Toroki and Minzelinskoe), in sapropel sequences, high content of organic matter of allochthonous origin has been established (the average values are  $C/N \geq 12$ ), the source of which are mainly semisubmerged macrophytes (bulrush, thipa). In typical phytoplankton lakes of the eastern Baikal Region (Kotokel', Dukhovoe, and Ochki), organic matter of mainly autochthonous origin ( $C/N < 12$ ) predominate, with phytoplankton as the source. When using this organic-geochemical index of the contribution of autochthonous and allochthonous material into the composition of bottom sediments (C/N), it should be understood that it is approximate due to the uncertainty of the degree of diagenetic transformation of organic matter.

### **Conclusions**

West Siberian Lakes Bol'shie Toroki and Minzelinskoe in which the main role in formation of organic matter in recent biocoenoses belong to aquatic higher plants (macrophytes), are assigned to the typical macrophyte lakes. In the lakes of the Baikal Region: Dukhovoe and Kotokel', organic matter formation in recent biocoenoses was mainly due to microscopic algae of phytoplankton. In Lake Ochki, the main role is played by phytoplankton during synthesis of autochthonous organic matter; the contribution of also allochthonous organic matter source such as sphagnum and hypnum mosses is significant.

Having studied the problem of sources and genesis of organic matter buried in stratified sequences of lacustrine sediments, it is advisable to use the proposed complex of organic-geochemical indicators for more reliable conclusions.

Having studied the problem of sources and genesis of organic matter buried in stratified sequences of lacustrine sediments, it is advisable to use the proposed complex of organic-geochemical indicators for more reliable conclusions. As a priority, we propose to apply the direct method of quantitative layer-by-layer calculation of preserved remains of organisms and based on the data of biological analysis to give the structure of a bog body and biostratification of sediments. The authors first represent the biostratification of sapropel deposits of Lake Ochki (the Baikal region) and Lake Minzelskoe (south of West Siberia).

The molecular composition of normal aliphatic hydrocarbons (*n*-alkanes) and components of protein-carbohydrate complex is recommended to be used as biomarkers of sources and genesis of buried organic matter, which are defined as complex molecular “imprint” of previously living organism formed from biochemical components. Investigation of bioproducts and sapropel from Lake Ochki by pyrolytic methods in versions RE-pyrolysis has shown that the main autochthonous organic matter source in the lake was phyto- and zooplankton.

The ratio organic carbon/organic nitrogen (C/N) is recommended for use for approximate estimation (due to uncertainty of the degree of diagenetic organic matter transformation) of the proportion of organic matter shares of the terrigenous (allochthonous) and aquagenic (autochthonous) genesis in lacustrine sediments.

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