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Methodological Approach to the Identification of the Sources and Genesis of Buried Organic Matter in Holocene Sections of Lake Sapropels (Southern West Siberia and Eastern Baikal Area)[1]

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Abstract—We present results of study of the chemical composition of organic matter (C, H, N, and S) from Holocene sections of lake sapropels with undisturbed stratification penetrated by vibratory drilling of the bottom sediments, down to the underlying rocks, of Lakes Bol'shie Toroki (1.8 m), Minzelinskoe (5 m), Ochki (4.5 m), Dukhovoe (7 m), and Kotokel' (6 m). We consider methodological approaches to the identification of the sources and genesis of buried organic matter in marine and lacustrine sediments by a number of organo-geochemical 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. Pyrolytic study (RE pyrolysis and Pyr–GC–MS analysis) of bioproducers and sapropel from Lake Ochki has shown that phytoplankton and zooplankton were the main autochthonous source of OM in the lake, which is confirmed by the identified macromolecules of nitrogen compounds of different compositions. The presence of hopanes indicates the contribution of microorganisms to the formation of OM in the sediments. The organic matter of sapropel has a terrigenous component, which is confirmed by the presence of high-molecular odd-numbered *n*-alkanes, ketones, and methyl esters of fatty acid. The C/N ratios in the stratified sections of macrophytogenic sapropels of Lakes Bol'shie Toroki and Minzelinskoe fall in the range of values specific to higher aquatic and terrestrial vegetation (C/N = 15–18), whereas the C/N ratios in planktonogenic sapropel of Lake Dukhovoe are typical of marine and lake plankton (C/N = 5.7–8.6).

Keywords: lake sapropel, genesis of organic matter, bioproducers, hydrocarbon biomarkers, biostratification, authohtonous source of organic matter, diagenetic transformation of organic matter

INTRODUCTION

Revealing sources of supply and genesis of buried (fossilized) organic matter (OM) in depths of stratified sequences of lacustrine sapropel is an extremely challenging task, which requires an integrated approach with the involvement of special methods of analysis and criteria (genesis indicators) indirectly conforming the genetic link of buried OM with initial organic matter (autochthonous matter such as aquatic organisms or allochthonous one, i.e., terrestrial vegetation) (Leonova et al., 2018a).

It is necessary to distinguish the methods for determination of recent organic matter supply into bottom sediments of lakes and those of sources of organic matter buried in a sapropel strata formed during the Holocene period (11 ka) and late glacial period (11.7–15 ka). The methods for determination of recent OM sources are relatively simple: recent biocoenoses dominated by species of producers such as phytoplankton, or highest terrestrial vegetation (macrophytes) are revealed (Zarubina, 2013; Ermolaeva et al., 2016). The content of carbon of organic matter (C_{org}) in vertical profiles of bottom sediments characterizes the proportion of carbon buried at different depths during the whole historical period of sedimentary strata formation (Lisitzin, 1955; Vetrov and Romankevich, 2008; Romankevich et al., 2009). However,

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the variation of C_{org} in vertical profiles of bottom sediments is complicated by uncertainty of the degree of diagenetic transformation of organic matter losing a part of labile compounds due to their different resistance to decomposition (Vetrov et al., 2008; Melenevskii et al., 2011, 2015, 2017; Leonova et al., 2018b, 2019).

In water bodies of final runoff (inland seas and oceans), OM of plankton detritus is remineralized on the way to bottom sediment due to great depths (in seas) or almost completely (in oceans) (Lisitzin, 2004). In contrast to this fact, OM in shallow marginal seas, coming from land, as well as newly formed OM, rapidly reaches sea bottom, and the main diagenetic transformation (microbial OM destruction) occurs in the upper layer of sediments and on their surface (Vetrov et al., 2008; Lein et al., 2011; Belyaev, 2015). In small shallow lakes, OM of detritus is not completely destroyed as it is sinking to the bottom, that provides, under certain conditions, formation of significant strata of lacustrine organogenic deposits (sapropels) (Korde, 1960, 1968; Leonova and Bobrov, 2012; Leonova et al., 2015). In sapropels enriched with organic matter of small lakes, diagenetic transformation is extended to almost the entire depth of multimeter sedimentary strata. During this process initial OM both autochthonous and allochthonous is transformed, losing some labile compounds.

The aim of this study is to reveal sources and genesis of organic matter buried in the stratified Holocene sequences of sapropel deposits of some small lakes in the Siberian Region by a complex of organic-geochemical indicators: (1) data of biological analysis (biostratification based on layer-by-layer determination of preserved remains of organisms in sequences of lacustrine sapropels in sequences); (2) by application of hydrocarbon biomarkers (molecular composition of normal aliphatic hydrocarbons (*n*-alkanes)) and components of protein-carbohydrate complex; (3) by the C_{org}/N_{org} ratio reflecting differences in biochemical composition of producer organisms.

REVIEW OF METHODOLOGICAL APPROACHES TO REVEALING SOURCES AND GENESIS OF ORGANIC MATTER OF MARINE AND LACUSTRINE SEDIMENTS

Methodological approach to revealing sources and genesis of organic matter in marine sediments. Organic material buried in bottom sediments is a reflection of the record of past age conditions of deposition and sources of OM. Organic material is formed in marine environment from autochthonous sources, mainly at the expense of primary phytoplankton production (Vetrov and Romankevich, 2008; Romankevich et al., 2009). The considerable contribution of allochthonous material is represented by terrigenous erosion discharge and river run-off (Gordeev and Rachold, 2004; Vonk et al., 2012). For the most effective revelation of OM sources, biochemical reconstruction of the original organic substance is used with the help of biomarkers, which, in one of key foreign research articles summarizing multiyear studies of biomarkers (Peters et al., 2008) are defined as complex molecular "imprints" of previous organisms, formed from biochemical components.

Revealing sources and genesis of OM in marine sediments is performed by the complex of organic-geochemical indicators: normal aliphatic hydrocarbons (*n*-alkanes), isotope composition of carbon $\delta^{13}C$ and applying the ratio of organic carbon to organic nitrogen (C/N). Quantitative estimation of the proportion of terrigenous and marine OM in bottom sediments by organic-geochemical indicators is complicated by the diversity of composition of the initial OM and uncertainty of the degree of its diagenetic transformation (Semiletov et al., 2005; Vetrov et al., 2008; Lein et al., 2011; Belyaev, 2015; Melenevskii et al., 2017; Gershelis, 2018).

Molecular composition of normal aliphatic hydrocarbons (n-alkanes) and their molecular ratios are the most widely useable molecular biomarker reflecting sources, genesis and OM transformation during the diagenesis process (Vetrov et al., 2008; Romankevich et al., 2009; Belyaev, 2015; Melenevskii et al., 2017). Molecular-mass distribution of *n*-alkanes is individual for different types and it is used as a tool for recreation of sedimentation conditions. Thus, for example, long-chain odd *n*-alkanes with maxima ascribed to C_{23}, C_{25}, C_{27}, C_{29}, C_{31}, C_{33}, and $C_{35,}$ contained in wax coatings of plants, point to dominating contribution of higher vegetation and these can serve as biomarkers of terrigenous OM including both recent and ancient OM of abrasion products of ancient rocks. Marine algae produce mainly low-molecular weight *n*-alkanes (to *n*-C_{17}) of smoothed distribution, i.e., without obvious predominance of even and odd structures. Low-molecular homologues of hydrocarbons C_{12}–C_{19} are peculiar to hydrobionts and planktonogenic OM. The smoothed distribution of *n*-alkanes within the range from *n*-C_{15} to *n*-C_{28} (Vetrov et al., 2008; Melenevskii et al., 2017; Gershelis, 2018) is also peculiar to bacteria.

The molecular ratio of *n*-alkanes (the ratio of the sum of odd to the sum of even *n*-alkanes)—the odd/even ratio (CPI) is also an indicator of the OM genesis: high CPI values indicate the terrigenous genesis of OM and point to the initial stage of diagenetic transformations under oxidative conditions that is typical for shallow depths of shelf seas (Vetrov et al., 2008; Melenevskii et al., 2017; Gershelis, 2018).

Values of hydrogen (HI) and oxygen (OI) indices mark sources of primary OM (aquatic or terrigenous). Thus, the values HI < 100 mg/g, along with a high oxygen index OI indicate predominantly terrigenous, deeply oxidized OM (humus substance), while high values of HI (from 300 to 800) are typical for marine genesis of OM and predominance of reductive conditions (sapropel substance). High values of the hydrogen index HI are the indicator of marine (autochthonous) nature of OM (Tissot and Welte, 1978; Melenevsii et al., 2017).

Carbon ($\delta^{13}C$) isotope composition is also an indicator of OM genesis. Lighter composition of carbon is characteristic for terrigenous OM: sediments of terrigenous origin have $\delta^{13}C$ = –26–28‰. Organic carbon of marine origin contains large amount of the isotope ^{13}C due to worse conditions of fractionation of carbon isotopes by algae during photosynthesis (Galimov, 1968; Lein et al., 2011; Vonk et al., 2012). According to (Semiletov et al., 2005), the isotope composition of carbon $\delta^{13}C$ from the upper layer of sediments of the East Siberian Sea varies from –22.9‰ on the eastern shelf, where highly productive waters of Pacific origin penetrate, to –27‰ in regions of thermoabrasive coasts that indicates its mixed genesis with the prevalence of a terrigenous source. The isotope composition of carbon ($\delta^{13}C$) was used in (Gershelis, 2018) to clarify the contribution of various sources into the composition of organic carbon of bottom sediments from seas of Eastern Africa: the values of $\delta^{13}C$ varied within the range –27.4‰ and –20.85‰ demonstrating the trend of sediment enrichment with stable carbon isotope with increasing distance from the shoreline and increasing influence of autochthonous (marine) OM component. This made it possible to reveal, in general, the tendency of a relatively uniform substitution of terrigenous organic carbon by marine substance with distance from the coastline with respect of increase of the fraction of the heavy C_{org} isotope. The values established for studied sediments indicate the marine origin of OM ($\delta^{13}C$ = – 24 ± 3.0‰).

The ratio organic carbon/organic nitrogen (C/N) is often used for approximate (due to uncertainty of the degree of diagenetic OM transformation) estimation of the relation between sediments of terrigenous (allochthonous) and marine (autochthonous) genesis. The C/N ratio reflects the proportion of nitrogen-rich and depleted in nitrogen terrigenous substances in suspension and marine sediments. The data available in literature on marine sediments indicate an increase in the C/N ratio in the deposits with depth, which evidences disintegration of OM with advance decomposition of nitrogen-containing organic compounds (Starikova, 1956; Bordovskiy, 1964). The mean C/N ratio in suspended OM (mainly represented by plankton) from the White Sea was 7.7 according to (Belyaev, 2015), Marine plankton according to (Vetrov et al., 2008) is characterized by C/N ratio ~6÷7 and it can be used in estimations of C/N_{mar}. However, it should be taken into account that nitrogen-rich proteins of plankton pertain to easily hydrolysable compounds and the C/N ratio changes during the process of diagenesis. In order to estimate the fraction of terrigenous OM in marine sediments applying the C/N ratio the linear equation is used (Bordowskiy, 1965): OM_{ter} (%) = $(C/N_{sampl} - C/N_{mar})/(C/N_{ter} - C/N_{mar})$. Terrigenous OM may include recent organic matter, as well as organic matter subjected, in varying degree, to transformation, on the way to marine environment, and OM of abrasion products of ancient rocks including easy decomposable permafrost rocks. Usually C/N_{ter} are approximately estimated by values >15, according to (Bordowskiy, 1965). According to the data (Stein and Macdonald, 2004), the C/N ratio of terrigenous sediments in the Arctic basin varies within the range 10–20. The C/N ratio in superficial deposits of the East Siberian Sea varies from 6.5 to 14.7, more than 80% of samples have C/N > 9, that evidences the mixed genesis of OM with predominance of the terrigenous one (Vetrov et al., 2008).

Methodical approach to revealing sources and genesis of buried organic matter in the stratified Holocene sequences of lacustrine sapropels. Revealing sources and genesis of lacustrine sapropels of OM buried 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 (Korde, 1960, 1968). The data of the integrated biological analysis allow us to judge not only about sources of buried OM, 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 OM 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) and oxygen (OI) indices, isotope composition of carbon $\delta^{13}C$, and the ratio C_{org}/N_{org} (Leonova et al., 2018a).

Hydrocarbon biomarkers. The traditional method for study of molecular OM 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 OM in samples of soils and recent lacustrine sediments (Disnar et al., 2003). This is based on the fact that pyrolysis of OM sample of recent sediment may be represented as a complex process comprised of a series of consecutive stages of decomposition of some OM 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 OM components. The method of deconvolution (factorization) of some experimental OM pyrograms into pyrograms of separate OM components proposed in (Sebag et al., 2006) allows the contribution of each OM component in recent sediments to be estimated quantitatively.

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 (Meyers, 2003; McKirdy et al., 2010).

Reliable identification of OM source (primary bioproducers) can be performed only applying a complex interpretation of several organic-geochemical criteria: (1) composition of *normal aliphatic hydrocarbons (n-alkanes)* including the ratio of homologues with even and odd numbers of carbon atoms in the molecule; (2) oddness index (CPI), high values of which ($\gg 2$) indicate weak genetic transformation of OM, and therefore, marking a permanent input of fresh OM, not aquagenic, but of terrestrial (terrigenous) origin into sediments; (3) Pr/Ph (the interrelation of isoprenids with *n*-alkanes), increased values of which confirm preferentially oxidative environment of sedimentation, i.e., significant contribution of terrestrial (terrigenous) OM (Melenevskii et al., 2011, 2015).

One of the most important relict hydrocarbons retaining their biochemical structure are *hydrocarbons of the hopane series*. In immature OM of recent sediments, biohopanes (products of diagenetic chain biosynthesis of hopanoid of bacteriohopanepolyol transformation) predominate. In immature OM 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 (Blumenberg et al., 2006). *Steranes*, saturated tetracyclic hydrocarbons C_{27}–C_{35}, along with hopanes, are also the most important relict biologically marking hydrocarbons (Petrov, 1984).

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 (Vinogradov, 1938). It is known that higher terrestrial plants are depleted in nitrogen and have a high C/N (20–40) value (Skopintsev, 1950). This ratio for diatomic plankton is 5.5–7.0 (Harvey, 1945) and for Baikal phytoplankton (*Melosira baicalensis*) it is close to 10 (Votintsev, 1961). 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) (Veber et al., 1956).

The C/N ratio not only points to a genetic link of OM sediments with initial organic material, but also reflects the degree of its mineralization during the transformation process of sediments at the stage of early diagenesis. It is observed in Lake Baikal both as an increase in the C/N ratio downsection (in sediment columns from shallow coastal bays in the northern Baikal), and as a decrease from top to bottom in sediment columns of the middle and southern Baikal (the latter is likely due to accumulation of the most persistent nitrogen-containing organic matter (Vykhristyuk and Lazo, 1972; Vykhristyuk, 1980).

Surface sediments of the most part of the northern and middle basins of Lake Baikal have C/N values close to those in phytoplankton, which indicates the formation of organic matter in open areas of the Northern and Middle Baikal mainly at the expense of plankton organisms and, foremost, phytoplankton (Vykhristyuk, 1980). Planktonogenic genesis of OM from deep-water Baikal sediments is also confirmed in (Votintsev, 1967; Tarasova, 1971). Increased C/N values (from 12 to 16) were denoted in bottom sediments of Lake Baikal foreshore areas: in all appearance, the great role belongs to allochthonous organic vegetative detritus supplied with waters of tributaries and overland runoff (Vykhristyuk, 1980). In small lakes, influence of the input factor of allochthonous organic remains on the composition of organic matter has an impact on the whole lake bottom area. The C/N values in the surface layer of sediments of small lakes is higher than 10 (Koyama, 1954; Wieckowski, 1969).

SUBJECTS AND METHODS 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). 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.

Lake Bol'shie Toroki (55°25′ N, 80°36′ E) is situated in the forest-steppe zone of the Novosibirsk Region (Kargat District) within the East Baraba lowland plain (Fig. 1). This is a typical lacustrine alluvial plain where there are no bedrock outcrops since those are buried under a layer of sedimentary rocks of various ages: from Precambrian to Quaternary. The Quaternary part of the geological sequence includes sediments of different facies. The lower horizons are represented by the Kochki Formation made up of very dense clay marls and clay loams, and inequigranular sands of lacustrine and of subaerial genesis. Interlayers of buried soils are found in clays. The upper part of the Quaternary sediments is represented by the Fedosovo Formation of Barbara, made up of lacustrine-alluvial loam and clays (Krasnov, 1984). The presence of buried soils and the clay composition of sediments allows the significant part of the Quaternary sequence to be referred to the loess-soil series of sediments (Zykina et al., 1981). Loess was accumulated during cold stages of glaciation whereas soils were formed during warm interglacial periods. Subaerial cover loess-like sediments of the last glaciation are distributed everywhere on the Ob'–Irtysh interfluve, except of the first river terrace and flood plain, and these form a layer with the thickness of 5–6 m (Volkov et al., 1969; Volkov, 2003).

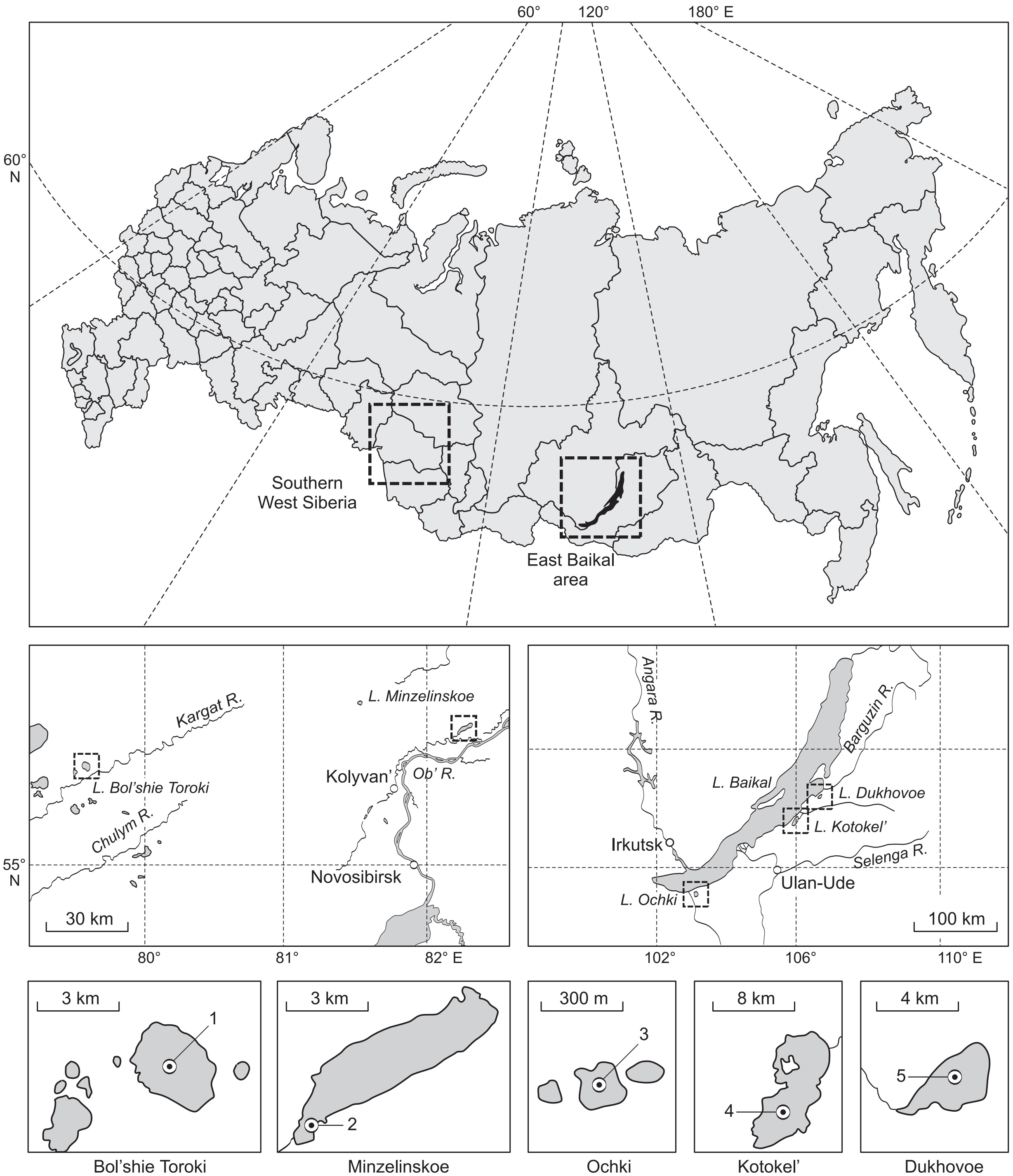

Fig. 1. Schematic location map of lakes of the south of Western Siberia (Bol'shie Toroki, Minzelinskoe) and lakes in the East Baikal Region (Kotokel', Dukhovoe, and Ochki). Locations of boreholes are shown below.

The lake is of relict origin and it represents a postglacial water body, its basin is flat and flanks are gently sloping. The lake length is 4.2 km, its width is 2.9 km, and its depth is 0.5 m, the water area is 9.5 km^2, the drainage area is 49.2 km^2, the specific drainage factor is 5.7. The thickness of sediments is 1.8 m (Bgatov et al., 1990). The lake is an internal-drainage basin; feeding is due to spring floods and atmospheric precipitations. The lake is located on the area of peat deposits (Bgatov et al., 1990).

Lake Minzelinskoe (55°33′ N, 83°16′ E) is located in the subzone of the southern forest–steppe landscape zone in the Novosibirsk Region in the northeastern part of the Kolyvan' District (Fig. 1). The lake occupies an intrazonal position: it is on the Ob' floodplane-terrace geomorphologic area on the first ancient terrace on the left Ob' River bank (the terrace is elevated above the floodplain by 5–7 m). This terrace was formed by layered sands and sandy loam sediments (Bgatov et al., 1990). The Ob' River terrace complex, and generally, the left bank of river value, where Lake Minzelinskoe is located, includes two terrace steps (Tolmachevo and Kudryashi terraces). The Tolmachevo terrace of about 20 m high was formed during the second half of the late Pleistocene of about 50–30 ka. The alluvium of the terrace is overlapped by a 10–15 m layer of loess-like loam of the time of the Sartan glaciation that is its outstanding feature. The Kudryashi terrace is 10 m high and it not overlapped by loess, which limits the time of its formation by the late glacial time about 13–11 ka (Orlova, 1990). The Kudryashi terrace has an atypical sequence for fluvial deposits. It is devoid of argillaceous floodplain facies and is entirely composed of sand, often coarsely crossbedded. Such features allow one to assume terrace formation as a result of catastrophic run-offs during the discharge of waters from giant lake reservoirs in Altai (Volkov, 2007).

The lake length is 12 km, the greatest width is 2.2 km, the average depth is 0.35–0.65 m, maximum depths in the northeastern part of the lake are 2.0–3.5 m. The water area changes from 13.8 to 19.8 km^2 at various levels of water (mean water area is 15 km^2), the drainage area is 31.9 km^2, according to the low indicator of the specific drainage area (1.6–2.3), the lake belongs to accumulative water bodies, the thickness of sediments is, on average, 4.5. The lake is an internal-drainage basin; feeding is due to spring floods and atmospheric precipitations. The lake has no tributaries, the Krutishka River flows from it, and the river then flows into the Ob' River. On the drainage area, cedar, pine, and birch forests are widespread, as well as extensive swamps (Bgatov et al., 1990).

Lake Kotokel' (52°49′ N, 108°09′ E) occupies the most of the Kotokel' depression located in the southeastern coast of the Middle Baikal at a distance of 2 km (Fig. 1). Morphologically, the Kotokel' depression represents the southern section of Kika–Kotokel'–Kotochik–Turka group of depressions within the transition zone from the Baikal rift to the forefront step of the Ulan-Burgasy range (Solonenko et al., 1968). Various stratified formations participate in the geological structure of the mountain frame and bottom of the Kotokel' depression: Archean, Proterozoic, and Cenozoic groups, late Proterozoic intrusions of the Barguzin complex and others (Shobogorov, 1981).

The lake length is 15 km, the mean width is 4.6 km, the mean depth is 4.0–4.5 m (maximum depth is 14 m), the surface area of water is 70 km^2, the water drainage area is 183 km^2, according to the low drainage factor (2.6), the lake is qualified as accumulative water body, the thickness of bottom sediments is ~20 m, of which organic sapropel accounts for ~6 m (Pronin and Ubugunov, 2013).

Lake Dukhovoe (53°17′ N, 108°52′ E) is located on the coast of the Barguzin Bay, 18 km from the mouth of the Barguzin River and 2 km from Lake Baikal near the Maksimikha Village (Fig. 1). The geology of the Barguzin Bay is represented by various formations such as upper Proterozoic–lower Paleozoic granitoids, Paleozoic granitoids, syenites, diorites, and others (Galazii, 1993). Lake Dukhovoe is located in a vast, almost round basin, closed in mountains. The lake was formed in the Late Glacial Period after formation of the fourth terrace of Lake Baikal. The lake length is 2.5 km, its width is 1.6 km, its depth is no more than 2.8 m, the area of water surface is 7.15 km^2. Several streams flow into the lake on the eastern and southern shores; the small Dukhovaya River flows out of the lake, then the latter flows into Lake Baikal (Dumitrashko, 1949).

Lake Ochki (51°30′ N, 104°53′ E) is located on the southeastern Baikal coast on the right bank of the Vydrinnaya River. It occupies the topographic lowering in the moraine rise of glacial origin at the foot of the Khamar-Daban Range (Fig. 1). This is one of the typical ridges of terminal moraines abundant in the mouths of trough valleys descending to Lake Baikal from surrounding mountain ranges. The trough line was formed during one of stages of the late Pleistocene glaciation and it has a horseshoe-like form about 7 km wide and about 50 m in height. The Vydrinnaya River cuts it in the middle and it has a rather developed valley of width about 200 m in the narrowest place. The moraine surface is leveled, and the upper layer is reworked by denudation. Inversion depressions specific for moraine lines are shallow and filled with boulder-sandy material confined mainly to a bar crest and form a chain of lakes and peats (Krivonogov, 2010).

The depth of Lake Ochki is about 3 m, the Vydrinskoe upland bog is located in adjacent topographic lowering with peat thickness of 4.5 m. The lake, in conjunction with the upland bog, makes up the lake-marsh system complementary to each other. Feeding of both the lake and marsh is preferentially atmospheric (Leonova et al., 2015), input of deep underground waters trough zones of tectonic dislocations is also not inconceivable (cold nitrogen and methane waters are characteristic for this region (Galazii, 1993).

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 IGM 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, dericks 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. After drilling, the cores were unloaded from a sampler, then pH and Eh of lacustrine sediments were measured using the

"Anion 4151" analyzer, previously described and photographed, packed entirely into polyethylene and plastic cases and transported, in the undisturbed state, into a laboratory for further study.

The applied vibration drilling technology makes it possible to uncover the entire lacustrine sediments and to penetrate underlying rocks by 15–20 m (Krivonogov et al., 2012). Obtaining long drill cores is an undoubted advantage of this approach to study of geochemistry of lacustrine sediments that gives a possibility for study of diagenetic OM transformation not only in the upper layer on the boundary "suprabottom water–sediment" on the model of (Belkina, 2003), but throughout the whole sequence of a sapropel strata opened by drilling, most commonly stratified and made up of primary organic matter heterogeneous in origin. 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 OM sources of OM inputs, and, consequently, different OM genesis in stratified layers of sedimentary strata.

Pyrolytic analysis in the Rock-Eval version (RE-pyrolysis approach) has been performed on a SR Analyzer (Humble Instr.Ins.™, US) for thermal decomposition of OM into macromolecules. The essence of the method is in step-wise heating of studied bioproducer samples and lacustrine sediments in a helium current using a temperature program (at the first stage, a sample is kept at the isotherm $T = 250$ °C (3 min) and then it is heated at the isotherm $T = 650$ °C with the rate of 50 °C/min). The gas flow supplied from reactor into flame-ionization transducer, which records only the hydrocarbon component in the gas. Chromatography-mass spectrometry (Pyr-GC-MS, according to (Kashirtsev et al., 2001). was used for study of the distribution of hydrocarbons and labile components of the protein-carbohydrate-lignin substances in thermodesorbates of bioproducers and lacustrine sediments (analyst Dr V.N. Melenevskii, IPGG SB RAS). Identification of compounds was carried out using the reference data (NIST) of mass-spectra and retention times of mass-spectra and retention times (Ralph and Hatfield, 1991).

Elemental analysis of organic sapropel matter was performed in accordance with the procedure (Fadeeva et al., 2008) on automatic CHNS-Analyzer (analyst Dr. V.D. Tikhova, NIOCH SB RAS). Oxygen was determined by calculation.

Determination of organic carbon (C_{org}) in sapropel samples was performed in accordance with the Tyurin procedure (Vorob'eva, 1998) (analyst L.D. Cherepakhina, ISSA SB RAS).

Biological analysis of sapropel deposits from lakes Ochki and Minzelinskoe (quantitative layer-by-layer calculation of remains of aquatic organisms, terrestrial and bog vegetation in stratified sequences) has been carried out according to the Korde method (Korde, 1960, 1968) (analyst Dr. T.A. Kopoteva, IWEP FEB RAS).

Study of micromorphology and material composition of sapropel samples and biological species (plankton, macrophytes) has been carried out on a scanning electron microscope TESCAN MIRA 3 LMU at the MULTIPLE-ACCESS Center for common use for multielement and isotope investigations at IGM SB RAS (MAC MII SB RAS). Different detection modes were used: mode of secondary electrons providing insight into morphology of topography; mode of back scattered electrons or phase contrast regime, which gives a possibility to obtain a distribution pattern for electron density in a studied sample; mode of characteristic X-ray radiation, which gives a possibility to carry out X-ray spectroscopic microanalysis and to obtain data on elemental composition of the sample (Goldstein et al., 1981).

The age of sapropel strata was dated by the radiocarbon method using accelerator mass-spectrometry (AMS ^{14}C) at the laboratory "Cenozoic Geochronology" of the MAC MII SB RAS (analysts L.A. Orlova, E.V. Parkhomchuk, and A.V. Petrozhitskii). Determination of residual carbon activity has been performed using the instrument QUANTULUS-1220 (Liquid Scintillation Counters).

PRODUCTION TYPES OF STUDIED LACUSTRINE ECOSYSTEM AND RECENT SOURCES OF ORGANIC MATTER

Synthesis of primary OM, which is realized by autotroph: phytoplankton, perephyton, and aquatic higher plants (macrophytes), are of great importance for the formation of organogenic bottom sediments. The contribution of each group of autotroph organisms to amount of primary production being created depends on the degree of their development in biocoenosis of water body. Formation of lakes with the prevailing role of either macrophytes, or phytoplankton in OM synthesis is the result of historical development of lakes affiliated to certain combinations of physical-geographic (outside water body) and limnic (inside water body) conditions (Pokrovskaya et al., 1983).

Among the studied water bodies, lakes in the south of the West Siberia: Bol'shie Toroki and Minzelinskoe (Fig. 2*A*) belong to the production-macrophyte type of ecosystems in accordance with the typification (Pokrovskaya et al., 1983). According to the morpholimnic classification (Savchenko, 1997) and based on the ratio of littoral (coastal) zone area to profundal (deep) one, these lakes belong to the littoral type. The average depth in Lake Bol'shie Toroki was 0.50 m in August of 2012 and it was 0.35 m in Lake Minzelinskoe. These lakes have a gently sloping bottom with a suitable substance for rooting submerged vegetation. Shallowness, the absence of water stratification, and the flat bottom give rise to mass development of macrophytes in these lakes. Thus, in Lake Bol'shie Toroki, macrophytes are distributed throughout the entire bottom area forming a continuous carpet, and they form thick underwater terraces in the littoral water body zone. Production of OM in these lakes occurs

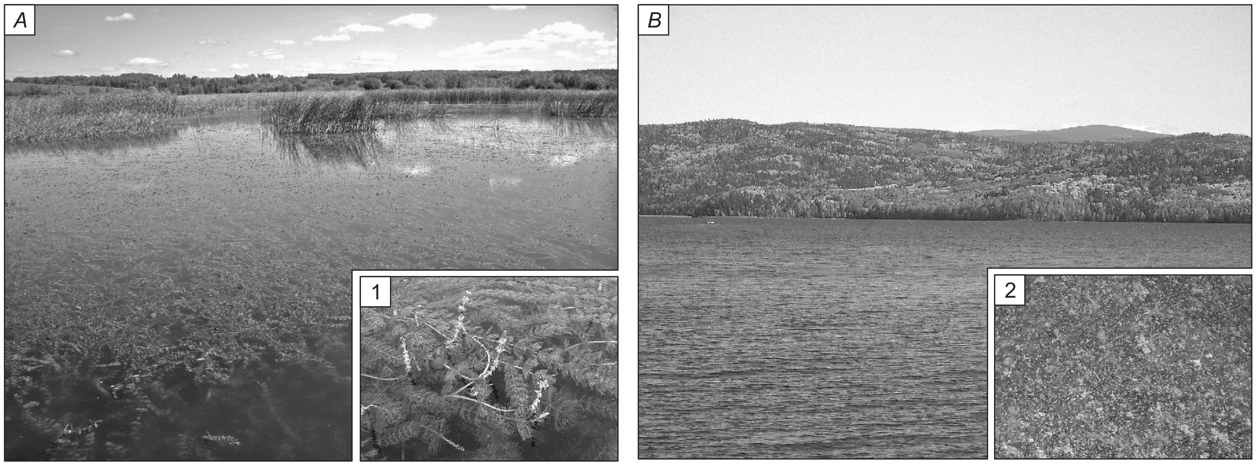

Fig. 2. Production-macrophyte type of lake ecosystems of the south of West Siberia (Lake Minzelinskoe) (*A*), where macrophytes (*1*) are the main producers of OM. Production-phytoplankton type of lake ecosystems in the East Baikal region (Lake Kotokel') (*B*), where phytoplankton (*2*) is the main OM producer.

mainly due to photosynthetic activity of submerged macrophytes (Zarubina, 2013; Ermolaeva et al., 2016). According to (Pokrovskaya et al., 1983), such lakes are referred to as "macrophyte" meaning by this term certain properties of their production systems, in which the main role is assigned to submerged macrophytes, whereas the secondary role is assigned to phytoplankton when united intra-water reservoir process of OM synthesis is implemented.

As opposed to studies of West Siberian macrophyte lakes (Pokrovskaya et al., 1983), the lakes adjacent to the Lake Baikal Region, i.e., Lakes Dukhovoe and Kotokel' are referred, in accordance with the typification, to *production-phytoplankton type of lake ecosystem*, the main producer of organic matter in which is phytoplankton (Fig. 2*B*). According to the morpholimnic classification (Savchenko, 1997), Lake Dukhovoe belongs to the littoral type, whereas Lake Kotokel' belongs to the profundal-littoral type, characterized by decrease of the area of shallow littoral to 10–30% and, correspondingly, increase in the area of the profundal type, i.e., deep-water zone. In Lakes Dukhovoe and Kotokel', favorable conditions for the mass development of phytoplankton occur, which is the main producer of OM in recent biocoenoses of these lakes (Leonova et al., 2014, 2015; Romanov and Ermolaeva, 2014).

Lake Ochki can be attributed to neither phytoplankton, nor to macrophyte lakes. During synthesis of autochthonous organic matter, the main role is played by phytoplankton. In spring and summer, this true mosses and diatomic algae dominate, and in autumn—chrysophytes. The source of allochthonous OM are sphagnum mosses and chlorophytes, fragments of which enter the lake from peat floating mats forming the shores, that is due to the close proximity with the lake of the Vydrinskoe upland bog. However, if the allochthonous OM source is excluded, the main producer of autochthonous OM is phytoplankton (Leonova et al., 2015).

Consequently, having revealed dominant types of producers in terms of their primary production in recent lacustrine biocoenoses, we can infer only the recent source of OM supply into bottom sediments, as well as speculate about the genesis of OM (macrophyte or plankton) only from the upper (recent) horizons of sapropel deposits. Thus, the main source of supply of recent autochthonous OM into the upper sapropel horizons being formed in studied lakes in the south of the West Siberia (Bol'shie Toroki and Minzelinskoe) are macrophytes according to (Zarubina, 2013; Ermolaeva et al., 2016). In the studied lakes of the Lake Baikal Region, the main supply source of recent autochthonous OM into the upper sapropel of Lake Dukhovoe (Leonova et al., 2014), Lake Kotokel' (Leonova et al., 2014; Romanov and Ermolaeva, 2014), and Lake Ochki (Leonova et al., 2015) are various groups of phytoplankton. In order to reveal sources and genesis of OM buried throughout the whole depth of the strata of lacustrine deposits, it is necessary to confirm OM genesis by applying a complex of organic-geochemical criteria, indicators of genesis, as well as by investigating the material composition of stratified sapropel sequences by the methods of scanning electron microscopy (SEM).

SOURCES AND GENESIS OF BURIED ORGANIC MATTER IN SEQUENCES OF SAPROPEL OF STUDIED LAKES

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.

The biostratification of the sapropel Holocene sequence of Lake Ochki (Fig. 3*A*) revealed heterogenic origin of organic matter: remains of phyto- and zooplankton as autochthonous source of OM, and humus flakes, remains of green algae (*Drepanocladus*), and Sphagnum as allochthonous OM 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%.

Of three components attributed to allochthonous OM, 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 peat-forming 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.

According to the data of pigment analysis of Dr. M.A. Klimin, sediments of Lake Ochki were accumulated and preserved over the Holocene and remained practically unchanged (or with slight changes) of pigment characteristics. This is possible only in the case of rapid conservation of vegetative matter due to the presence an oxygen-free zone at the lake bottom. We also noticed the good preservation of zooplankton remains in the lower part of a sequence. The sapropel layer at the depth 75–150 cm, formed during the period from 3585 to 2880 years ago, is not only characterized by maximum sedimentation rate, but also corresponds to the period of maximum productivity of water body ecosystem.

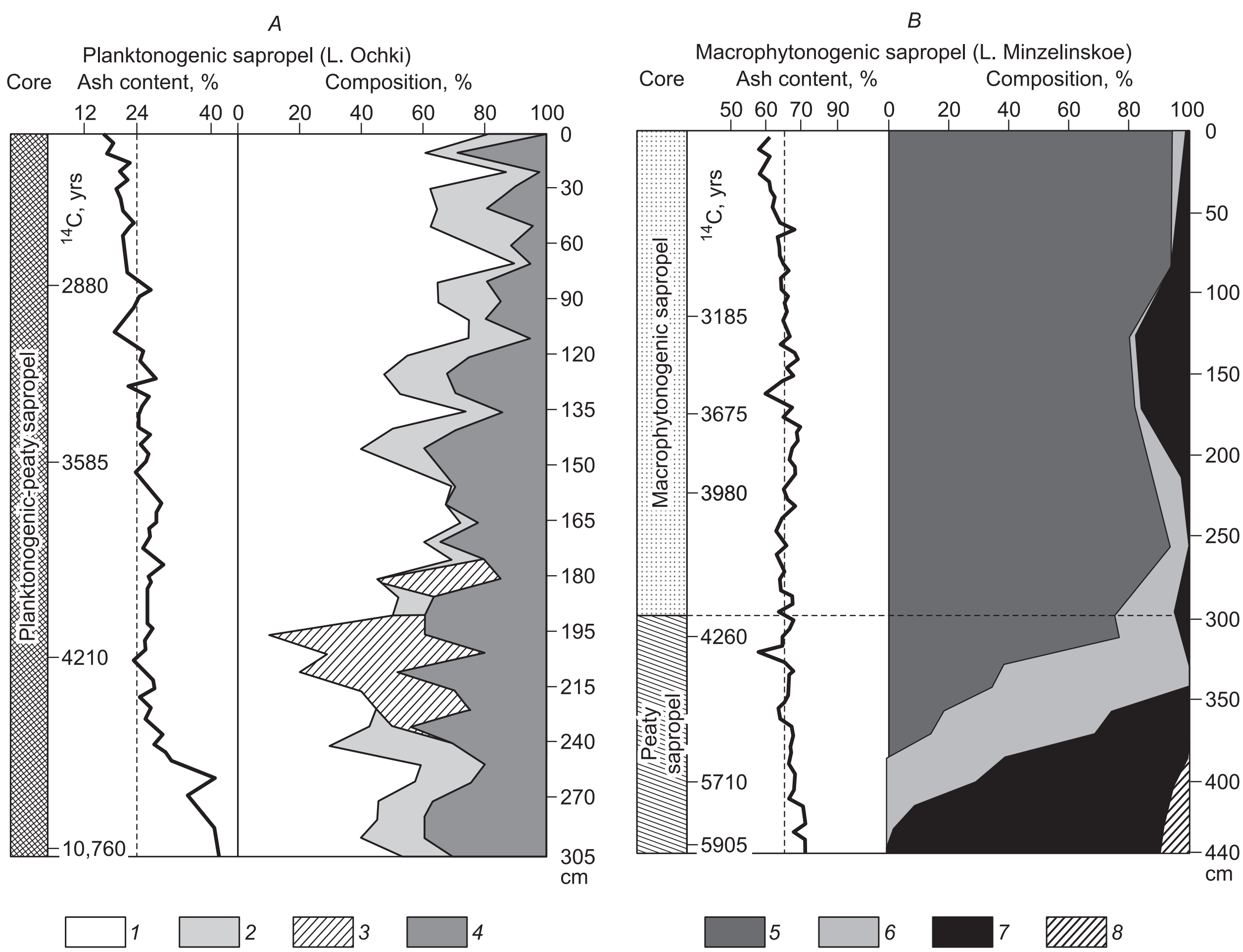

Fig. 3. Biostratification of Holocene sapropel sequences of Lakes Ochki (*A*) and Minzelinskoe (*B*) according to the data of complex biological analysis performed by T.A. Kopoteva (Institute of Water Problems, FEB RAS). *1*, plankton; *2*, green mosses; *3*, sphagnum mosses; *4*, humus flakes; *5*,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

Thus, during 10,760 years, planktogenous sapropel (autochthonous source of OM) 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 OM source).

The biostratification of the Holocene sequence in Lake Minzelinskoe (Fig. 3*B*) also points to the heterogeneous origin of organic matter: remains of submerged (hydrilla, water soldier) and semisubmerged (typha, bulrush) macrophytes (autochthonous OM source), remains of green mosses (allochthonous OM source). The upper strata of sediments up to the horizon 300 cm are represented by macrophytogeneous 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, macrophytogeneous 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 m 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 bulruch, 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 macrophytogeneous 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 OM composition obtained with using scanning electron microscope (SEM).

Material composition of lacustrine sapropels according to data of scanning electron microscopy. The main part of the sapropel substance from Lakes Bol'shie Toroki and Minzelinskoe makes up amorphous detritus, which represents an assemblage of small (from several μm to several mm) amorphous particles of plants (mainly submerged macrophytes, so-called "soft" vegetation). The content of detritus reaches 80% of organic matter (Fig. 4*A*, *C*). The remaining fraction of OM accounts for undecomposed plant remains of semi-submerged macrophytes (bulrush, thypa, so-called "hard" vegetation) and mosses. Single frustules of benthonic diatoms (*Pinnularia*) are present.

The data of scanning electron microscopy have shown that sapropels from Lakes Kotokel' and Dukhovoe represent a structureless mass of colloidal organic matter of the planktogenic genesis (phytoplankton and cyanobacteria) including single fragments of macrophytes and a large number of siliceous diatom frustules 10–30 μm in size (Fig. 4*D*, *E*). In the sapropel substance from Lake Kotokel', cells of centric diatoms *Cyclotella* sp. (Fig. 4*D*, the large, round entity at the center) and cysts of Chrysophyte algae were also found. The sapropel substance from Lake Ochki is composed of interlayers of finely dispersed organic matter, frustules of benthonic diatoms, and weakly-decomposed green mosses (Fig. 4*D*).

Data of pyrolytic analysis of organic matter. Organic matter of recent lacustrine sediments, by definition, is immature and it is represented by the continuity from remains of dead organisms to products of their reworking such as biopolymers (cellulose, proteins, lignin, fulvic and humic acids, and others), and geopolymers, the relationship between them depends on the diagenetic transformation of OM. Pyrolysis of immature OM represents a complex process including several studies of thermal destruction of its separate components with different chemical properties and thermal stability (Melenevskii et al., 2015).

RE-pyrolysis. The analysis results of bioproducers) (zooplankton and phytoplankton), as well as sediments of Lake Ochki are shown in Fig. 5*A*. The pyrogram of zooplankton is a superposition of three peaks: low-temperature S_1 with $T_{peak} < 250$ °C, intermediate peak (S_2') with two partially separated peaks lying on the right limb of intermediate peak at $T_{peak} \approx 440$ °C. Phytoplankton is mainly represented by intermediate peak S_2' assigned to the decomposition of thermolabile OM components.

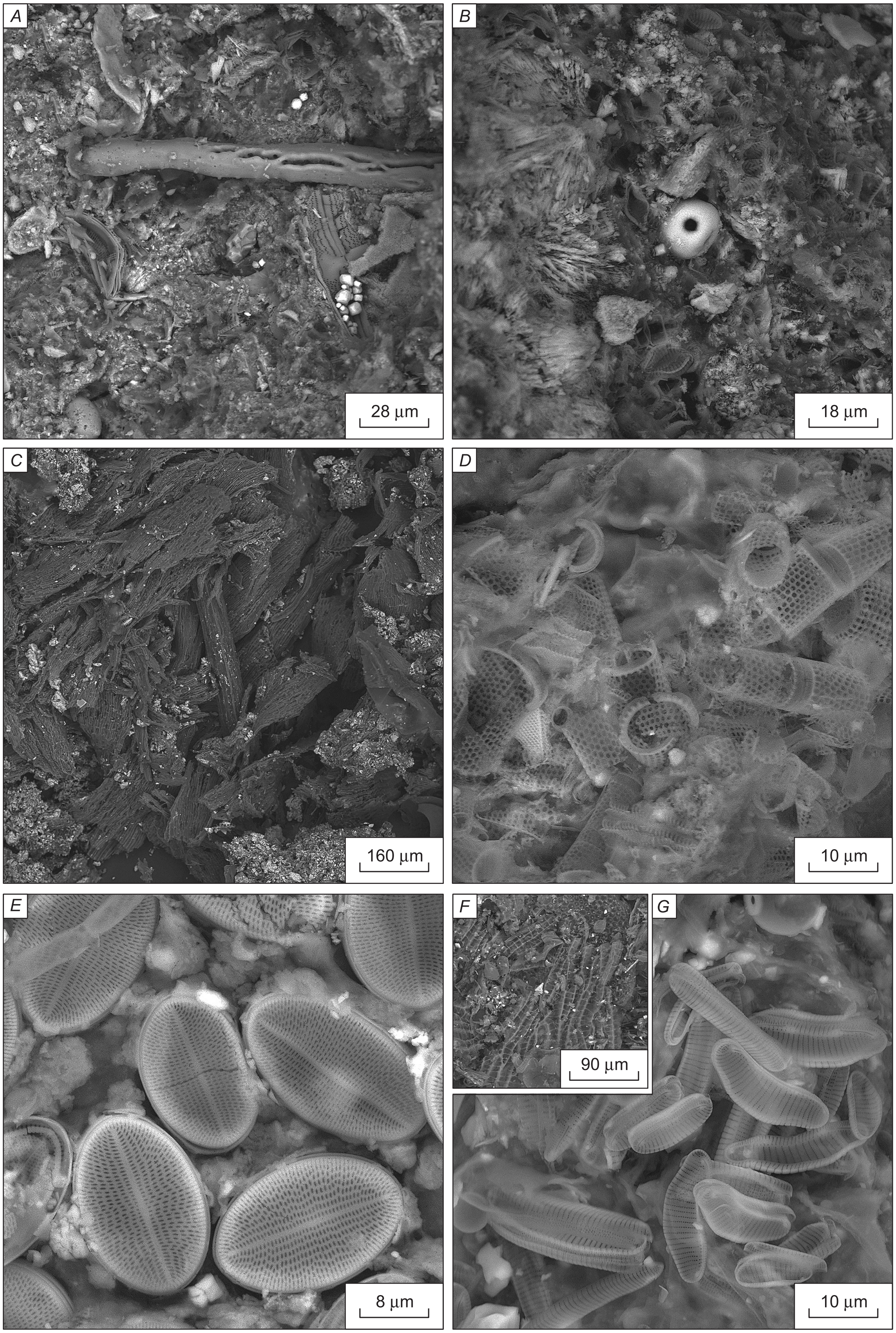

Fig. 4. Material composition of lacustrine sapropels: macrophytogenic of Lake Bol'shie Toroki (*A*) and Lake Minzelinskoe (*B*), peaty Lake Minzelinskoe (*C*), planktonogenic (Lake Kotokel' (*D*) and Lake Dukhovoe (*E*), and planktonogenic-peaty (Lake Ochki: peat (*F*) and plankton (*G*)). Scanning electron microscope TESCAN MIRA 3LMU.

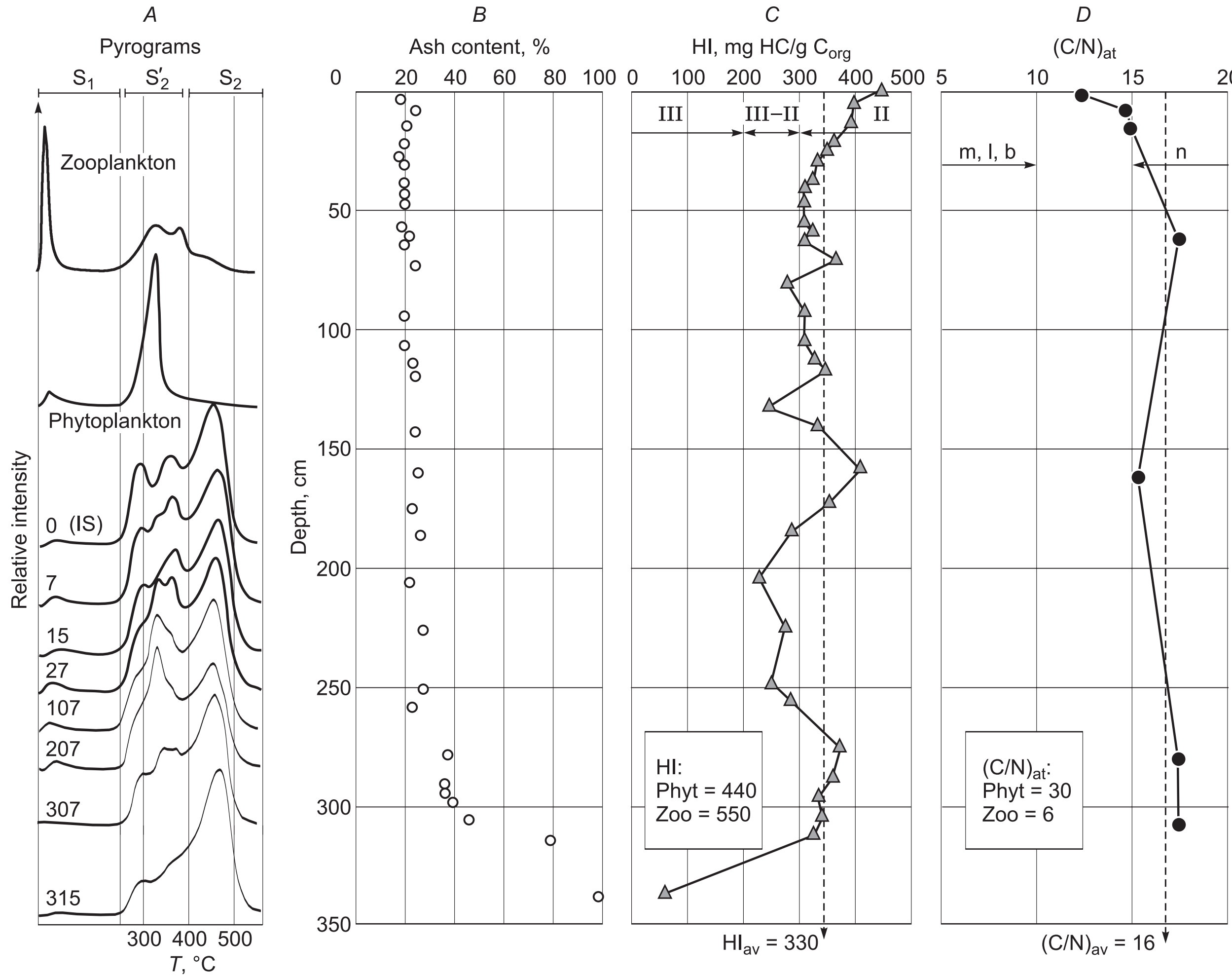

Fig. 5. Pyrograms of zooplankton, phytoplankton, and samples of sediment from Lake Ochki (*A*). Figures in the pyrograms correspond to the depths (cm) of analyzed sediment samples. IS, incoherent sediment. Along the *y*-axis the intensity of a signal of flame-ionization detector normalized at temperature 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 $(C/N)_{at}$ for phytoplankton and zooplankton (*D*). HI was calculated by the formula HI = $\Sigma HC \times 100/C_{org}$, where ΣHC is the total amount of pyrolytic hydrocarbons ($S_1 + S'_2 + S_2$) (mg HC/g of sediment); $(C/N)_{at}$ 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.

In pyrograms of sediments (Fig. 5*A*), beginning from incoherent sediment (IS) the predominance of a high-temperature "kerogen" peak is clearly seen at $T_{peak} \approx 460$ °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 OM composition and, consequently, to increase of the degree of its diagenetic transformation. The peculiarity of pyrolysis of bioproducers and organic matter of sediments of Lake Ochki (Fig. 5*A*) is in the sharp dissimilarity of pyrograms of bioproducers and organic matter from pyrograms of bottom sediments: if the thermal decomposition for the former occurs mainly to 400 °C (peaks S_1 and S'_2), 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 OM is rather constant within the interval of sediments 0–250 cm and is about 80, the ash content is, correspondingly, 20% (Fig. 5*B*), however, the data of pyrolysis and elemental analysis demonstrate qualitative changes of organic matter (Fig. 5*C*, *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. 5*C* and 5*D*. The most oxidized OM is observed at the base of the sapropel strata (339 cm). This may be due to

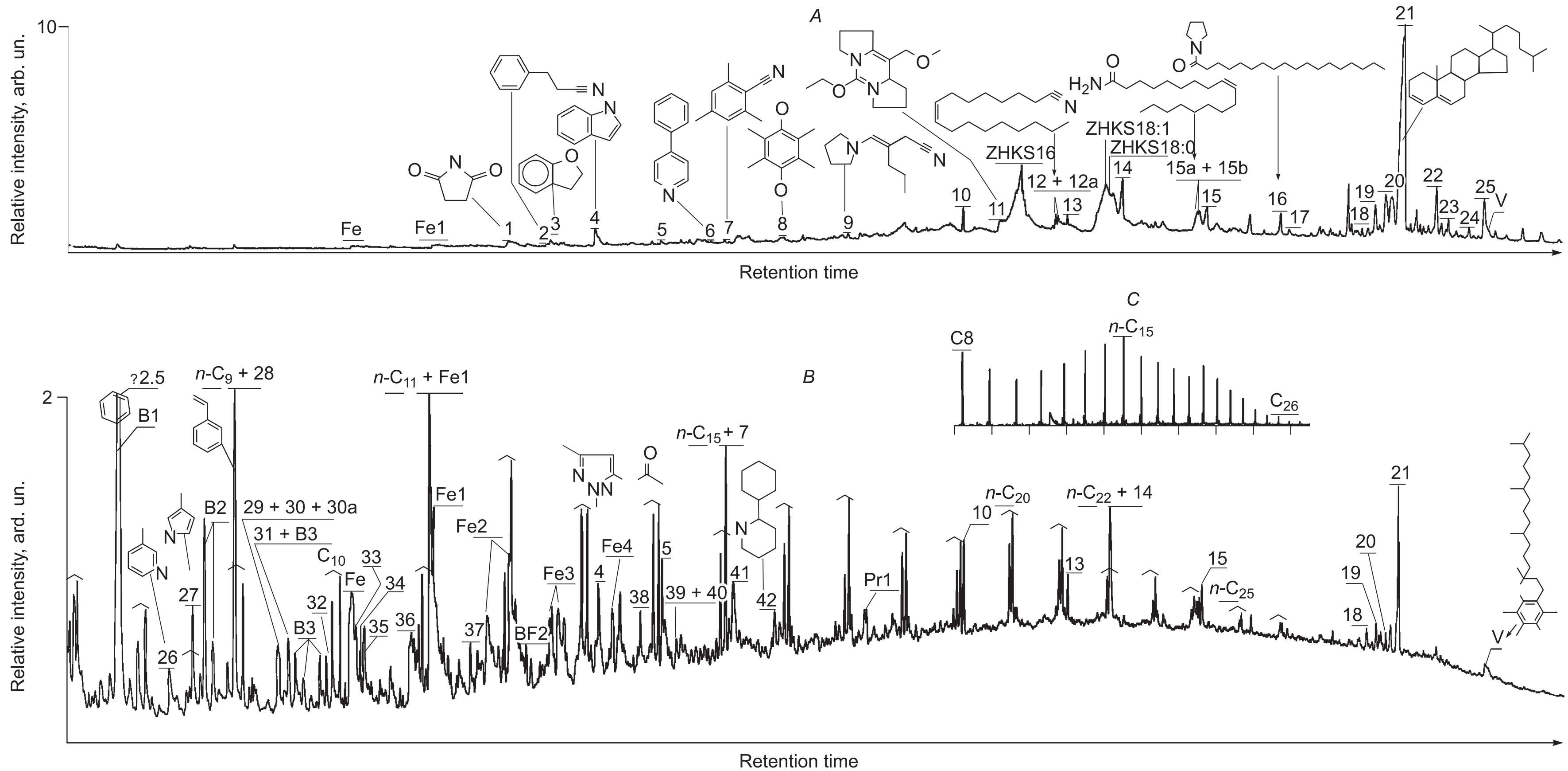

Fig. 6. Total ion current (TIC) mass-chromatograms of products of zooplankton pyrolysis: (*A*) at 330 °C; (*B*) at 610 °C (sequential heating of the same sample); (*C*) ion current mass-fragmentation of *n*-alkanes. The scale of ion current intensities of mass-spectrometer on mass chromatogram is given in arbitrary units (Melenevskii et al., 2015).

both the oxidative sedimentation conditions and the oxidation processes during early diagenesis, for example, owing to contacts with aerated groundwaters (Melenevskii et al., 2015).

PYR-GC-MC (chromatography-mass spectrometry). Total ion current chromatograms of zooplankton and phytoplankton from Lake Ochki are demonstrated in Figs. 6 and 7. In the thermodesorbate of zooplankton (Fig. 6*A*), unsaturated diene-cholestane isomer prevails (peak 21). Carbonic acids (ZHKC16, ZHKC18:0, ZHKC18:1) are present in subordinate amounts. Hopanes and *n*-alkanes are not found in analytically definable amounts. Among the remaining compounds, nitrogen compounds of different compositions dominate, their structures are given in Fig. 6*A*. Homologues series of indoles (peaks 4, 5), alkylnitriles (even) (peaks 10, 12, 12a, 13), alkylamides (even) (peaks 14–15, 17, 18) of the C_{16}–C_{22} composition with maximum at C_{16}–C_{18} in the molecular-mass distribution have been identified.

In pyrolysate of zooplankton (Fig. 6*B*), low molecular weight cracking products prevail: toluene (peak B1), then in order of decreasing peak intensity is styrole (peak 28), phenolmethyl (peak Fe1), and doublets of *n*-alkanes/alkenes forming a homologue series within C_9–C_{27} with maximum in the distribution at *n*-C_{15} (Fig. 6*C*) (Melenevskii et al., 2015).

In pyrolysate of phytoplankton (Fig. 7*B*), low molecular oxygen-bearing compounds are most common: furanmethyl (peak F1), furfurol (peak 5), furphurmethyl (peak 9), and medium- and high molecular compounds (Fig. 7*A*): cyclotene (peak 13), fatty acids (ZHKS14 and ZHKS16:0. Identified lipid components include unsaturated isoprenoid pristens (Pr1, Pr2), diphytenes (Pht1–Pht3), and sterenes (St27:2, peak 42, St29:3, peak, and St29:2, peak 45). Pyrolysate also contains aromatic compounds of benzene (B) and phenol (Phe) with different degree of methyl substitution (Fig. 7*C*). Unlike zooplankton, in pyrolysate of phytoplankton, only three nitrogen compounds (peaks 25, 34, and 35) have been identified. They predominate in zooplankton and, vice versa, oxygen compounds predominate in phytoplankton. The main differences between the organic matter of zooplankton and phytoplankton are the predominance of nitrogen compounds of protein complex in the former, and products of decomposition of polysaccharides in the latter (Melenevskii et al., 2015).

Thus, investigation of phytoplankton, zooplankton, and sediments of Lake Ochki applying pyrolytic methods in the version of RE-pyrolysis and Pyr-GC-MS allows the following conclusions to be drawn. The main sources of autochthonous organic matter in the lake are phytoplankton and zooplankton. The presence of hopanoid hydrocarbons and the elemental composition of OM indicate the contribution of biomass of microorganisms into formation of OM sediments. The terrigenous OM component of terrestrial vegetation is present in bottom sediments, that is confirmed by the presence of high-molecular weight odd *n*-alkanes, ketones, methyl ethers of fatty acids, high-molecular weight even homologues of fatty acids, amides, nitriles, and metaxyphenol compounds as well as by a lower value of the hydrogen index. Formation of the macromolecular structure of kerogen begins at the early stages of diagenesis in incoherent lacustrine sediment by inheritance of aliphatic structures of OM producers. The decomposition degree of OM into molecular components increases with depth.

Distribution of biogenic elements (C_{org}, H, N, S, P) and C/N over sapropel sequences. Living organisms are the most important agent of migration of chemical elements in the hypergenesis zone since the living matter accumulates the main biogenic (constitutional) elements C_{org}, H, N, S, and P, as well as H_2O, CO_2 and mineral compounds (Vernadsky, 1978). Therefore, the distribution over sequences of biogenic elements inherited from producers of autochthonous and allochthonous organic matter, are its important geochemical characteristics.

In Lake Bolshie Toroki, the content of C_{org} in macrophytogeneous sapropel (0–120 cm) varies within the range 14–24%, average contents of H and N are 2.5 and 1.7%, respectively (Fig. 8*A*). In peaty sapropel (120–140 cm), the average C_{opr} content is 23%, H and N are 2.9 and 2.0%, respectively. The average content of S is 2.2%, it is higher in peaty sapropel than it is in overlying layers of macrophytogeneous sapropel (0.7%). The increased sulfur contents in the peaty sapropel is due to the presence pyrite in it (Mal'tsev et al., 2014a).

In Lake Minzelinskoe, the C_{org} content in macrophytogeneous sapropel (0–300 cm) varies from 9 to 23 % with the maximum in the highest horizon 0–5 cm (Fig. 8*B*). Within the range of peaty sapropel (300–460 cm), contents of C_{org} vary within 9–19 %. The average contents of H and N in macrophytogeneous sapropel are 4.7 and 2.7%, respectively, and close to those in peaty sapropel (H is 4.2 and N is 2.5%), that may be explained by the similarity of sources of OM formation: aquatic and near-water (marsh) vegetation. Very low S contents have been revealed throughout the whole sequence (<0.2%), with the exception of the interval (420–425 cm), where the S content was 0.96%, what is associated with accumulation framboidal pyrite in this layer (Maltsev et al., 2014b).

In Lake Kotokel', the C_{org} content in the upper 600 cm sequence of planktonogenic sapropel varies insignificantly from 26 to 31%, the contents of H, N, and S vary from 2.0 to 4.0%, from 2.0 to 3.0%, 2.2 to 2.7%, respectively (Fig. 8*C*). It is noteworthy the rather high sulfur contents throughout the section of sapropel (2.2–2.7%) compared with very low concentrations in producers of organic matter ($S < 0.2\%$), which may be due to the presence of iron sulfide (pyrite).

In Lake Dukhovoe, the C_{org} content in the upper interval of the planktonogenic sapropel (0–180 cm) varies from 35 to 42%, the average contents of H and N, are 5.0% and 3.0%, respectively, the S content varies within 0.7–1.3%, and the average P value is 0.07% (Fig. 8*D*).

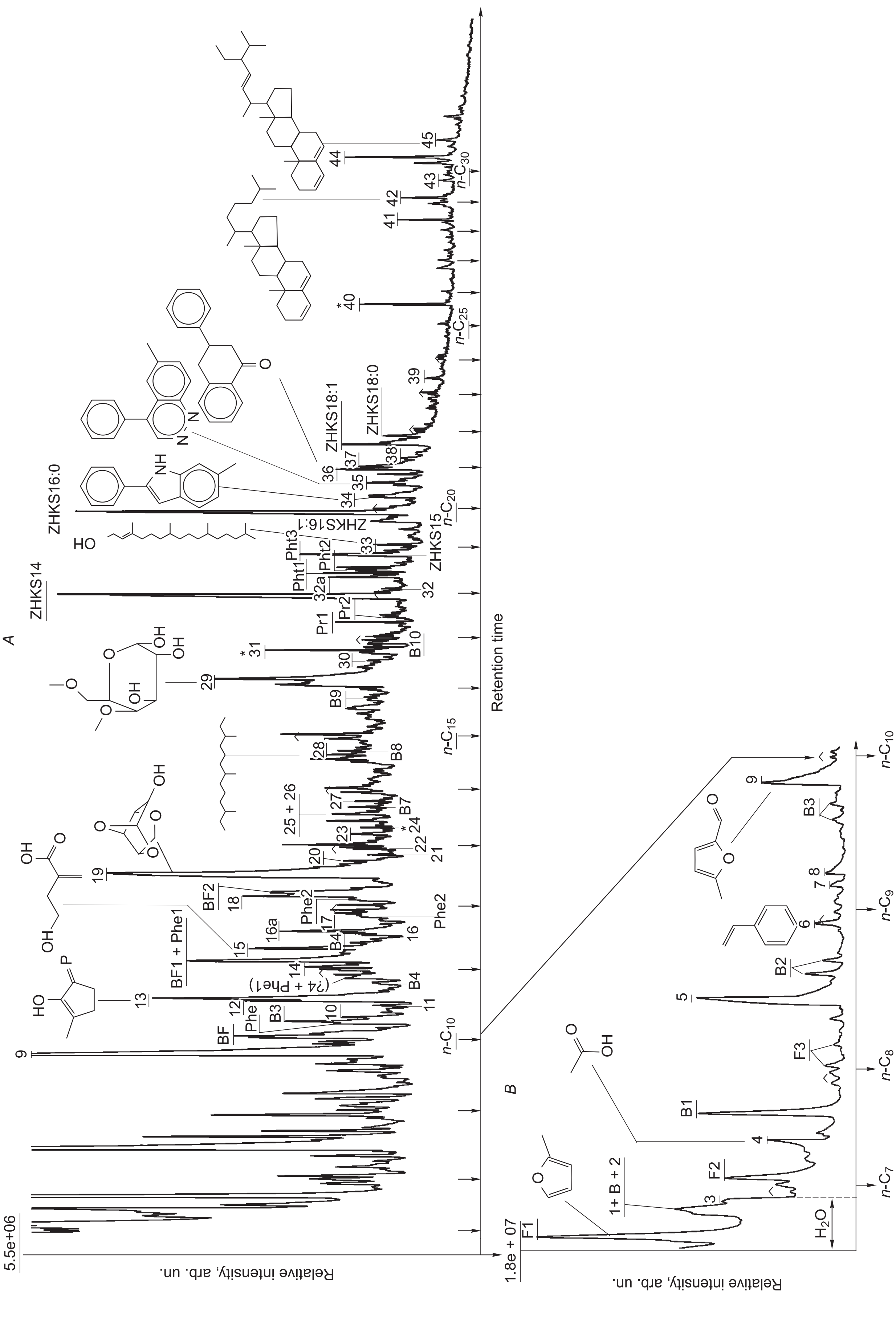

Fig. 7. TIC mass-chromatograms of products of phytoplankton pyrolysis: at 610 °C. Inset of the low-molecular section of the mass chromatogram (*A*); esignations: time stamps of release of *n*-alkanes are shown by vertical arrows on the *x*-axis (*B*) (Melenevskii et al., 2015).

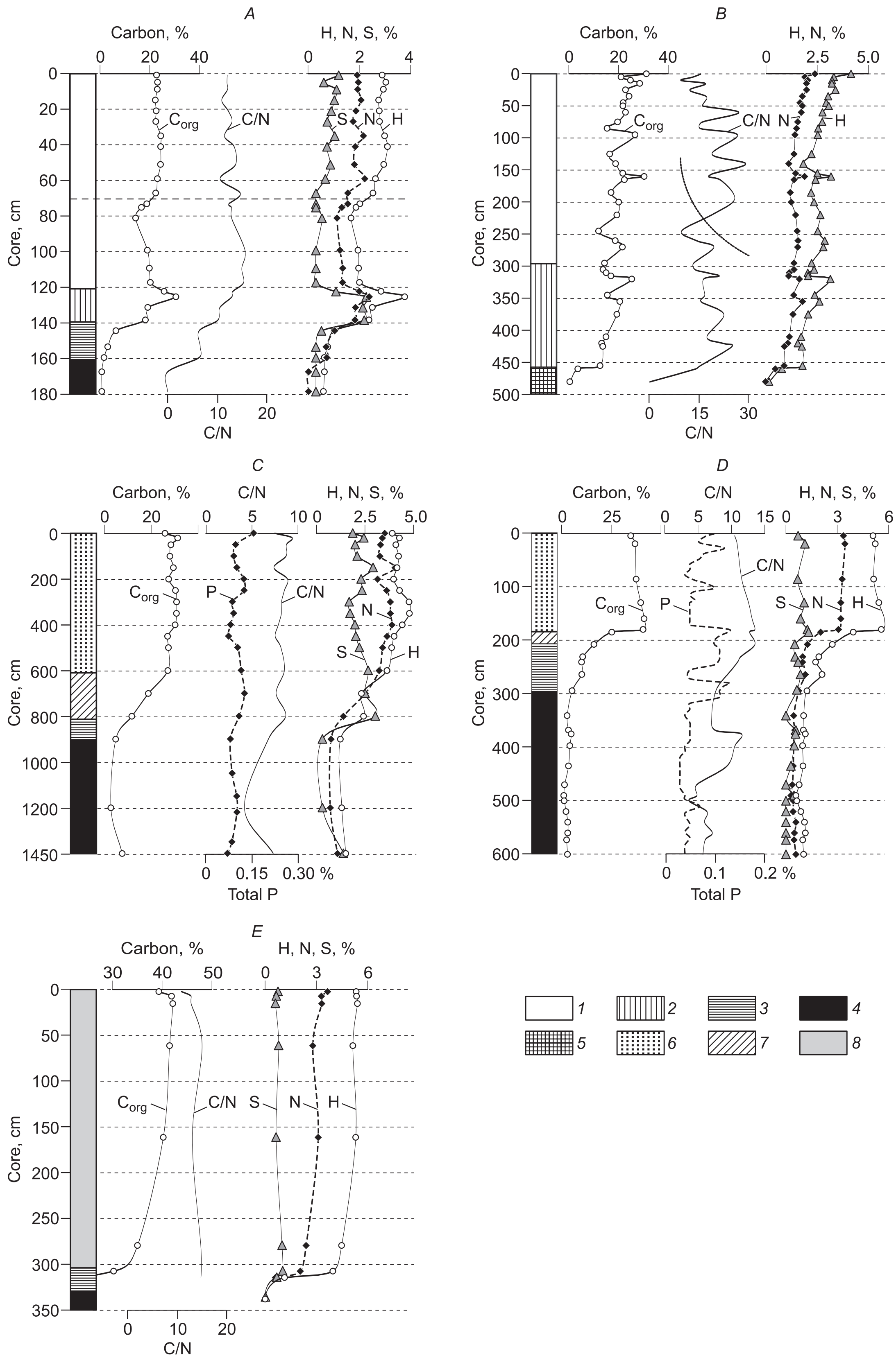

Fig. 8. Vertical distribution profiles of organic carbon (C_{org}), H, N, S, P, and C/N in the Holocene sediments of Lakes Bol'shie Toroki (*A*), Minzelinskoe (*B*), Kotokel' (*C*), Dukhovoe (*D*), and Ochki (*E*). *1*, Macrophytogenic sapropel; *2*, peaty sapropel; *3*, organic-mineral sediment; *4*, underlying clays; *5*, underlying sands; *6*, planktonogenic sapropel; *7*, high ash planktonogenic sapropel; *8*, sapropel of mixed genesis (planktonogenic-peaty).

In Lake Ochki, the distribution of C_{org} over the section of planktonogenic sapropel (0–310 cm) is relatively persistent, the content varies from 39 to 42 % (Fig. 8*E*). The mean H values over the section are 5.0, N and S contents are 2.9 and 0.8%, respectively. Only in the transition interval of sediment (from planktonogenic to peaty) we observe a sharp decrease in the contents of H, N, and S to 1.1, 0.6, and 0.7%, respectively.

The comparative analysis shows that the contents of C_{org}, H, and N are, in total, significantly higher in organic planktonogenic sapropel from Lakes Kotokel', Dukhovoe, and Ochki as compared with organomineral macrophytogenic sapropel from Lakes Bol'shie Toroki and Minzelinskoe.

The C/N ratio *as a marker of sources of autochthonous and allochthonous organic matter.* According to the data (Gashkina et al., 2012) with the C/N ≤ 12 ratio, OM of the autochthonous origin predominate in bottom sediments of small lakes over allochthonous OM in its proportion from 12 to 40–47. In other publications (Koyama, 1954; Wieckowski, 1969), the authors place emphasis on the fact that in small lakes, the influence of the input factor of allochthonous OM into common OM 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. 8*A*, *B*) as compared with typical phytoplankton Lakes Kotokel', Dukhovoe, and Ochki (Fig. 8*B*, *D*, *E*) are significantly different. In Lake Bol'shie Toroki and Minzelinskoe, the C/N values vary in the sequences of macrophytogeneous 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 OM of sapropel from Lake Minzelinskoe in which, according to the data of biological analysis, the share of allochthonous OM (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 OM 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 OM formed mainly by phytoplankton, the share of allochthonous OM, 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 OM of allochthonous origin has been established (the average values are C/N ≥ 12), the source of which are mainly semisubmerged macrophytes (bulrush, thipa). In typical phytoplankton lakes of the eastern Baikal Region (Kotokel', Dukhovoe, and Ochki), OM 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.

Diagenetic transformation of organic matter in lacustrine sediments according to pyrolysis data. The degree of diagenetic transformation of bottom sediments can be judged from the pyrolysis data (in Rock-Eval version). Analysis of pyrogram forms of sediments of studied lakes gives insight into the change of OM composition during diagenesis (Fig. 9). Pyrograms have distinctive peaks: high-temperature (500 °C), indicating the presence of sediments of macromolecular aliphatic structures: kerogen (associations of heterogeneous detrital and finely dispersed organic remains reworked, for the greater part, under anaerobic conditions) and low-temperature peaks (300–400 °C and less) representing labile components of protein-lipid substances).

According to the pyrolysis data, it has been established that already in the uppermost intervals of sapropel sediments, organic matter is subject to deep transformation and differs significantly in composition from bioproducers of OM: macrophytes, phytoplankton, cyanobacteria, and zooplankton (Fig. 9*A*–*D*).

The comparative analysis of pyrograms of bioproducers and sapropel indicates that in the sapropel composition (already in the first 5 cm), labile protein-lipid substances, characteristic for bioproducers (low-temperature peaks ~330 °C) are absent, whereas high-temperature peaks (~500 °C) responsible for the presence of kerogen are present. This indicates that the decomposition of OM falling into sediments (and as consequence, formation of kerogen) begins already at the earliest stages of diagenesis, regardless of genesis of organic matter of sapropels (planktonogenic or macrophytogenic). With further deposition, it seems that very persistent OM remains in the deeper layers of sapropel strata, the further decomposition of which proceeds extremely slowly.

As mentioned above, transformation of buried OM planktonogenic and macrophytogenic sapropels proceeds in a similar way: all labile OM components are subjected to destruction at the early stages of diagenesis, that is reflected in a character of pyrograms, which have high-temperature peaks marking the presence of kerogen in sapropel lying

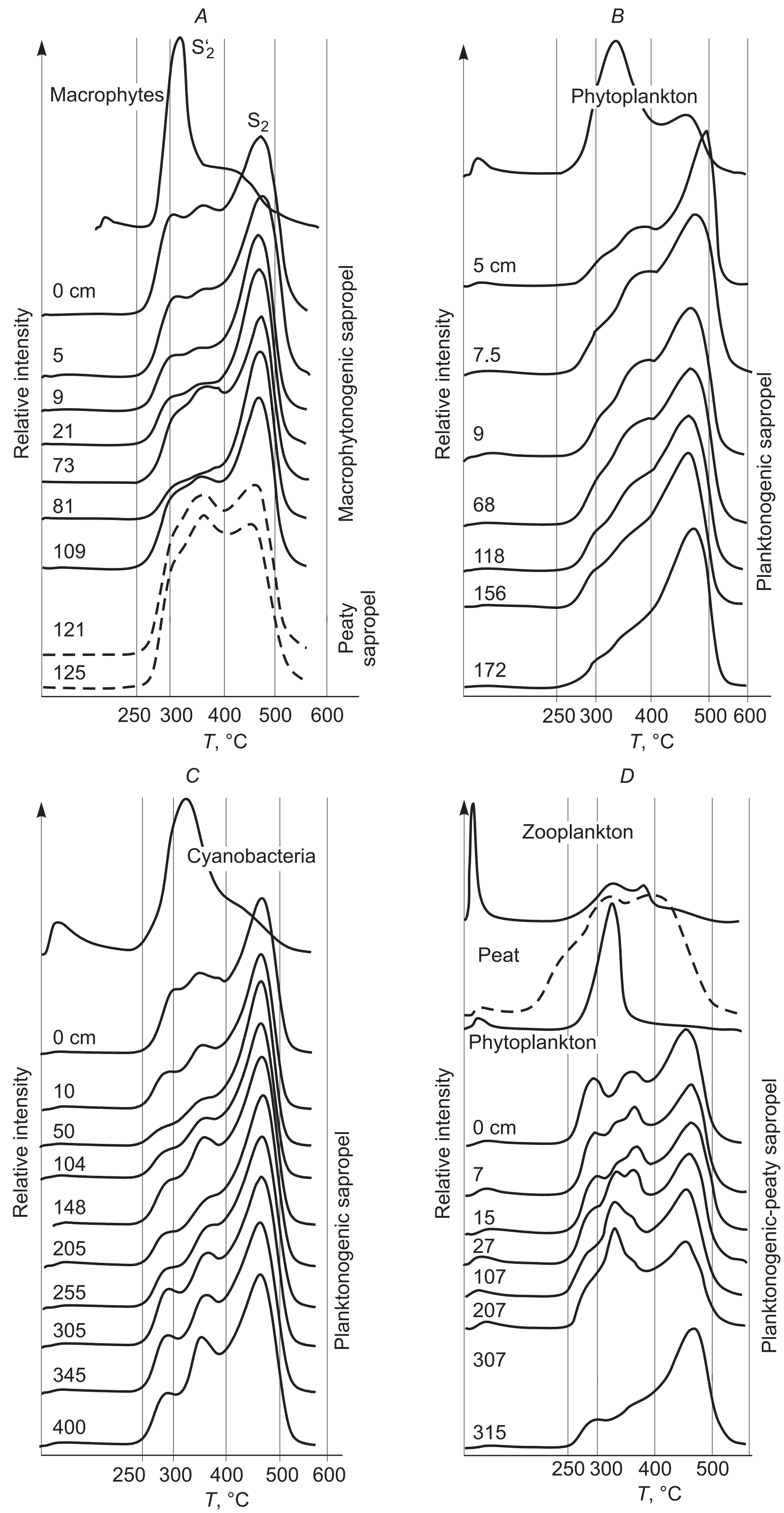

Fig. 9. Pyrograms of bioproducers and sapropel of Lakes Bol'shie Toroki (*A*), Dukhovoe (*B*), Kotokel' (*C*), and Ochki (*D*). As an example, the pyrogram of Lake Ochki shows peat from the Vydrinskoe upland bog incoming from coasts into the lake sediments. The relative intensity in the release rate of a substance per unit of time at a given temperature. Conditional peak S_2' is characteristic peak for OM producers within the range of temperatures ~300–400 °C. The peak S_2 is assigned to hydrocarbon products of kerogen pyrolysis at 300–650 °C. Analyst V.N. Melenevskii (IPGG SB RAS).

within the lower intervals of a section of Lake Bol'shie Toroki and formed by "hard" coastal vegetation (bull rush, typha, mosses) have no temperature peaks. (Fig. 9*A*). Pyrograms of peaty sapropel have well-defined plateau within temperature range from 300 to 500 °C. The absence of high-temperature peaks evidences a lesser fossilization degree of buried OM of peaty sapropel. Forms of pyrograms of peaty sapropel from Lake Bol'shie Toroki are very similar to the form of the Sphagnum pyrogram (Fig. 9*A*) taken from the upper interval (0–5 cm) of the Vydrinskoe upland bog located in the sapropels close proximity from Lake Ochki. Obviously, coastal vegetation consisting mainly of hardly hydrolysable substances, contains, in particular, a large amount of lignin and cellulose (Potekhin et al., 2010), requires a lot of time for fossilization and is slightly susceptible to microbial destruction, which is directly indicated by the similarity of pyrogram forms of the top 5 cm of peat and buried peaty sapropel from the depth 121–125 cm. Heterogeneous sapropel from Lake Ochki (Fig. 9*D*), formed both by OM of autochthonous genesis (phyto- and zooplankton), and by OM of allochthonous genesis (sphagnum from an upland bog) that have the "intermediate" form of pyrograms: mean between macrophytogeneous and planktonogenic sapropels at one side, and peat and peaty sapropel at another side. In particular, pyrograms of sapropel from Lake Ochki, although they have a well pronounced kerogen plateau, include also a high-temperature kerogen peak. Consequently, labile OM of phyto- and zooplankton, as well as cyanobacteria are subjected to destruction during the process of early diagenesis, and decomposition-resistant OM of the allochthonous nature does not undergo significant changes.

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 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 OM 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.

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 Minzelinskoe (south of West Siberia). It has been established that in Lake Ochki, planktonogenic sapropel (phyto- and zooplankton—autochthonous OM source) was formed during 10,760 years, and remains of sphagnum an hypnum moss (allochthonous OM source) were supplied from marshy shores. In Lake Minzelinskoe during the time period dating back to 5905 years ago, peaty sapropel was formed at the stage of flooded hypnum swamp, and accumulation of macrophytogenic sapropel occurs in subsequent time periods (3980–3185 years ago) and up to the present time. Based on the data of biological analysis, sources of supply of organic matter into lacustrine sediments have been established over the entire Holocene period that is confirmed by the results obtained with help of a scanning electron microscope for studying the material composition of sediments from different layers of stratified sequences.

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 OM, which are defined as complex molecular "imprint" of previously living organism formed from biochemical components. Investigation of bioproducers and sapropel from Lake Ochki by pyrolytic methods in versions RE-pyrolysis and Pyr Gc-MC has shown that the main autochthonous OM source in the lake was phyto- and zooplankton.

In pyrolysate of zooplankton, nitrogenous compounds of different compositions are dominant: members of homologue series of alkylnitriles (even), alkylamides (even) of the $C_{16} \div C_{22}$ composition with maximum at $C_{16} \div C_{18}$ in the molecular-mass distribution. In pyrolysate of phytoplankton, mainly low-molecular weight oxygen-bearing compounds: furanmethyl, furfurol, and moderate to high-molecular weight compounds—cyclotene are common. Identified lipid OM components also include unsaturated isoprenoid hydrocarbons: pristenes, diphytenes, and unsaturated sterenes. The presence of hopanoid hydrocarbons indicates the contribution of biomass of microorganisms to formation of OM sediments. In sapropel of Lake Ochki, the terrigenous component of OM (remains of terrestrial vegetation: mosses, sedges and others), that is confirmed by the presence of high-molecular weight odd *n*-alkanes, ketones, methyl ethers of fatty acids, high-molecular weight even homologues of fatty acids, amides, nitriles, and, methoxy-phenol compounds (derivatives of lignin), as well as by the lower value of the hydrogen index (HI). Formation of macromolecular kerogen structure (fossilized OM) begins at early diagenesis in incoherent lacustrine sediment by inheritance of aliphatic structures of OM producers and due to reactions of melanoidin formation. The degree of OM decomposition into molecular components increases with depth.

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

In studied typical macrophyte lakes of the south of West Siberia (Bol'shie Toroki and Minzelinskoe), the high content of OM of allochthonous origin (average values C/N ≥ 12) have been established in sapropel sequences, the source of which are predominantly semi-submerged macrophytes (bulrush, typha), and in typical phytoplankton lakes of the eastern Baikal Region (Kotokel', Dukhovoe, and Ochki), OM predominantly of autochthonous origin (C/N < 12), the source of which is mainly phytoplankton.

The distributions of C/N in stratified sapropel sequences in typical macrophyte Lakes Bol'shie Toroki and Minzelinskoe, as compared with typical phytoplankton Lakes Kotokel', Dukhovoe, and Ochki differ significantly. The C/N values in lakes Bol'shie Toroki and Minzelinskoe vary in depth of sequences of macrophytogeneous sapropel from 10 to 15, in layers of peaty sapropel these values are within 15–18. Generally, for the entire sapropel strata of Lakes Bol'shie Toroki and Minzelinskoe, the C/N values fall into the interval of such values for organic matter just as of aquatic (submerged and semi-submerged macriohytes), so terrestrial vegetation (sedges, cotton grass, and mosses).

Lake Kotokel' is characterized by the lowest C/N values, which vary throughout the depth of the 8 m sapropel sequence from 7.4 to 9.3. In lake Dukhovoe, the C/N values vary throughout the depth of the 180 cm sapropel sequence from 5.7 to 8.6 falling into interval of C/N values of marine and lacustrine plankton. Based on these data, it may be concluded that the main OM source of sapropel from Lakes Kotokel' and Dukhovoe is phytoplankton.

In Lake Ochki, the C/N values in depth of sapropel sequence (0–310 cm) vary within 0.8–14.9, on the average 13.6. The rather high C/N values are explained by the material composition of the sapropel, which is mixed (along with the autochthonous OM, formed mainly by phytoplankton, the share of allochthonous OM represented by remains of mosses is significant.

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