

Benthic algae as major precursors of oil-prone kerogen – A case study from the Hungarian Middle Miocene

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Received: February 21, 2016; accepted: September 6, 2016

This study is intended to clarify the depositional environment of a 180-m-thick, immature, limy Middle Miocene oil source rock interval, cored in the Zala Basin, western Hungary. For this purpose, a highly interdisciplinary approach was applied combining simple, standard micropaleontological, isotopic, and organic geochemical methods, rarely applied together. Foraminifera were studied for estimating bottom oxygenation and water depth, while nannoplankton biostratigraphy permitted for estimating the rate of sedimentation. The studied source rocks were deposited in a rather shallow sea, below well-oxygenated bottom water. The abundant epiphytic foraminiferal fauna proves that the bottom was densely inhabited by benthic algae, while the high $\delta^{13}\text{C}_{\text{org}}$ ($>-22\text{‰}$) clearly indicates massive benthic algal contribution to the kerogen. Mass accumulation rate of the limy upper part of the NN5 nannoplankton biozone, the oil source interval included, was very high ($551 \text{ t/m}^2/\text{Ma}$). In spite of moderate productivity and good oxygenation of the bottom, rapid accumulation of carbonate, produced partly by benthic algae, assured both the great relative weight of the marine organic components and their good preservation. Our results provide the first proof for the possibility of a major contribution of benthic algae to oil-prone kerogen.

Keywords: benthic algae, epiphytic foraminifera, nannoplankton, $^{13}\text{C}_{\text{org}}$, oil-prone

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Introduction

The Neogene Pannonian Basin System (PBS), surrounded by the Carpathian, Dinarides, and Alps (Fig. 1), is a classical petroleum province. Apart from the Jurassic-sourced oil and gas in the Vienna Basin (Ladwein 1988) and the Triassic-sourced oil in the Zala Basin (Koncz 1990; Clayton and Koncz 1994), the bulk of the petroleum accumulated in the PBS is considered to be the product of marly, limy Miocene sediments, widely developed in the basin system.

The Miocene-sourced oils of Hungary form two distinctly different groups: while oils discovered in SE Hungary have been sourced from Late Middle Miocene and Late Miocene brackish and lake sediments, those discovered in the Zala and Drava Basins (SW Hungary) have been generated mostly in the Middle Miocene marine sediments (e.g., Koncz 1990; Šarković et al. 1992; Clayton and Koncz 1994; Clayton et al. 1994; Barić et al. 2000; Hasenhüttl et al. 2001; Lučić et al. 2001; Troskot-Corbić et al. 2009; Badics et al. 2011; Király et al. 2011). On the other hand, according to the available data (Koncz 1990; Clayton and Koncz 1994; Clayton et al. 1994; Fekete et al. 2010), the $\delta^{13}\text{C}$ of oils accumulated in SE Hungary is more negative than that of the Miocene-sourced Zala Basin oils.

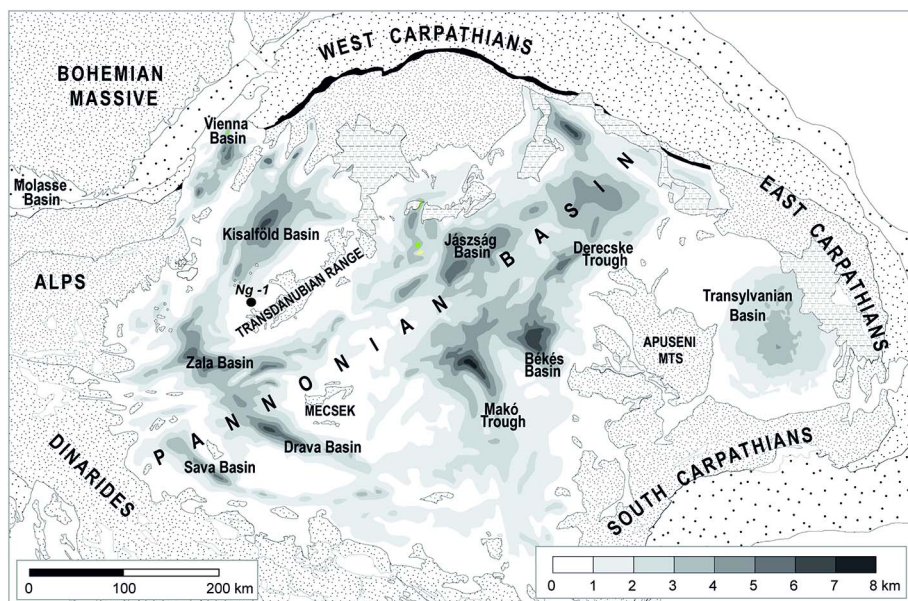


Fig. 1
Iso-thickness map of the Pannonian Basin System with location of the Ng 1 well (after Horváth and Royden 1981)

Many case studies deal with the hydrocarbon potential (HC_{pot}) of the Miocene source sediments in different parts of the PBS (e.g., Alajbeg et al. 1990, 1996; Šarković et al. 1992; Clayton and Koncz 1994; Clayton et al. 1994; Pap and Pap 1997; Barić et al. 1998; Badics et al. 2011), but sedimentological, paleontological, or even lithological information about them are rather parsimoniously provided.

Unfortunately, the few papers focusing on their depositional environment are mostly free of source rock considerations (e.g., Pavelić et al. 1998; Magyar et al. 2004). The only exception is the work of Malvić and Majstorović-Busić (2012), dealing with Upper Badenian–Pannonian source rocks of the Bjelovar Depression, Sava Basin, Croatia. Due to the restricted scope of applied methods in previous work in studying the depositional environment of the PBS source rocks, it is still poorly understood.

An additional difficulty for deciphering the relationship between source rock quality and depositional environment derives from the fact that some of the case studies referred to above deal with mature or marginally mature sediments, from which a part of the generated hydrocarbons have been already expelled, meaning that their HC_{pot} decreased during late burial.

Preliminary studies (Vető et al. 2010; Badics and Vető 2012) revealed that the Nagyörbő 1 (Ng 1) scientific well, cored at the northeastern margin of the Zala Basin (for location see Fig. 1), penetrated Middle Miocene (Badenian age) fair to good quality but immature source rocks of limy marl/limestone lithology in more than 100 m thickness. In spite of its marginal position in the basin, this fully cored Badenian section offers a unique possibility to understand the relationship between depositional environment and source rock quality.

In this study, our results are obtained by a combined application of standard geochemical and micropaleontological methods on the marine Middle Miocene (Badenian age) sediments cored between 400 and 700 m depth by the Ng 1 well. The biostratigraphy and the trophic status of the sea are based on the calcareous nannoplankton record, while recognizing water depth and the oxygenation of the bottom water is based on foraminifera. Organic carbon isotopic composition ($\delta^{13}C_{org}$) and Rock-Eval indices are used as proxies to estimate the relative contribution of planktonic, benthic, and land plant components to kerogen.

Geologic setting

This section is based on the Open File Report No. 1775/1 of the Hungarian Office for Mining and Geology (HOMG) dealing with the Ng 1 well, cored in 1972 to a depth of 1,517 m.

Oligocene sediments are discordantly covered by Miocene ones at 1,067 m depth. The bottom part of the Miocene, penetrated between 1,067 and 923 m, is built up by clay, marl, and gravel, mostly of terrestrial facies.

The bulk of the Miocene, developed between 923 and 350.7 m, consists of fine-grained, non- or slightly layered marine marl and limestone with traces of bioturbation

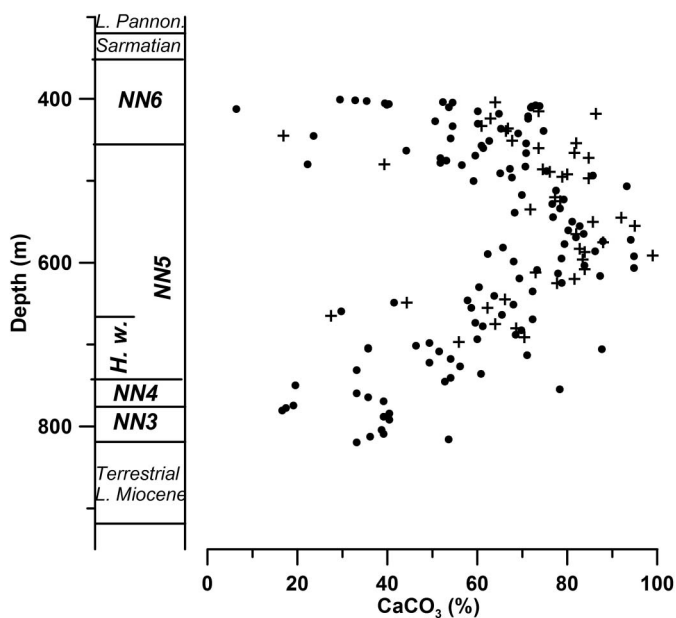


Fig. 2

Variation of the carbonate content with depth in the 350–850 m interval, Ng 1 corehole. The data points are from Open File Report No. 1775/1, HOMG

in some parts. Carbonate content (Fig. 2) gradually increases upward from 20–30% to 80–95% between 800 and 550 m, then decreases with a high scatter to 30–70% between 550 and 400 m. Several red alga-rich limestone beds of 0.1–1 m thickness were penetrated between 706 and 410.4 m. Above 407.5 m, several sand layers were penetrated, while the uppermost 40 m of the marine Middle Miocene consists of sand. The discordantly overlying uppermost Middle Miocene (Sarmatian) and Late Miocene (Lower Pannonian) strata are built up mainly of clastic sediments of brackish and lacustrine origin.

Samples and techniques

Altogether, 28 samples were taken from 762.9 to 404 m for studying the calcareous nannoplankton with the aim of refining the stratigraphy and tracing paleoecological changes. The 44 samples taken from 697 to 404 m were split into subsamples to study foraminiferal assemblages and Rock-Eval indices or determine carbonate content. Organic and inorganic carbon isotope measurements were carried out on about the half of this sample set.

Preparation and determination of calcareous nannoplankton

Smear slides were analyzed using a light microscope (1,000× magnification) at normal and crossed nicols. To trace paleoecological changes, at least 300 specimens from each sample were counted. For biostratigraphic interpretation, the standard nannoplankton zonation of Martini (1971) was used.

Preparation and determination of foraminifera

Samples were soaked in hydrogen peroxide solution for several weeks. In spite of this aggressive treatment, samples taken from below 612 m did not disintegrate. Hence, only the upper two-thirds of the 697–404 m interval were studied for foraminifera.

The 125 µm–0.6 mm fraction was picked for benthic foraminifera and mounted on slides from the split reaching the statistically required minimum of 200 specimens. Altogether, only 14 samples taken between 612 and 404 m were eligible for further statistics by satisfying this requirement. Determining foraminifera to the species level was difficult due to foraminiferal tests frequently covered by calcareous lining hiding characteristic features of the test. This resulted in the high number of undetermined calcareous specimens.

As will be discussed in more detail further on, the far from ideal preservation of foraminifera resulted in some limitations for the utility of multi-proxy approach to reconstruct depositional environment.

Chemical and isotope measurements

About 20–30 g of core sample was ground in a Fritsch ball mill.

Rock-Eval indices – Hydrogen Index (HI), characterizing the hydrogen richness of kerogen, and the temperature of the maximal intensity of the pyrolytic hydrocarbon liberation (T_{\max}), characterizing the maturity of the kerogen – together with the total organic carbon (TOC) content were determined on a Delsi Oil Show Analyser under standard conditions at the Department of Mineralogy, Geochemistry and Petrography in the University of Szeged.

Amounts of inorganic constituents were weighed in the laboratory of the Geological Institute of Hungary. Major element contents were measured with the inductively coupled plasma atomic emission spectroscopy technique after fusion with Li metaborate. The inorganic carbon content was measured volumetrically after HCl treatment. Uncertainties of the measurements were 10–20 relative percentages, respectively.

Organic carbon content of a set of decarbonated samples was oxidized at 480 °C in a Pyrex tube under vacuum following the method of Sofer (1980). The carbonate content of the same sample set was digested using the conventional H₃PO₄ method (McCrea 1950). In both cases, isotope ratios of the CO₂ generated were measured with a Finnigan MAT Delta S mass spectrometer at the Institute for Geological and

Geochemical Research in Budapest. The reproducibility of the $\delta^{13}\text{C}$ data is better than 0.2‰.

Organic carbon content of another set of decarbonated samples mixed with CuO was oxidized at 850 °C. The carbonate content of the same sample set was digested using the conventional H_3PO_4 method (McCrea 1950). In both cases, isotope ratios of the CO_2 generated were measured with a Thermo Finnigan Delta plus XP mass spectrometer at the Institute of Nuclear Research in Debrecen. The reproducibility of the $\delta^{13}\text{C}$ data is better than 0.15‰.

Results

Biostratigraphy

The observed calcareous nannoplankton species are listed in Supplementary Material A.

Based on calcareous nannoplankton, the Ng 1 Middle Miocene sequence can be subdivided into three nannoplankton zones: NN4, NN5, and NN6.

The last occurrence (LO) of *Sphenolithus heteromorphus*, which defines the NN5/NN6 boundary, was observed between samples 436 and 472 m. This biostratigraphically important species has its last appearance datum (LAD) at 13.53 my (Lourens et al. 2004), which fits well with the data of Abdul Aziz et al. (2008). The accompanying assemblages in the *Discoaster exilis* Zone (nannoplankton Zone NN6) are characterized by high percentages of reticulofenestrids: *Reticulofenestra gelida*, *Reticulofenestra haqii*, *Reticulofenestra minuta*, and *Reticulofenestra pseudoumbilica*. The species *Coccolithus pelagicus*, *Coronosphaera mediterranea*, *Cyclicargolithus floridanus*, *Geminilithella rotula*, *Helicosphaera carteri*, *Helicosphaera walbersdorfensis*, and *Umbilicosphaera jafari* occur regularly.

The absence of *Helicosphaera ampliaperta* and the presence of *S. heteromorphus* in the interval from 472 downhole to 731 m indicate the *S. heteromorphus* Zone (nannoplankton Zone NN5). Nannoplankton assemblages from this part of Ng 1 contain high percentages (up to more than 90%) of small reticulofenestrids: *R. minuta* and *R. haqii*. The following species also occur: *C. pelagicus*, *C. mediterranea*, *C. floridanus*, *G. rotula*, *H. carteri*, *H. walbersdorfensis*, *Pontosphaera multipora*, *R. gelida*, *R. pseudoumbilica*, *Sphenolithus moriformis*, *Thoracosphaera spp.*, and *U. jafari*. In the lowermost part of NN5, from 675 to 731 m *Helicosphaera waltrans* occurs. This form was used for biostratigraphic subdivision of nannoplankton Zone NN5 into subzones by a few authors in the Mediterranean (Theodoridis 1984; Fornaciari et al. 1996) and Central Paratethys (Švábenická 2002) bioprovinces. The occurrences of *H. waltrans* were also reported from Lower Badenian (Lower Lagenidae Zone) sediments of the Styrian Basin (Rögl et al. 2002) and the Alpine–Carpathian Foredeep (Čorić and Rögl 2004; Čorić and Švábenická 2004). In the upper part of the Retznei section (Styrian Basin), *H. waltrans* co-occurs

with *Orbulina suturalis* (Rögl, personal communication) within the Lower Lagenidae Zone. The Upper/Lower Lagenidae Zone boundary is placed approximately between the first occurrence (FO) of *O. suturalis* and LO of *Praeorbulina sicana*. The FO of *H. waltrans* in Ng 1 can be dated as 15.47 Ma (Abdul Aziz et al. 2008), whereas its LO can be presumed to be around 14.36 Ma (Abdul Aziz et al. 2008) (between FO of *O. suturalis* and LO of *P. sicana*, within the Lower Lagenidae Zone). Recent results indicate that *H. waltrans* disappears below the Lower/Upper Lagenidae Zone boundary in the Central Paratethys. *H. ampliaperta* co-occurs with *S. heteromorphus* in the 762.9 m sample. Therefore, this sample can be attributed to the *H. ampliaperta* Zone (nannoplankton Zone NN4). The accompanying taxa are *C. pelagicus*, *C. mediterranea*, *C. floridanus*, *H. carteri*, *Helicosphaera scissura*, *H. walbersdorfensis*, *P. multipora*, *R. gelida*, *R. haqii*, *R. minuta*, *R. pseudoumbilica*, *S. moriformis*, and *U. jafari*.

We used the latest calibration of Abdul Aziz et al. (2008) for the range of *H. waltrans*; it is much longer and seems to be more realistic than the earlier one after Lourens et al. (2004).

Abdul Aziz et al. (2008) give the following ages [“The bioevent ages through interpolation of subchron ages according to Astronomically Tuned Neogene Time Scale (ATNTS04)”]:

<i>S. heteromorphus</i>	LO	13.419+–7
	LCO	13.628+–3
<i>H. waltrans</i>	LCO	14.357+–4
	FCO	15.476+–5
<i>O. suturalis</i>	FAD	14.561+–4
<i>H. ampliaperta</i>	AS	15.899+–24

LO, last occurrence; LCO, last common occurrence; FCO, first common occurrence; AS, abundance spike; FAD, first appearance datum.

The age of LAD of *S. heteromorphus* at 472 m in the Ng 1 corehole is 13.53 Ma (after Lourens et al. 2004), which fits well with the ages of LO and LCO of the species after Abdul Aziz et al. (2008).

Depths suggested for the zone/subzone boundaries are displayed in Fig. 2.

Paleoecology

Sediments from the upper part of the *H. waltrans* biohorizon and NN5 Zone contain high percentages of small reticulofenestrids (*R. minuta* and *R. haqii*). The reconstructed oligotrophic paleoconditions of a well-stratified water column enabled the bloom of these small forms during this interval.

Paleo water depth

Almost all samples taken from 433 to 612 m are dominated by epiphytic benthic foraminifera such as *Asterigerinata planorbis*, *Cibicides lobatulus*, *Hansawaia boueana*, and *Heterolepa dutemplei*, with the exception of the sample taken from 480.3 m, dominated by *Bulimina elongata*. The dominating species are listed in Supplementary Material B. The species *B. elongata* is an inbenthic species living relatively deep in the sediment and profiting from high food availability, while able to tolerate dysoxic conditions. However, the majority of the samples are characterized by an epiphytic assemblage, where foraminifera were living on the blades or leaves of benthic plants. Although the benthic plants hardly ever fossilize, the epiphytic foraminiferal fauna reveals the presence of these plants and opens ways of paleo-ecological interpretations (Mateu-Vicens et al. 2010). More specifically, these benthic plants could have been seagrass or more likely benthic algae based on the foraminifera morphotypes present in the material (Langer 1993; Drinia et al. 2005; Mateu-Vicens 2014). This vegetation, providing a home to the epiphytic foraminifera, needs light for photosynthesis, directly indicating the maximum depth of deposition as inside the euphotic zone. In the present-day ocean, the light able to support vegetation penetrates 200 m at most, but optimum depth for benthic plants is even shallower, usually less than 50 m [e.g., *A. planorbis*; see Mendes et al. (2013), among others].

The paleo water depth estimation based on the presence of epiphytes was refined by using the corrected planktonic/benthic (P/B) ratio. Here, we followed the approach of van der Zwaan et al. (1990) as species level determination is less necessary in their corrected P/B ratio than in the more recently published proxy of Hohenegger (2005) based on depth distribution of individual species. Results are displayed in Fig. 3.

The P/B ratio shows a progressive decrease of the water depth from 140–120 m to about 40 m during deposition of the sediments penetrated in the 612 to 480 m interval. This shallowing trend is in accordance with the regionally observed uplift across the NN6/NN5 boundary (Filipescu and Girbacea 1997; Báldi et al. 2002). During the deposition of the rest of the studied sequence, the water depth was about 40 m. This shallow depth is supported by the rather uniform fauna dominated by the epiphyte *A. planorbis*.

The estimated paleo water depth is not given for the sample at 479.3 m, as the dominant species of the sample is *B. elongata*. This species lives deep in the sediment, where accumulated organic matter (OM) provides an ample food resource and is thus independent of the OM flux reaching the seafloor from surface water (Murray 2006).

Since soaking in hydrogen peroxide was not sufficient to disintegrate the samples from below 612 m, it was not possible to calculate the P/B ratio for the lower third of the interval studied. The *Anomia–Cardium–Pitaria–Ostrea* and *Algae–Ostrea–Pecten* assemblages observed between 612 and 642 m and at around 700 m, respectively (Selmecezi et al. 2004) suggest shallow water sedimentation for the 612–697 m interval.

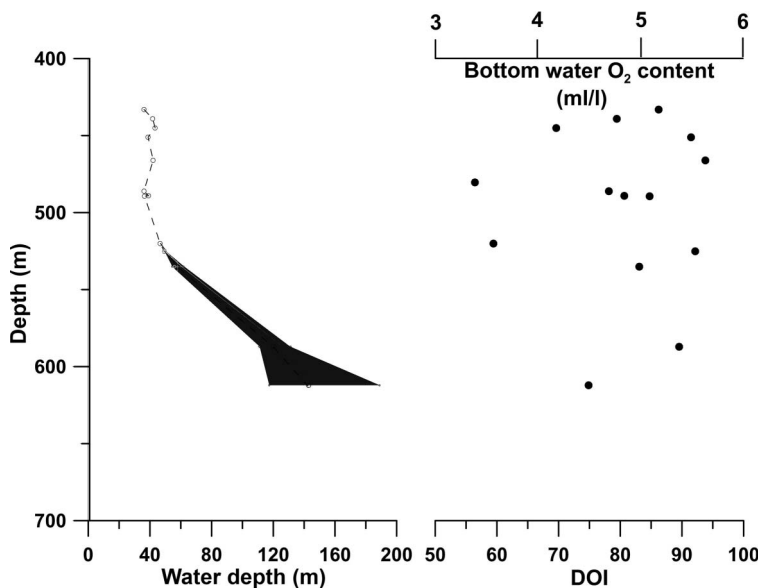


Fig. 3

Changes of water depth and bottom water oxygenation during deposition of the sediments penetrated in the 400–620 m depth interval, Ng 1 corehole. Above 550 m, the minimum and maximum water depth are practically identical. The darker belts represent the more probable depth interval. DOI: dissolved oxygen index of Kaiho (1994)

Oxygen content of the bottom water

The dissolved oxygen index (DOI) of Kaiho (1994) is used here for estimation of O_2 content expressed in ml/l. The DOI was calculated in all possible ways (DOI*1–4) supposing all undetermined specimens to be all oxic, or none of them oxic, all dysoxic or none of them dysoxic. Values obtained are listed in Supplementary Material B and displayed in Fig. 3. All DOI values are higher than 50, falling into the category of “high oxic” according to Kaiho’s classification (1994). Based on this proxy, the bottom was very well ventilated (minimum 3 ml/l dissolved O_2 content) during deposition of the sediments penetrated above 612 m.

The *Anomia–Cardium–Pitaria–Ostrea* assemblage observed in the 612–642 m interval (Selmecezi et al. 2004), the common presence of *Amussium* (Open File Report No. 1775/1) and the red alga containing limestone beds suggest that the sediments below 612 m were deposited in similarly oxygenated water.

OM and carbonate

The TOC content, Rock-Eval parameters, the organic and inorganic $\delta^{13}C$, and the carbonate content are listed in Table 1, while variations of some of these parameters along depth are displayed in Fig. 4.

Table 1
Organic geochemical data, isotope ratios, and carbonate content (n.d. = not determined)

Depth (m)	TOC (%)	T_{\max} (°C)	HC _{pot} (kg HC/t)	HI (mg HC/g TOC)	$\delta^{13}\text{C}_{\text{org}}$ (‰)	$\delta^{13}\text{C}_{\text{carb}}$ (‰)	Carbonate (%)
404.00	0.360	422	0.87	219	n.d.	n.d.	63.99
415.00	0.320	423	0.91	253	n.d.	n.d.	73.64
418.00	0.320	425	1.20	331	n.d.	n.d.	86.36
424.00	0.910	424	2.41	247	n.d.	n.d.	62.95
433.00	1.120	417	2.30	188	-20.10	0.80	60.91
436.00	1.420	422	2.45	162	n.d.	n.d.	66.82
439.00	1.320	426	3.65	256	n.d.	n.d.	66.36
445.00	0.240	400	0.66	231	-20.30	0.20	16.86
451.00	1.070	422	2.79	243	-19.80	0.90	67.73
454.00	0.850	422	2.01	220	n.d.	n.d.	81.99
460.00	1.040	425	2.52	222	n.d.	n.d.	73.64
466.00	0.940	427	2.93	288	-20.30	1.00	81.59
472.00	0.300	396	0.83	213	n.d.	n.d.	84.77
480.00	0.370	410	0.90	207	n.d.	n.d.	39.32
486.00	1.160	428	3.26	262	-19.30	1.70	74.55
489.00	0.950	423	3.53	347	-20.20	1.50	76.14
492.00	0.990	424	3.51	330	n.d.	n.d.	80.00
495.00	0.860	426	3.76	410	n.d.	n.d.	78.86
497.00	0.620	425	2.76	412	n.d.	n.d.	84.77
520.00	1.360	420	3.61	247	-17.70	1.90	77.27
525.00	0.860	429	3.75	408	-19.00	1.60	78.41
535.00	0.780	425	2.59	310	-17.50	1.80	71.82
545.00	0.330	428	2.97	806	n.d.		91.99
550.00	1.020	423	3.47	316	-19.71	0.60	85.68
555.00	0.360	421	1.01	256	-20.49	0.13	95.00
565.00	0.880	420	2.80	292	-20.86	0.55	81.99
575.00	1.430	421	4.76	306	-19.70	0.59	87.95

Table 1 (continued)

Depth (m)	TOC (%)	T_{\max} (°C)	HC _{pot} (kg HC/t)	HI (mg HC/g TOC)	$\delta^{13}\text{C}_{\text{org}}$ (‰)	$\delta^{13}\text{C}_{\text{carb}}$ (‰)	Carbonate (%)
583.00	1.150	419	3.61	286	-20.20	0.45	82.73
587.00	1.600	427	5.65	331	n.d.	n.d.	83.86
591.30	0.060	403	0.20	262	n.d.	n.d.	98.99
596.00	0.800	423	4.66	532	n.d.	n.d.	83.41
608.00	1.360	419	4.05	272	-19.95	0.94	83.86
612.00	1.420	423	3.97	258	-20.90	0.70	72.95
620.00	1.250	423	4.20	308	n.d.		81.59
625.00	0.950	416	2.45	233	-21.09	0.62	77.73
645.00	1.550	418	4.26	249	-20.98	0.92	66.14
648.60	0.070	364	0.13	96	n.d.		44.32
655.00	1.120	414	2.17	175	-21.81	1.00	62.27
665.00	0.280	389	0.68	158	n.d.	n.d.	27.50
675.00	1.930	422	7.02	332	n.d.	n.d.	63.99
680.00	1.600	419	4.15	235	-20.86	0.20	68.64
691.00	1.760	417	5.35	278	n.d.	n.d.	70.45
697.00	0.700	419	1.92	252	n.d.	n.d.	55.91

The amount of carbonate expressed in CaCO_3 varies mostly between 40% and 95%. It increases upward in the lower 120 m of the section, then decreases in its upper part. carbonate content shows a high scatter in the lower 120 m and the upper 100 m. Some samples, lying beyond the above depth trends, contain only 20–30% carbonate.

TOC varying between 0.4% and 2% decreases upward with a great scatter in the lower half of the section, then does not show a definite depth trend (Fig. 4).

T_{\max} varying between 410 and 429 °C clearly shows that the kerogen is immature. HI shows a co-variance with the carbonate content and ranges between 150 and 350 mg HC/g TOC in the great majority of the samples; hence, they contain mostly Type II/III and Type II kerogen (Fig. 4).

The $\delta^{13}\text{C}_{\text{org}}$ ranges between -22‰ and -17.5‰ (Fig. 4); in the lower half of the section, it moderately increases upward. Then, after a shift of 2.2‰ between 555 and 520 m, it shows a marked decrease.

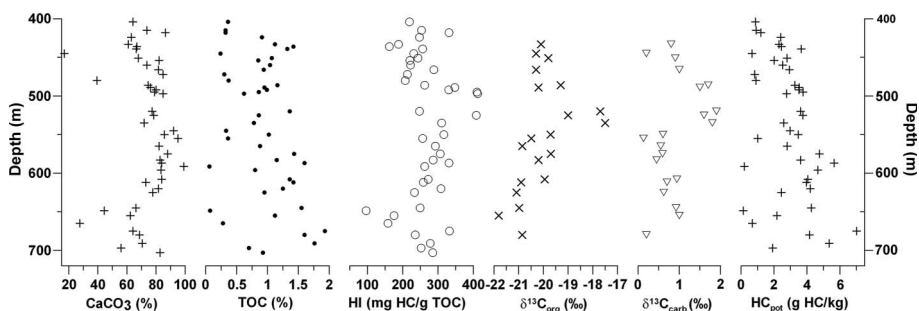


Fig. 4 Variations with depth of carbonate content, Rock-Eval indices, HC_{pot}, and organic and inorganic C-isotope ratios, 400–700 m depth interval, Ng 1 corehole

The $\delta^{13}\text{C}_{\text{carb}}$ ranges between 0.13‰ and 1.90‰; in the lower half of the section, it slowly decreases upward. Then, after a 1.2‰ jump between 555 and 520 m, it shows a marked decrease (Fig. 4).

Hydrocarbon potential

A significant part of the samples shows HC_{pot} and TOC values above 3 kg HC/t and 1%, respectively (Fig. 4). According to Peters and Cassa (1994), these samples represent fair oil source rocks.

Taking into account that for carbonate oil source rocks 0.4% is the lower limit of TOC content (Palacas 1978), an even higher portion of the samples, being mostly in the carbonate dominated 625–454 m depth interval, represents fair or even good oil source rocks.

Discussion

Relative weights of the kerogen precursors

The relatively high HI values – they are below 200 mg HC/g TOC in only 10% of the samples, while in more than 30% of them they are above 300 mg HC/g TOC – suggest that in the studied section, marine OM is at least one of the main sources of the kerogen. The common occurrence of epiphytic foraminifera living on benthic algae (see Paleo Water Depth subsection) confirms that beyond phytoplankton, benthic algae have also contributed to some degree to the kerogen. On the other hand, in view of the proximity of the dry land, some admixture of terrestrial plant matter must also be assumed. Pollen and terrestrial plant debris observed in significant amount in the palynological concentrates of four samples studied from the 400–700 m depth interval support this premise (Vetó 1973).

Isotopic studies carried out on recent plants (Deines 1980; Meyers 1994; Reuning et al. 2005) revealed that marine benthic plants, marine phytoplankton, and land plants typically exhibit $\delta^{13}\text{C}_{\text{org}}$ values between -12‰ and -5‰ , -22‰ and -20‰ , and -26‰ and -25‰ , respectively.

Although the massive contribution of isotopically very light terrestrial plant matter is obvious, $\delta^{13}\text{C}_{\text{org}}$ is less negative than -22‰ in all of the samples studied and even less negative than -20‰ in some of them. This finding strongly suggests that the isotopically very heavy benthic algal material was an important kerogen precursor.

In the following, a semi-quantitative estimation of the benthic algal contribution to the kerogen is attempted. Since no Rock-Eval and $\delta^{13}\text{C}_{\text{org}}$ data are available from immature Badenian sediments of the Zala Basin, corresponding data reported from nearby Austrian basins are used for this purpose.

According to Reischenbacher et al. (2007), the HI of the great majority of the 40 samples studied from an immature marine Badenian section of the Lavanttal Basin (Austria) is significantly lower than 100 mg HC/g TOC, and their kerogen is of predominantly terrestrial origin. Similarly, Wagreich et al. (2008) report HI ranging between 30 and 90 mg HC/g TOC from the immature marine Badenian of the Baden-Soos section, Vienna Basin, containing kerogen of predominantly terrestrial origin.

The average and range of $\delta^{13}\text{C}_{\text{org}}$ of 10 samples studied by Johns and Hoefs (1985) from the immature marine Badenian of the well Aderkla 78 (Vienna Basin) are -24.7‰ and -25.8‰ to -23.7‰ , respectively. The kerogen of these samples is also dominated by terrestrial components.

In the following, the kerogen of the Badenian samples studied from the Ng 1 core is considered as a mixture of terrestrial and marine end-members. For the calculations, 60 mg HC/g TOC and -24.7‰ will be used for HI and $\delta^{13}\text{C}_{\text{org}}$ of the terrestrial end-member, respectively, while 408 mg HC/g TOC, the greatest HI found on the Ng 1 samples analyzed for $\delta^{13}\text{C}_{\text{org}}$ (Table 1), will be used for HI of the marine one.

Applying a simple mixing model (Fig. 5a), we have determined that the marine end-member represents 35–75% of the kerogen in the great majority of the Ng 1 samples, studied for $\delta^{13}\text{C}_{\text{org}}$.

In the second step, we assessed the $\delta^{13}\text{C}_{\text{org}}$ of the marine end-member, using in the calculations -24.7‰ for $\delta^{13}\text{C}_{\text{org}}$ of the terrestrial end-member and the relative weights of the marine end-member obtained above. The obtained values range between -19.2‰ and -11.7‰ .

Supposing that benthic algal and planktonic components of the kerogen have the same HI, and using -8.5‰ and -21‰ for their $\delta^{13}\text{C}$, we can assess their relative weights in the marine end-member. We have found that benthic algal components represent 20–55% of the marine end-member of the kerogen in the great majority of the Ng 1 samples, studied for $\delta^{13}\text{C}_{\text{org}}$ (Fig. 5b). We believe that in spite of the obvious simplifications applied in the above calculations, the obtained results give a good idea of the relative weight of benthic algal material in the marine, oil-forming part of the kerogen.

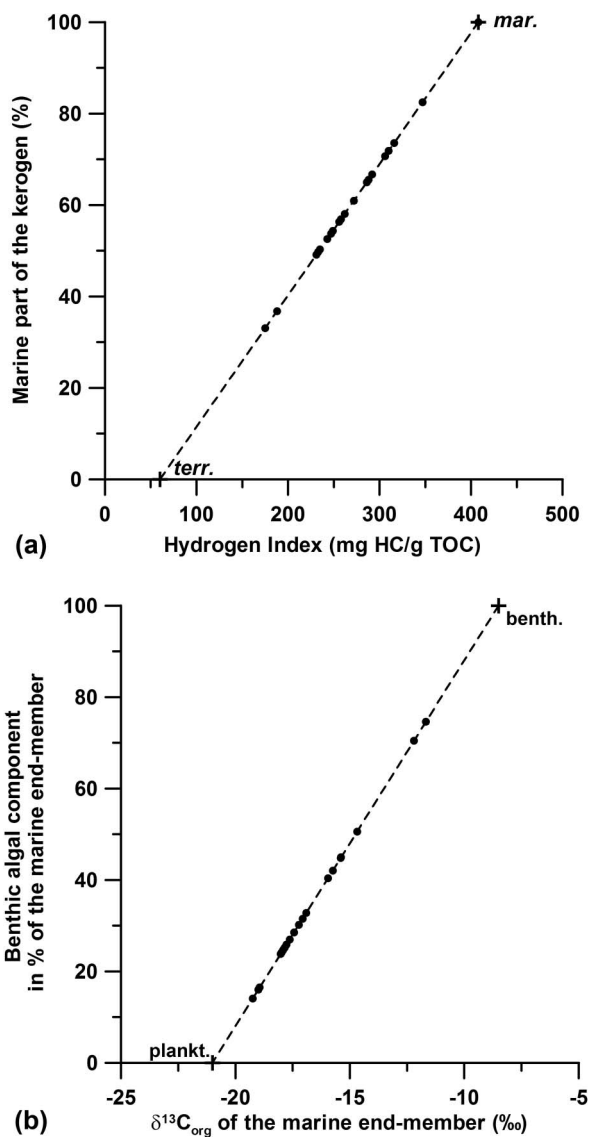


Fig. 5
 (a) Amount of marine components expressed in percentage of kerogen (dashed line: mixing line of the terrestrial and marine kerogen end-members). (b) Amount of benthic algal component expressed in percentage of marine kerogen (dashed line: mixing line of the benthic algal and planktonic kerogen end-members)

The depositional environment and its control on carbon isotopy and preservation of the kerogen

Planktonic productivity, bottom water O₂ content, and mass accumulation rate (MAR) are considered as of prime importance for the formation of oil source rocks (see [Littke et al. 1997](#) and references therein).

Since sediments of the 700–450 m depth interval were deposited from oligotrophic water, poor in nutrients (see above), planktonic productivity was certainly low during the corresponding time.

The bottom water was rich in O₂ during deposition of the sediments of the 700–400 m depth interval (see Oxygen Content of the Bottom Water subsection).

MAR was calculated for three parts of the studied section using nannoplankton zone/subzone ages documented in the section on Biostratigraphy and 2.17 g/cm³ for dry density of the sediments ([Dudko and Ódor 1980](#)). The obtained values, expressed in weight of dry sediment deposited on a unit area of bottom by time, are listed in Table 2.

High O₂ content of the bottom water is commonly considered as enhancing degradation of the freshly deposited OM (e.g., [Canfield 1994](#) and references therein) and the oligotrophic conditions prevailing during deposition of the bulk of the section studied suggest a relatively low flux of planktonic remains to the bottom. Hence, the trophic status of the sea and the redox conditions of the bottom both worked against the formation of oil source rocks.

On the other hand, the high MAR (Table 2) was certainly favorable for preservation of the freshly deposited OM (e.g., [Canfield 1994](#) and references therein) and in this way, for formation of oil source rocks. In view of the high carbonate content of the studied section (Table 1), the documented high MAR implies high rate of carbonate deposition as well. It is worth noting that the intense benthic algal life was certainly accompanied by intense carbonate precipitation, thus enhancing the preservation of the OM.

Table 2
Mass accumulation rates during the NN6 and NN5 nannoplankton zones

Nannoplankton (zone/subzone)	Depth interval (m)	Age boundaries (Ma)	Duration (Ma)	Average SR (m/Ma)	Average MAR (t/m ² /Ma)
NN6	454–350.7	13.53–12.7*	Max. 0.83	Min. 124	Min. 269
Upper part of NN5	665–454	14.36–13.53	0.83	254	551
<i>H. waltrans</i>	747–665	14.91–14.36	0.55	149	323

*12.7 Ma is the age assigned to the beginning of Sarmatian regional stage. Since the contact between Sarmatian and Badenian is of discordant nature (see Geologic Setting section), it is probable that sediments penetrated between 454 and 350.7 m do not represent the entire NN6; hence, the corresponding average SR and MAR are likely minimum values

The isotopic composition of the dissolved inorganic carbon (DIC) obviously exerted strong control on that of the phytoplankton and the benthic algae. On the other hand, it is commonly admitted that $\delta^{13}\text{C}$ of the syngenetic carbonate closely reflects isotopic composition of the DIC.

In spite of some admixture of diagenetic carbonate (e.g., foraminifera lining; see Preparation and Determination of Foraminifera subsection), the bulk of the high carbonate content of the studied samples is certainly of syngenetic origin, preserving the isotopic signal of the DIC. Hence, variation of $\delta^{13}\text{C}_{\text{carb}}$ along the section reflects changes of the isotopic composition of DIC occurred during its deposition. For this reason, an analysis of the relationship between depth trends of $\delta^{13}\text{C}_{\text{carb}}$ and $\delta^{13}\text{C}_{\text{org}}$ (Fig. 4) is promising for a better understanding of the cause(s) of variation observed in carbon isotopic composition of the kerogen.

$\delta^{13}\text{C}_{\text{carb}}$ shows a $<1\%$ upward decrease between 655 and 555 m, suggesting that a similar decrease of the DIC $\delta^{13}\text{C}$ took place during deposition of the corresponding sediments. On the other hand, $\delta^{13}\text{C}_{\text{org}}$ shows an about 1.5% upward increase in the same depth interval.

For interpreting these opposite isotopic trends, at least two factors must be taken into account:

- (a) The slight upward increase of HI in the 655–555 m depth interval (Fig. 4) makes likely an upward decreasing contribution of the isotopically relatively light land plant components to the OM.
- (b) The shallowing of the sea during deposition of the sediments of the 612–555 m depth interval (Fig. 3) made progressively easier the access of sunlight to the bottom, resulting in an intensification of the benthic photosynthesis. Hence, an upward increasing contribution of the isotopically very heavy benthic algal matter can be expected in the corresponding depth interval.

The enrichment of DIC in ^{12}C was not able to counterbalance the overall isotopic effect of these two factors.

The $\delta^{13}\text{C}_{\text{carb}}$ excursion between 555 and 520 m records an approximately 1.2% enrichment of the DIC in ^{13}C . The 2% increase of $\delta^{13}\text{C}_{\text{org}}$ observed in this depth interval is at least partly caused by this shift in DIC isotopic composition. In this depth interval, HI does not show a clear trend (Fig. 4); the cause of the difference between the organic and inorganic carbon isotope shifts is unknown. We note that Báldi (2006) and Gonera et al. (2000) studying the carbon isotope composition of foraminifera from sediments deposited during the NN5 biozone in two sections of the Central Paratethys, did not find such an excursion. Hence, this was a local phenomenon and its interpretation is beyond the scope of this work.

Finally, the well pronounced upward decrease of $\delta^{13}\text{C}_{\text{carb}}$ above 520 m is accompanied by an even stronger upward decrease of $\delta^{13}\text{C}_{\text{org}}$. The difference between the organic and inorganic carbon isotope shifts can be explained by an upward

increasing relative weight of the isotopically light land plant matter, suggested by the upward decrease of HI (Fig. 4).

Regional significance

Holzhaecker et al. (1981) report that the third of a sample set (70 samples), representing the marine Middle Miocene of the Hungarian part of the PBS, contains kerogen with $\delta^{13}\text{C}$ ranging between -22‰ and -18‰ or even less negative. According to the same authors, the $\delta^{13}\text{C}$ of the chloroform-soluble bitumen extracted from the third of the Middle Miocene marine sediments (60 samples) studied from Hungary is less negative than -24‰ . Hence, the very heavy organic C-isotopic composition of the Ng 1 Badenian sediments is not a pure curiosity, and benthic algae likely are important kerogen precursors in the marine Middle Miocene of the PBS.

Koncz (1990) reports that some oils of the Zala Basin are characterized by high pristane/phytane ratio (1.95–3.46) and relatively heavy C-isotopic composition ($\delta^{13}\text{C}$ of the saturate fraction ranges between -23.5‰ and -21‰). Based on these indices and by making comparison with extracts of marine Middle Miocene sediments, Koncz (1990) and Clayton and Koncz (1994) think that the latter are the likely source rocks of the oils.

Since pristane/phytane ratio above 1 is commonly considered as an index for deposition from oxic bottom water and the very heavy C-isotopic composition of the saturate fraction suggests that benthic algal matter acted as source for the oils, we think that Middle Miocene source rocks, deposited under similar conditions as the Ng 1 Badenian sediments, produced oil in commercial quantity in the Zala Basin.

A novel approach for recognition/estimation of benthic algal contribution to kerogen

Due to the poor fossilization potential of the benthic algae, it is difficult to study them in sedimentary rocks by microscopic techniques. As far as we know the significant contribution of benthic algae to kerogen has been suggested only for Jurassic and Miocene source rocks of some northwest Chinese basins. Li et al. (2012) speculated about the massive presence of benthic algae in the Miocene source rocks of the Tarim Basin, but their suggestion is not supported by data. Cao et al. (2009) reported UV-microscopic observations of red algal material and elevated concentration of the diahopane biomarker as independent lines of evidence for the contribution of benthic algae to the kerogen of different Jurassic source rocks of the Qaidam Basin.

The original goal of this study was to contribute to a better understanding of the depositional environment of the Miocene source rocks in the PBS. For this purpose, a new approach was followed using combined micropaleontological, organic geochemical, and isotopic techniques.

In the course of the work, we found that if we use these independent techniques, especially the combined study of benthic foraminifera and measurement of $\delta^{13}\text{C}_{\text{org}}$, we are able to estimate, at least semi-quantitatively, the weight of the benthic algal material in the kerogen.

Our combined paleontological, organic geochemical, and isotopic results provide the first sound demonstration of major benthic algal contribution to oil-prone kerogen. This simple new multi-proxy approach seems to be promising for a better understanding of the formation of oil source rocks in carbonate-dominated shelf environment.

Conclusions

Benthic algae represent 20–55% of the marine kerogen in the Badenian section studied from the Ng 1 core. Benthic algae are important kerogen precursors in effective oil source rocks in the Middle Miocene of Hungary. The combined study of benthic foraminifera, carbon isotopic composition of kerogen, and Rock-Eval indices is a powerful multi-proxy approach for recognition/assessment of contribution of benthic algae to kerogen.

Acknowledgements

This study was funded by grants of the Hungarian Scientific Foundation (OTKA T 032260 and D 42191). Discussions with and comments of Mária Bálđi-Beke, Margit Bohn-Havas, András Németh, and Ildikó Selmeczi are gratefully acknowledged. We would like to thank two anonymous reviewers for suggestions and remarks.

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