

Upper Cretaceous/lower Tertiary black shales near the North Pole: Organic-carbon origin and source-rock potential

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Abstract

During IODP Expedition 302 (Arctic Coring Expedition—ACEX), the first scientific drilling campaign in the permanently ice-covered central Arctic Ocean, a 430 m thick sequence of upper Cretaceous to Quaternary sediments has been drilled. The lower half of this sequence is composed of organic-carbon-rich (black shale-type) sediments with total organic carbon contents of about 1–14%. Significant amounts of the organic matter preserved in these sediments is of algae-type origin and accumulated under anoxic/euxinic conditions. Here, for the first time detailed data on the source-rock potential of these black shales are presented, indicating that most of the Eocene sediments have a (fair to) good source-rock potential, prone to generate a gas/oil mixture. The source-rock potential of the Campanian and upper Paleocene sediments, on the other hand, is rather low. The presence of oil or gas already generated in situ, however, can be ruled out due to the immaturity of the ACEX sediments.

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1. Introduction and background

The present-day Arctic Ocean is fringed by several large sedimentary basins (e.g., North Alaska Slope, Arctic Canada, the Barents Shelf and western Siberia) that are highly productive in terms of gas and oil (Fig. 1). The hydrocarbons known from these circum Arctic basins probably derived from organic-carbon (OC)-rich mudrocks (“black shales”), which were deposited during Mesozoic and early Cenozoic times (Dixon et al., 1992; Leith et al., 1992; Bakke et al., 1998; Littke et al., 1999; Vyssotski et al., 2006). In the Barents Sea and northern North Atlantic off Norway, for example, organic-geochemical data from petroleum exploration drill holes representing Jurassic/Cretaceous time intervals indicate that these black shales are characterized by very high OC contents (typically 15–25% with peak values of >30%) of a mixed marine/terrigenous origin, probably related to anoxia and/or increased primary production (Leith et al., 1992; Langrock

et al., 2003; Langrock and Stein, 2004). For the central Arctic Ocean, however, in only four short cores (FL533, FL437, FL422, and CESAR-6; Fig. 1) obtained by gravity coring from drifting ice flows on the Alpha Ridge, some older pre-Neogene sediments were recovered. The upper Cretaceous sediments from the Alpha Ridge consist of OC-rich black mud containing immature, mixed terrigenous-marine type of organic matter, and almost pure laminated siliceous oozes with excellently preserved diatoms and silicoflagellates (Jackson et al., 1985; Clark et al., 1986; Dell’Agnese and Clark, 1994; Firth and Clark, 1998). In general, these data suggest a warmer (ice-free) Arctic Ocean with strong seasonality and high paleoproductivity, most likely associated with upwelling conditions (Clark et al., 1986; Jenkyns et al., 2004). Differences in sediment composition between the cores may have been caused by lateral and temporal nutrient conditions, oceanic currents, bottom-water oxygen levels, and basin topography (Firth and Clark, 1998).

In August 2004, an almost 430 m thick, Quaternary to late Cretaceous sedimentary sequence was drilled on the Lomonosov Ridge close to the North Pole (Fig. 1) during

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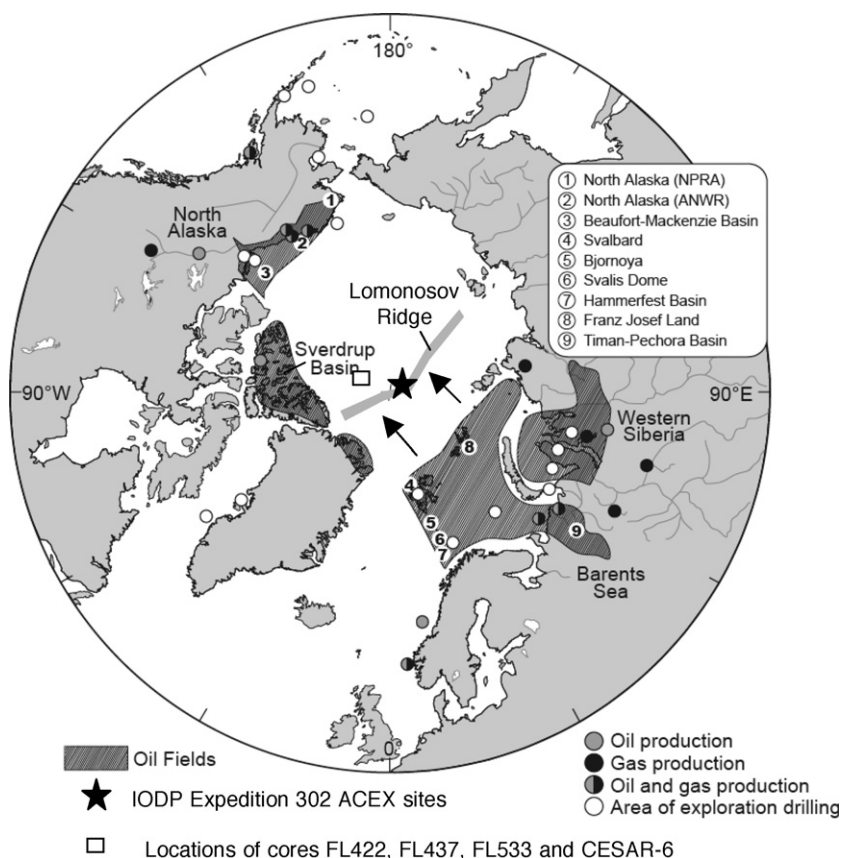


Fig. 1. Distribution of major circum-Arctic oil and gas fields (Bakke et al., 1998) and the location of IODP Expedition 302 ACEX sites. The area where the four short sediment cores containing older pre-Neogene sediments, were recovered, are also shown. Black arrows indicate the drift of Lomonosov Ridge, separated from the Eurasian continental margin near 56 Ma (Kristoffersen, 1990).

the IODP Expedition 302 (Arctic Coring Expedition—ACEX), the first scientific drilling in the permanently ice-covered central Arctic Ocean (Backman et al., 2006; Moran et al., 2006). Cores were recovered in five holes across three sites (Holes MSP0002A, MSP0003A, MSP0004A, MSP0004B, and MSP0004C) situated within 15.5 km of each other on seismic Line AWI-91090 (Jokat et al., 1992) and interpreted as a single site (composite section) because of the internally consistent seismic stratigraphy across that distance. Unfortunately, major hiatuses occur at about 200 mcd (meters composite depth) and 405 mcd, i.e., the upper middle Eocene to early Miocene and the upper Campanian to lower late Paleocene time intervals, respectively, are missing in the ACEX record (Backman et al., 2006; Moran et al., 2006). The lower 230 m of the ACEX sequence consist of very dark gray biosiliceous oozes and mudstones (“black shales” in a broader sense) of Campanian and Paleogene (late Paleocene to middle Eocene) age, which are distinctly enriched in OC (Fig. 2). The stratigraphy of this lower part of the ACEX sequence is based on dinocysts (Backman et al., 2006).

Directly after the recovery of these OC-rich sediments during the ACEX expedition, speculation started on “maybe oil underneath all the ice” (Revkin, 2004). With the new Rock-Eval records presented here, real facts on the source-rock potential of the Campanian/Paleogene central Arctic

Ocean sediments become available for the first time. All data are available in [doi:10.1594/PANGAEA.552044](https://doi.org/10.1594/PANGAEA.552044). More sophisticated studies using gas chromatography (GC) and gaschromatography/mass spectrometry (GC/MS) techniques as well as organic petrography, are in process to get more detailed information about the depositional environment as well as source-rock characteristics.

2. Methods

Total organic carbon (TOC) content and Rock-Eval parameters were determined on bulk ground samples to get information about the source-rock potential, presence of hydrocarbons, and maturity. TOC was determined by LECO elemental analyzer technique. Measured Rock-Eval parameters are (1) the amount of hydrocarbons already present in the sample (S1 peak in mg hydrocarbons per gram sediment), (2) the amount of hydrocarbons generated by pyrolytic degradation of the kerogen during heating of up to 550 °C (S2 peak in mg hydrocarbon per gram sediment), (3) the amount of carbon dioxide generated during heating of up to 390 °C (S3 peak in mg carbon dioxide per gram sediment), and (4) the temperature of maximum pyrolysis yield (T_{max} value in °C) (Espitalié et al., 1977; Tissot and Welte, 1984; Peters, 1986). As indicator for the composition of the organic matter

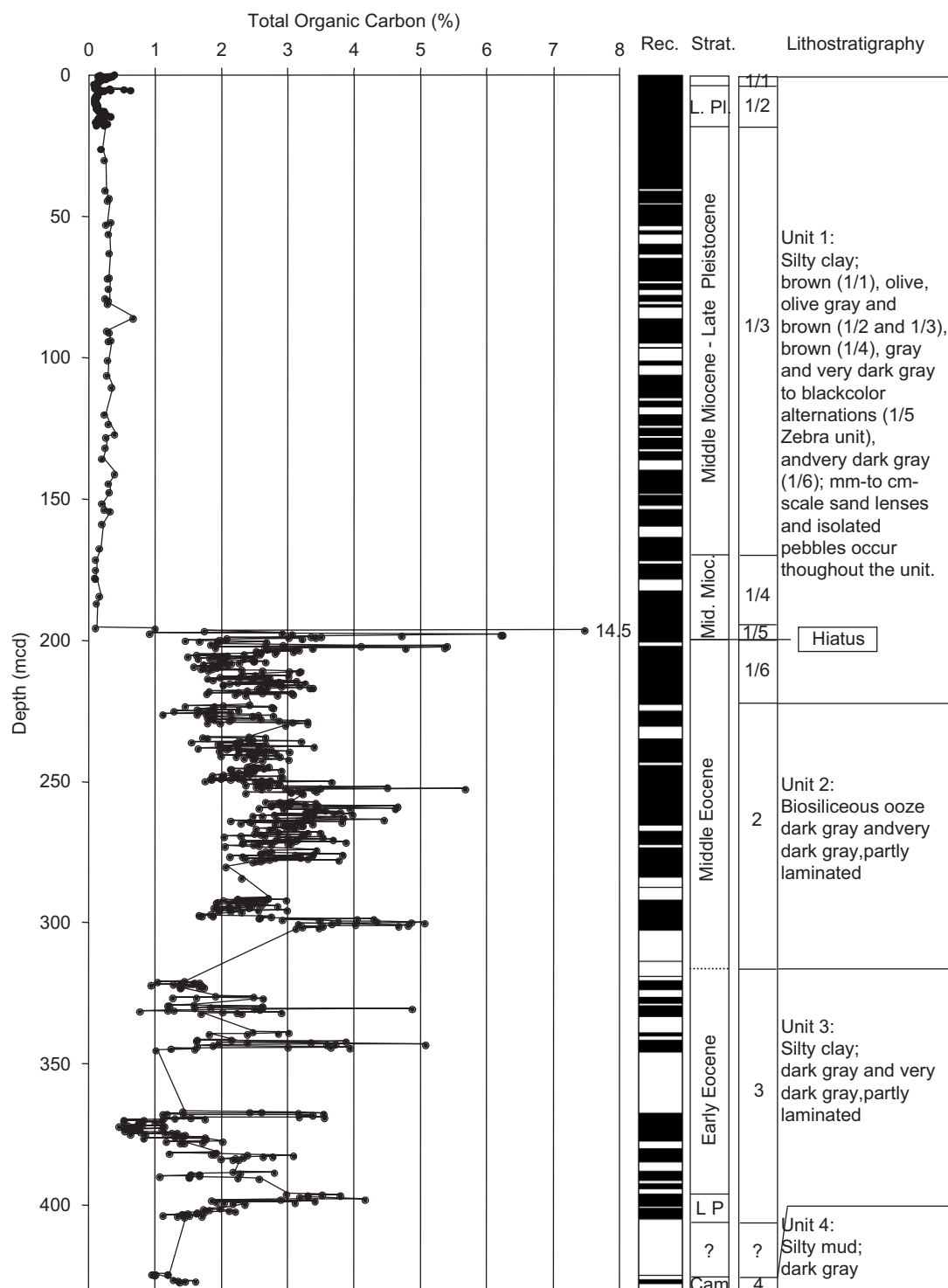


Fig. 2. Record of total organic carbon (TOC) contents as determined in the composite ACEX sedimentary sequence (0–17.85 mcd: Hole MSP0004C; 18–265.23 mcd: Hole MSP0002A, and 265.34–427.57 mcd: Hole MSP0004A). Data on recovery, stratigraphy, and lithological units (1–4) and subunits (1/1–1/6) from Backman et al. (2006). Cam = Campanian; LP = late Paleocene; Mid Mioc. = middle Miocene; L.Pl. = late Pleistocene.

(kerogen type), hydrogen (HI) and oxygen index (OI) values were calculated and shown in a “van-Krevelen-type” diagram (see Fig. 3). The HI value corresponds to the quantity of pyrolyzable hydrocarbons (S_2) per gram TOC ($\text{mg HCg}^{-1}\text{C}$), the OI value corresponds to the quantity of

carbon dioxide (S_3) per gram TOC ($\text{mg CO}_2\text{g}^{-1}\text{C}$). The S_1 , S_2 , S_3 , S_2/S_3 , $S_1 + S_2$ (Genetic Potential), and $S_1/(S_1 + S_2)$ (Production Index) values give further information about the OC source, source-rock potential, presence of hydrocarbons, and maturity (for further details see

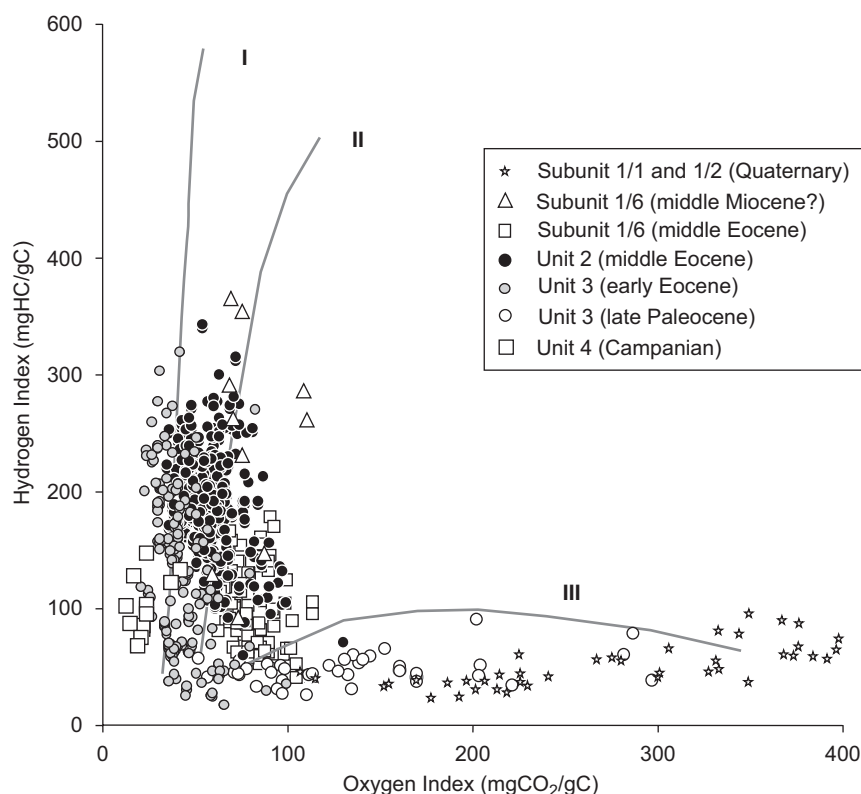


Fig. 3. Hydrogen index versus oxygen index (van-Krevelen-type) diagram of the Campanian, Paleogene, and Quaternary intervals of the ACEX record (Units 4, 3, and 2; Subunits 1/6, 1/5, 1/2, and 1/1), indicating the main kerogen types: type I/II is of aquatic algae and/or microbial biomass origin, type III is of terrigenous higher plant origin. A large number of samples from Subunits 1/1 and 1/2 have oxygen index values $>400 \text{ mg CO}_2\text{g}^{-1} \text{ C}$ and thus fall out of the plotted area (see data set in [doi:10.1594/PANGAEA.552044](https://doi.org/10.1594/PANGAEA.552044)).

Tissot and Welte, 1984; Peters, 1986). The T_{max} value from Rock-Eval pyrolysis can be used as maturity indicator. T_{max} values $<435^\circ\text{C}$ is an indication for fresh immature organic matter, whereas T_{max} values $>435^\circ\text{C}$ point to the presence of more mature and/or refractive organic matter.

The reflectance of the maceral vitrinite in reflected light (R_o) was used as additional maturity indicator, with mean R_o values $<0.5\text{--}0.7\%$ indicating immature and R_o values between $0.5\text{--}0.7\%$ and 1.3% indicating mature (main zone of oil formation) organic matter (Tissot and Welte, 1984).

3. Results and discussion

3.1. Amount, origin, and accumulation of OC

Whereas the upper (middle Miocene to Quaternary) part of this sequence is composed of silty clay with very low OC contents of $<0.5\%$, i.e., values very similar to those known from upper Quaternary records determined in gravity cores from Lomonosov Ridge (Stein et al., 2004), the Campanian and Paleogene sediments of the ACEX sequence are characterized by high TOC values of 1 to $>5\%$ (Fig. 2). In Subunit 1/5 (195–200 mcd; middle Miocene?) characterized by distinct gray/black color bandings, even TOC maxima of 7–14.5% TOC were measured in samples from the black horizons (Fig. 2).

The high abundances of biosiliceous microfossils and fine-scaled lamination typical for the Eocene part of the sequence (Backman et al., 2006) as well as the preservation of hydrogen-rich (algae-type) organic matter (kerogen types I/II and mixed II/III; Fig. 3) point to a high-productivity and anoxic paleoenvironment to be dominant in the middle Eocene and in parts of the early Eocene. Anoxia is also supported by excess (pyritic) sulfur values resulting in very low organic carbon/sulfur (C/S) ratios of <1 (cf., Leventhal, 1983), and the occurrence of small-sized framboid pyrite, determined in the lower/middle Eocene ACEX sedimentary section (Stein et al., 2006). In the late Paleocene interval, on the other hand, the organic matter is mainly of terrigenous origin (kerogen type III; Fig. 3). Due to oxic environmental conditions indicated by high C/S ratios and the absence of small-sized pyrite framboids (Stein et al., 2006), significant amount of algae-type organic matter is not preserved in the sediments. Based on the Rock-Eval data, the composition of the organic matter of the upper Paleocene sediments is similar to that of Quaternary sediments on Lomonosov Ridge (Fig. 3). The Campanian sediments deposited in a shallow-water neritic environment (Backman et al., 2006), probably contain high amount of reworked organic matter. Biomarker as well as organic petrography studies on ACEX material are in process to get more detailed

information on OC sources and paleoenvironmental conditions.

Taken into account the high TOC values of the Eocene interval of the ACEX section and the fact that sedimentation rates at that time were similar (or even twice) than during Holocene times (Stein et al., 2004; Backman et al., 2006), Eocene OC accumulation rates were 5–20 times higher than modern ones. Whereas very low OC accumulation rates of about $0.005 \text{ g C cm}^{-2} \text{ ky}^{-1}$ are typical for the modern (Holocene) central Arctic Ocean on Lomonosov Ridge (Stein et al., 2004), values of up to $0.1\text{--}0.15 \text{ g C cm}^{-2} \text{ ky}^{-1}$ were reached during Eocene times. Based on the correlation between accumulation rates of (marine) OC and primary productivity in recent anoxic environments, Bralower and Thierstein (1984) estimated that about 2% of the primarily produced OC is preserved and accumulated in the sediments under anoxic conditions (for more detailed discussion of estimates of primary productivity from sediment data in different environments see Stein, 1991). Using Bralower and Thierstein's approach for an Eocene anoxic Arctic Ocean, primary productivity

may have reached values of $50\text{--}75 \text{ g C m}^{-2} \text{ y}^{-1}$, i.e., values $>5\text{--}10$ times higher than modern ones (Sakshaug, 2004).

3.2. Source-rock potential and hydrocarbon formation

High S_2 values of $5\text{--}10 \text{ mg HC g}^{-1} \text{ Sed}$ ($\text{kg HC t}^{-1} \text{ Sed}$) clearly indicate that most of the OC-rich middle Eocene sediments have a good source-rock potential whereas in the Campanian and upper Paleocene sediments the source-rock potential is low (Fig. 4). In the early Eocene the potential is also generally low, except for short intervals with higher potential similar to the middle Eocene. HI values $>200 \text{ mg HC g}^{-1} \text{ C}$ and S_2/S_3 ratios >2.5 suggest that the intervals with a good source-rock potential are prone to generate a gas/oil mixture when the level of thermal maturity ("oil window") is reached. T_{max} values $<435^\circ\text{C}$, however, indicate an immaturity of the organic matter, also supported by vitrinite reflectance (R_o) values $<0.5\%$ (Fig. 4).

Although absolute values are depending on the source-rock potential, S_1 and $S_1/(S_1 + S_2)$ values generally

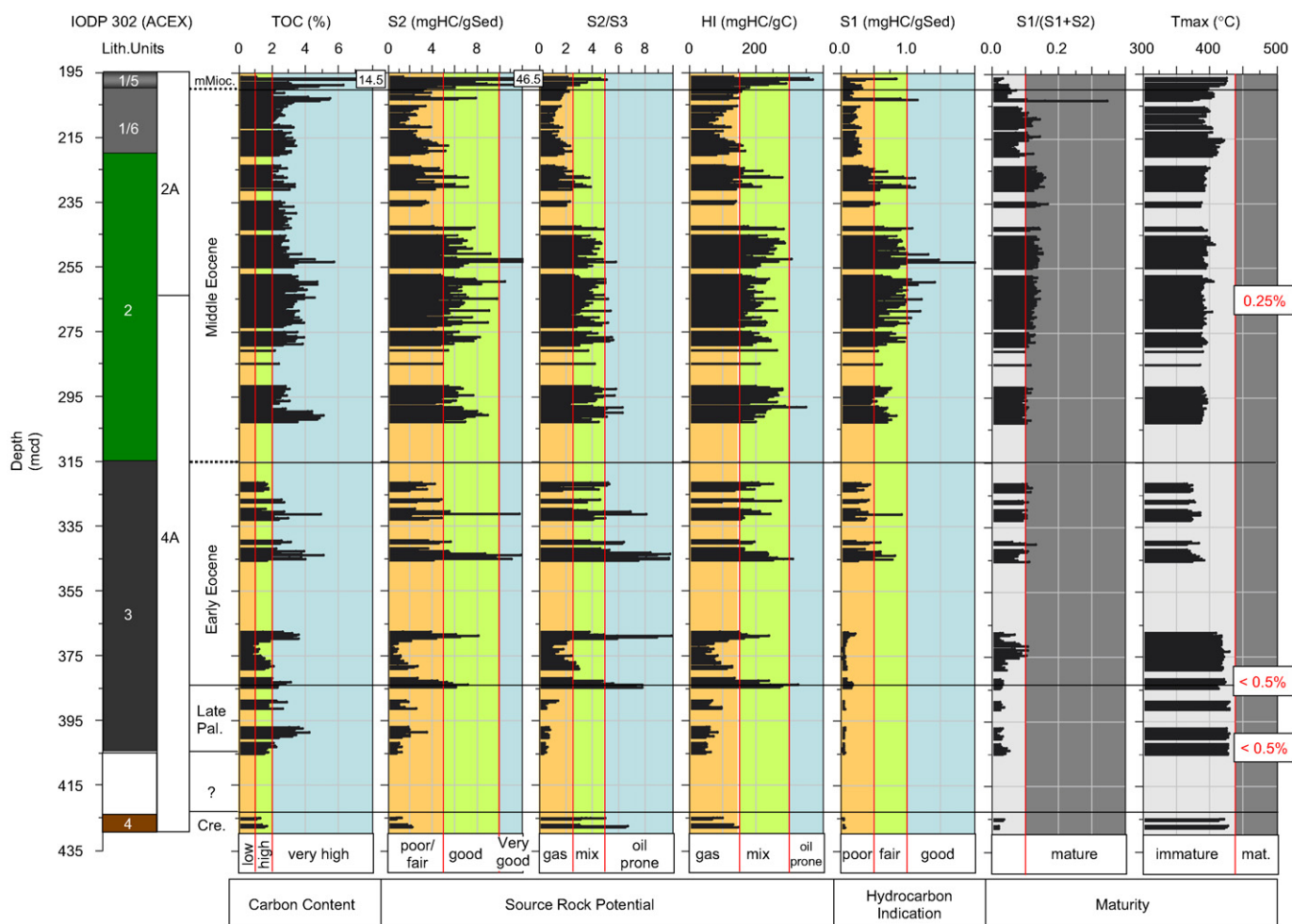


Fig. 4. Total organic carbon (TOC) content and Rock-Eval parameters S_1 , S_2 , S_3 , S_2/S_3 , $S_1/(S_1 + S_2)$, hydrogen index (HI), and T_{max} determined in lower 230 m of the ACEX drill site (Holes MSP0002A and MSP0004A), and interpretation in terms of source-rock potential, hydrocarbon indication, and maturity according to Peters (1986). Red numbers are vitrinite reflectance values (from Stein et al., 2006). Left, the stratigraphy and the lithological units are shown (Backman et al., 2006). For further description of lithological units, see Fig. 2.

increase with increasing maturity and may be used as indication for the presence of hydrocarbons generated in situ and stage of maturity, respectively (e.g., Tissot and Welte, 1984; Peters, 1986). The ACEX record, however, shows a different picture. High $S1$ and $S1/(S1 + S2)$ values occur in the middle part of Unit 2, and minimum values were determined in the lower part of the record (Fig. 4). This as well as the low maturity based on $T_{\max}$ and R_o values contradict an in-situ generation and may suggest that hydrocarbons were generated in other more mature sequences and transported to this site where the hydrocarbons accumulated. The biosiliceous sediments of Unit 2 have a higher porosity than the overlying silty clays of Unit 1/6 (Backman et al., 2006), which may support this explanation. The maximum $S1$ values, however, do not occur on top of Unit 2. Furthermore, the $S1$ maximum interval coincides with maximum TOC and $S2$ values, and a very low maturity (R_o of 0.25% and $T_{\max}$ values $<400^\circ\text{C}$). Thus, in Unit 2 characterized by maximum occurrence of aquatic OC, major amount of very labile OC is probably present, already cracked by low temperatures during pyrolysis and contributed to the $S1$ peak. Such a situation is often observed in young (Quaternary) immature sediments (e.g., Dean et al., 1994; Wagner and Dupont, 1999). Peters (1986) also mentioned that pyrograms of very immature sediments may show poorly separated $S1$ and $S2$ peaks, which can give anomalous (too high) $S1$ and $S1/(S1 + S2)$ values. Based on preliminary kerogen microscopy data (Stein et al., 2006), the presence of older reworked material which also could explain higher $S1$ values, is not indicated in this interval.

Based on these data, the presence of oil or gas already generated in situ from the Campanian and Paleogene ACEX sediments at this part of Lomonosov Ridge can be ruled out. If these sediments are buried more deeply, however, in-situ hydrocarbon formation is possible. This situation might occur in the more southern part of Lomonosov Ridge closer to the Eurasian continental margin, where sedimentation rates are significantly higher. Sedimentation rates at the ACEX sites area ($87^\circ56'\text{N}$, 140°E) are between about 1 and 3 cm ky^{-1} (Backman et al., 2006), but the rates increase to 3.5–6 cm ky^{-1} in Quaternary sediments recovered on Lomonosov Ridge at about 81°N (Polarstern Core PS2757-8, $81^\circ09.8'\text{N}$, $140^\circ12.0'\text{E}$, 1230 m water depth; Stein et al., 2001). The results of this study does also not mean that in the underlying deeper (Mesozoic) sedimentary rocks from the Lomonosov Ridge belonging to the rifted continental crustal block of the Eurasian continental margin (Kristoffersen, 1990; Jokat et al., 1992), hydrocarbons could not have been generated. This is even probable because, during pre-Tertiary times, the Lomonosov Ridge was part of the Eurasian continental margin with its large sedimentary basins filled with Mesozoic OC-rich sediments (Fig. 1). These sediments are the source rocks of the giant oil and gas fields explored in this area (e.g., Leith et al., 1992; Littke et al., 1999; Vyssotski et al., 2006).

Whether some of these hydrocarbons migrated into the overlying younger (ACEX) sediments can be proved by our future GC and GC/MS studies on ACEX material.

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