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Hydrocarbon potential and biomarker assemblages of Paleogene source rocks in the Cangdong sag, Bohai Bay Basin, China



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ABSTRACT

The organic geochemistry of the Paleogene-aged second and first members of the Kongdian Formation (Ek2 and Ek1) and the third member of the Shahejie Formation (Es3) in the Cangdong sag, Bohai Bay Basin, China, are characterized and the hydrocarbon generating potential is evaluated. The Ek₂ source rocks have a good to very good oil-generating potential, as indicated by the high abundance and good quality of organic matter within a mature stage. Although the Ek_1 mudstones have entered the oil window, they are organic lean and are dominantly composed of Type III kerogen. Accordingly, the Ek1 mudstones are considered to be poor source rocks. The Es₃ member is thermally mature and contains several intervals that have high contents of good-quality organic matter (mixed Type I and II kerogens), suggesting that these intervals also have a good potential for oilgeneration. Given this potential, new exploration plays sourced from the Es₃ member could be possible in the Cangdong sag. Biomarker distributions in the Ek2 and Es3 source rocks are both characterized by a dominance of medium- to high-molecular-weight *n*-alkanes (*n*-C₁₈ to *n*-C₂₉), a low sterane/hopane ratio, and a low diasterane abundance. The Ek₂ samples have low Pr/Ph ratios (< 1.0), relatively high gammacerane/ C_{30} hopane ratios (> 0.1) and β -carotane, trace amounts of C_{30} 4 α -methyl-24-ethlycholestanes, and a predominance of C_{29} regular steranes. Collectively, these characteristics indicate that abundant prokaryotic biomass mixed with some terrigenous organic matter was deposited under reducing conditions within a stratified water column. In contrast, the Es₃ source rocks are characterized by variable Pr/Ph ratios, low gammacerane and β -carotane, comparable C₂₇ and C29 regular steranes, and relatively high C30 4a-methyl-24-ethlycholestanes. These properties indicate that the Es₃ source rocks contain significant contributions from prokaryotic and algal sources, and were deposited under reducing to suboxic conditions within a relatively less stratified water column.

1. Introduction

The Cangdong sag is one of the most petroliferous regions within the Huanghua sub-basin, of the Bohai Bay Basin, China, where seven conventional oilfields, including two big oilfields (i.e., Zaoyuan and Wangguantun; with reserves > 1×10^8 tons), have been discovered (Ge et al., 2012; Xu et al., 2016). The basic geological and geochemical characteristics of the Cangdong sag have been previously documented (e.g., Li et al., 2006; Liu et al., 2010; Ge et al., 2012; Li et al., 2012; Liu and Jia, 2014; Xu et al., 2015; Pu et al., 2016). Specifically, the second member (Ek₂) of the Kongdian (Ek) Formation, and the third and first

member (Es₃ and Es₁) of the Shahejie (Es) Formation, were evaluated as potential source rock intervals. The Ek₂ member has previously been regarded as the primary source rock in the region, as it is organic rich with the largest thickness and most extensive distribution. Additionally, the Ek₂ member has a higher thermal maturity than either of the Es members, and preliminary oil-source rock correlations support the Ek₂ member as the primary source rock in the region (Liu, 2008). As such, previous regional petroleum exploration has focused on the Ek₂ member, while the Es Formation remains relatively unexplored, except for a few studies on the quantity, type, and thermal maturity of the organic matter in these potential source rocks (e.g., Liu et al., 2010; Ge

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et al., 2012; Xu et al., 2015). Indeed, minimal data on the molecular composition of organic matter has been reported with the exceptions of Ge et al. (2012) who observed slight pristane over phytane (Pr/Ph) predominance and predominate C_{29} regular steranes in the Ek source rock, and Xu et al. (2015) who observed low Pr/Ph ratios (< 1.0) for the Ek₂ source rocks.

The recent expansion of petroleum exploration to the Es Formation of the Cangdong sag has provided a rare opportunity to investigate the hydrocarbon potential and detailed biomarker distributions of this formation in order to elucidate the organic matter source and depositional characteristics. In this study, geochemical pyrolysis analyses were performed to re-evaluate the hydrocarbon generating potential of the Ek₂, Ek₁, and Es₃ members and provide a detailed assessment of new biomarker data generated for the potential source rocks. This work provides a critical assessment of the source rock potential, differences in organic matter sources and depositional environments across different source rock intervals, and diagnostic biomarker characteristics. The results may provide valuable information for future oil exploration in the Cangdong sag.

2. Geological setting

The Cangdong sag, which covers an area of 1.5×10^3 km², is located in the southern part of the Huanghua sub-basin of the Bohai Bay Basin (Fig. 1a–c). This northeast-trending sag is bounded by the Xuxi fault to the east, Cangdong fault to the west, Kongdian uplift to the north, and the Dongguang uplift to the south (Fig. 1c). The Cangdong sag can be divided into five distinct tectonic units, i.e., the (i) Kongdian tectonic belt, (ii) Shenvsi fault belt, (iii) Kongdong slope, (iv) Kongxi slope and (v) Nanpi slope (Fig. 1c; Pu et al., 2016).

The Cangdong sag underwent a syn-rift stage during the Paleogene and has been in a post-rift stage since the Neogene. From bottom to top, the stratigraphy consists of the Paleogene Ek, Es, and Dongying (Ed) formations, the Neogene Guantao (Ng) and Minghuazheng (Nm) formations, and the Quaternary Pingyuan (Qp) Formation (Fig. 1d). The Ek Formation unconformably overlies Mesozoic strata and is separated from the overlying Es Formation by another unconformity (Wang et al., 1987; Han, 2009). The Paleogene Ek, Es, and Ed Formations are each further subdivided into three respective members based on lithological changes. Stratigraphically from bottom to top the members are termed: Ek₃, Ek₂, and Ek₁; Es₃, Es₂, and Es₁; and Ed₃, Ed₂, and Ed₁, respectively (Fig. 1d). The detailed lithologies of each member have been previously described by Qu et al. (2008), so we focus on providing a detailed geochemical description of the following intervals:

- (1) The 400–600 m thick Ek_2 member deposited during the time of maximum flooding, which mainly consists of grayish-black shale intercalated with light-gray siltstone and dolomite.
- (2) The 1200–1500 m thick Ek_1 member, which has been further subdivided into six oil groups (Z0–ZV). From top to bottom the oil groups can be delineated as: Z0 and ZI near the top of Ek_1 that consist of thick, dark gypsum and red or green mudstones, respectively. Underlying these are the ZII, ZIII, ZIV, and ZV oil groups that are hosted in high-quality glutenite reservoirs.
- (3) The 200–400 m thick Es_3 member, the lower section of which is mainly composed of glutenite and arkose sandstones, while the middle and upper sections of the Es_3 member are characterized by a thick dark gray mudstone.

3. Samples and methods

Seventy-four core and cutting samples, obtained from nine wells (Fig. 1c) were used in this study. The samples include 35 from the Ek_2 member, 18 from the Ek_1 member, and 21 from the Es_3 member (Table S1, Supplementary data). To remove contamination, the samples were washed with deionized water, and then cleaned with a 9:1

dichloromethane methanol mixture (CH₂Cl₂:CH₃OH). The samples were finely ground (< 100 mesh) for programmed pyrolysis, total organic carbon (TOC) analysis, and Soxhlet extraction. Pyrolysis analyses were performed using an OGE rock pyrolyzer yielding results similar to the traditional Rock-Eval pyroanalyzer, including free hydrocarbons (S₁, mg HC/g rock), hydrocarbons generated by the decomposition of kerogen during pyrolysis (S₂, mg HC/g rock), and the temperature at maximum S₂ generation (T_{max}, °C). For determination of the TOC content, ~100 mg of each sample was decalcified with 5% HCl and analyzed on a Leco CS230 Determinator. Vitrinite reflectance (Ro, %) measurements were conducted on 12 samples (Table S1). The R_o measurements (random, oil immersion) were performed using a Leica DM4000M reflected light microscope equipped with a 25× objective and the Diskus Fossil System (Hilgers Technisches Büro, Germany) and using standard procedures (Stach et al., 1982; Taylor et al., 1998).

Owing to the small size of most samples, a subset of 17 were selected for biomarker analysis (Tables S1 and 1). The powdered samples were extracted for 72 h with chloroform using a Soxhlet apparatus. The extractable organic matter was then dried and weighed (Table 1). Aliquots of the extracts (12.4-90.1 mg) were precipitated with excess nhexane. After filtration through pre-extracted cotton wool, the maltenes were fractionated into aliphatic and aromatic fractions and polar compounds by column chromatography over activated silica gel and alumina (Cheng et al., 2016). The aliphatic hydrocarbons were analyzed by gas chromatography (GC) and gas chromatography-mass spectrometry (GC-MS). The GC analysis was performed using an HP 6890N GC equipped with a $30 \text{ m} \times 0.25 \text{ mm}$ i.d. (film thicknesses 0.25 µm) HP-5 ms capillary column. The starting temperature of the oven was 50 °C and increased to 100 °C at 20 °C/min, and then the temperature was increased from 100 °C to 310 °C at 3 °C/min with a final hold for 20 min. The GC-MS analyses were conducted using an HP 5973 mass selective detector coupled to an HP 6890 GC. The instrumental methodology for the GC-MS analysis has been previously documented in detail by Cheng et al. (2016).

4. Results

4.1. Bulk geochemical compositions

The TOC (%), hydrocarbon generative potential ($S_1 + S_2$, mg HC/g rock), hydrogen index (HI, mg HC/g TOC), temperature of the maximum pyrolysis yield (T_{max}, °C) and vitrinite reflectance (R_o, %) data for the Es₃, Ek₁ and Ek₂ units are detailed in Table S1, Figs. 2 and 3. The TOC contents of the Ek_2 samples range from 0.50 to 7.66%, with an average of 2.39%. The $S_1 + S_2$ and HI values of the Ek₂ samples range from 2.0 to 58.7 mg HC/g rock and from 68 to 882 mg HC/g TOC, respectively, while T_{max} values for these samples range from 440 to 450 °C. The Ek₁ member is characterized by low TOC, $S_1 + S_2$, and HI values that are < 0.46%, 0.56 mg HC/g rock, and 176 mg HC/g TOC, respectively. The T_{max} values for this member have a wide range (322–600 °C) due to the low S_2 value (0.01–0.38 mg HC/g rock). The overlying Es₃ samples show variable TOC values, ranging from 0.28 to 3.92%, but TOC values are generally > 1.0%. The $S_1 + S_2$ values of the Es₃ samples vary between 0.40 and 28.70 mg HC/g rock, indicating a strong potential for hydrocarbon generation. Hydrogen index values of the Es₃ samples are also highly variable and range from 105 to 719 mg HC/g TOC, while $T_{\rm max}$ values cluster between 434 and 440 °C.

4.2. n-Alkanes, acyclic isoprenoids and β -carotane

The representative gas chromatograms for the aliphatic fractions of the Ek₂ and Es₃ samples are shown in Fig. 4. The carbon number of the *n*-alkanes ranges from C_{15} to C_{37} , with maxima between *n*- C_{18} and *n*- C_{27} , and significant clustered around *n*- C_{21} -*n*- C_{23} . The *n*-alkanes display a unimodal distribution with a predominance of medium to high molecular weight compounds (*n*- C_{18} -*n*- C_{29}). A slight odd over even *n*-

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