

GEOCHEMICAL CHARACTERISTICS OF THE CRUDE OILS IN THE SOUTHEASTERN OFFSHORE MARGIN OF THE MEDITERRANEAN BASIN, NORTH SINAI, EGYPT

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Source rock characterization in the southeastern offshore margin of the Mediterranean basin was accomplished using five crude oil samples. Two samples were recovered from the Early Cretaceous Malha Formation reservoir in the Mango Basin. Three samples were recovered from the sand bodies within the Late Oligocene Tineh Formation in the Tineh Basin. Little is known about the geochemical characteristics of the organic matter in offshore North Sinai. These samples were tested for API gravity, sulphur content, and gas chromatography in order to infer the origin of organic matter, depositional environments, composition and thermal maturity of the source rock. The results showed that all the studied crude oil samples are characterized by low sulphur content ranging between 0.06 % and 0.24 %. Samples from the Mango-1 well (S4 & S5) shows high specific gravity (36 & 38 API), while S3 from the Tineh-1 well has moderate specific gravity (28.4 API). These results suggest the presence of two different types of oil families. Nickel and vanadium contents revealed that the studied crude oil types from the Mango-1 (S4 & S5) and S2 from Tineh-1 wells are grouped together and differ from S3 in the Tineh-1 well pinpointing to two different genetic oil families. Based on the carbon isotope composition, two main types of the studied crude oils were distinguished: waxy oils in the Tineh-1 well (S2) and the Mango-1 well (S4 and S5) and non-waxy oil in the Tineh-1 well (S3). The n-alkanes/isoprenoids ratios in S4 from the Mango-1 well revealed the highest contribution from the terrestrial organic matter during deposition, whereas the comparative ratios in S1, S2, and S5 showed a mixed source, and S3 showed the highest contribution from marine organic matter. This indicates that the source of the organic matter in the Mango-1 well was deposited under oxic to sub-oxic conditions, while in the Tineh-1 well; the organic matter was deposited under dysoxic to anoxic conditions. These results suggest the presence of two source rocks in the offshore North Sinai. All the samples (S1-S5) from the Mango-1 well and

the Tineh-1 well are derived from a mature source rock with variation in thermal maturity level from early mature in the Tineh-1 well (S1, S2, and S3) to the mature (oil-window) in the Mango-1 well (S4 and S5).

Key words: Source rock; Depositional environment; Thermal maturity; Gas chromatography; Offshore North Sinai; Egypt

1. INTRODUCTION

The North Sinai offshore represents the southeastern margin of the Mediterranean Sea (Fig. 1) covering an area of about 6631 km². Its location on the southern Tethyan margin of the Arabian-Nubian massif led to the accumulation of both terrestrial and shallow marine sequences (Guiraud and Bostworth, 1997), which are deeper than the North Sinai onshore. Hydrocarbon exploration in the North Sinai offshore began in 1979 by several companies under permission from the Egyptian General Petroleum Corporation (EGPC). Ten exploration wells were drilled in the North Sinai offshore area and most of them encountered good quality Lower Cretaceous turbidite sandstone reservoirs. Only two wells, the Mango-1 and Tineh-1 wells produced hydrocarbon with noncommercial amounts. The other eight wells were dry and some had limited gas shows (Hamdy et al., 2021). In comparison with the other petroleum provinces in Egypt, the geochemical studies that previously carried out on the North Sinai offshore are quite little (e.g., Al Sharhan and Salah, 1996; Nashaat, 2000; Dolson et al., 2001; Shabaan et al., 2006; and El Sheikh et al., 2016). The present study essentially aims to characterize some crude oil samples in the offshore North Sinai margin of the Mediterranean basin. This will be carried out using certain physical properties such as API gravity, sulfur content, nickel and vanadium and the gross composition of oil types, in addition to gas chromatography (GC) and the stable carbon isotopic composition to evaluate the genetic type of the organofacies and oil classes. Isoprenoids/normal alkanes (pr/n-C17 and ph/n-C18) give information on the type of organic matter and its thermal maturity, depositional environment, and redox conditions.

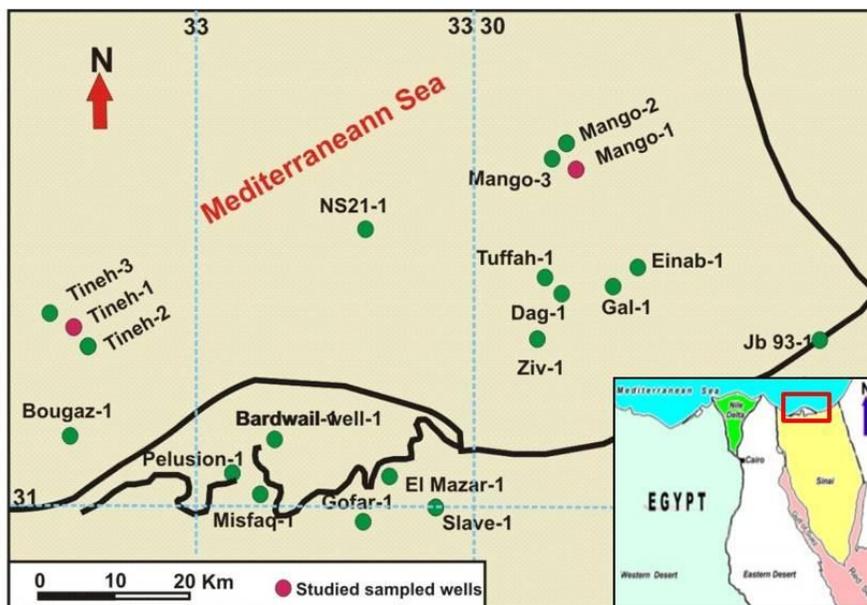


Fig. 1 Location map of the drilled wells in North Sinai offshore including the two studied Mango-1 and Tineh-1 wells (after Al Sharhan and Salah, 1996).

2. METHODOLOGY

Three crude oil samples from the Late Oligocene Tineh Formation in Tineh-1 well, and another two crude oil samples from the Early Cretaceous sandstones in the Mango-1 well in the offshore margin of the Mediterranean basin, North Sinai were investigated (Fig. 1).

1. API gravity is an indicator of crude oils' density at 15.6°C (60°F) and is calculated by $API = (141.5/Specific\ gravity) - 131.5$ (Waples, 1985).
 2. The sulphur content was detected by X-ray sulphur meter model RX-500 S using the ASTM D-42914 technique, and the results are reported as weight percent.
 3. Deasphalted crude oils were fractionated using open-column chromatography into saturates, aromatics and polar nitrogen, sulphur, and oxygen (NSO) compounds. A mixture of 1:2 wt % silica gel and alumina was used in the column packing of n-pentane introduced to the column.
 4. Saturates were recovered by n-pentane elution, aromatics with n-pentane and dichloromethane mixture 1:1. NSO compounds with methanol.
4. Gas chromatographic analysis was applied on saturated hydrocarbon

fractions using Perkin Elmer Instrument Model 8700, provided with a flame ionization detector (FID). The oven temperature was programmed for 100 to 320 °C at 3 °C/min and final time 20 min. SPB-1 capillary column of 60 m in length and 0.53 i.d. Nitrogen was used as a carrier gas, the optimum flow rate was 6 ml/min.

The analyses were performed in the Robertson Research International and Geoscience Technical Services, Oklahoma, USA, and were sincerely given by the British Petroleum (BP) company, Egypt.

3. RESULTS AND DISCUSSIONS

3.1 API gravity and sulphur content

API gravity and sulphur content of crude oils are classified into heavy with API gravity $< 27.3^{\circ}$, medium with API gravity ranging from 27.3° to 31.1° , and light with API gravity $> 31.1^{\circ}$ (Martinez et al., 1984). As a result, the Mango-1 oil samples (S4 and S5) have API gravities 36° and 38° , respectively (Table 1), reflecting light class, whereas the Tineh-1 oil (S3) has API gravity 28.4° pointing to a medium oil class. Paraffinic, paraffinic – naphthenic or naphthenic oils show a high waxy content with less than 1% sulfur, whereas the aromatic intermediate class shows a low waxy content with more than 1% sulphur (Tissot and Welte, 1984). The studied crude oils have a low sulphur content of less than 1% (Fig. 2, Table 1). Such data indicates that these oils are graded with a high maturity as paraffinic, paraffinic – naphthenic, or naphthenic groups (Makeen et al., 2015; Fig. 2).

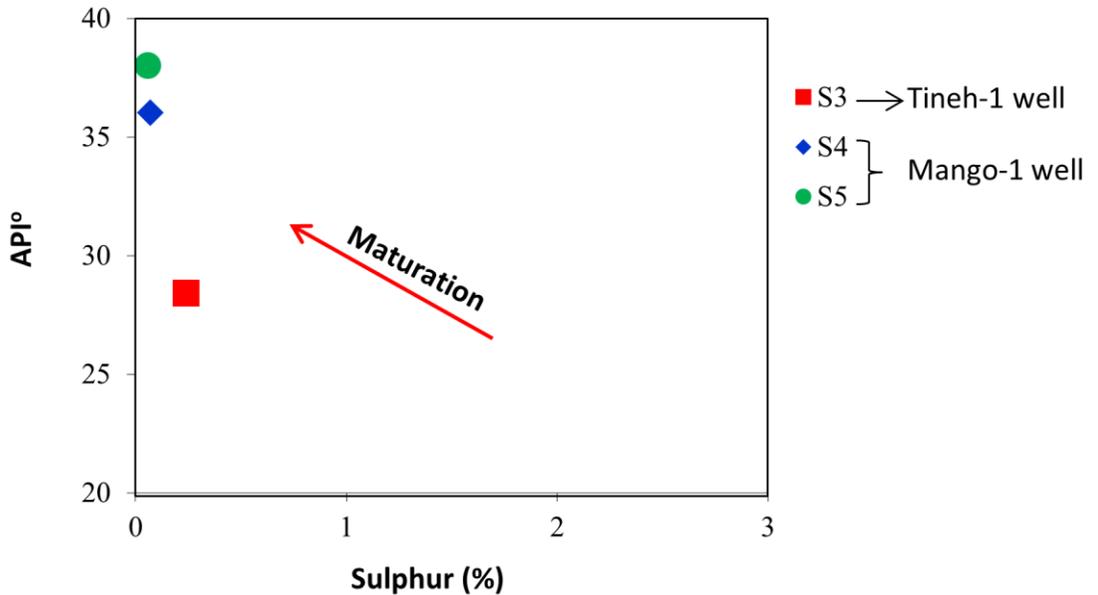


Fig. 2 Cross plotting API gravity and sulphur content in the Tineh-1 and Mango-1 oil samples, North Sinai offshore, Egypt (after Makeen et al., 2015).

3.2 Nickel and Vanadium contents

Nickel and vanadium elements remain constant during diagenesis, so their concentrations give a good implication on environmental conditions during the deposition of organic matter (Makeen et al., 2015). Barwise (1990) recognized high vanadium/nickel ratio (=1) as associated with oils from marine carbonate or siliciclastic deposits, low ratios (< 0.5) as involved in oils from lacustrine source rocks, whereas the extremely low ratio is found in higher plant organic matter. The investigated oil samples have a vanadium/nickel ratio approximately = 1 (Table1) indicating their derivation from siliciclastic or marine source rock. Cross plotting of V+Ni with sulphur content of samples (Fig. 3) has distinguished two groups of oils that emphasizing the dissimilarity between the Tineh-1 oils.

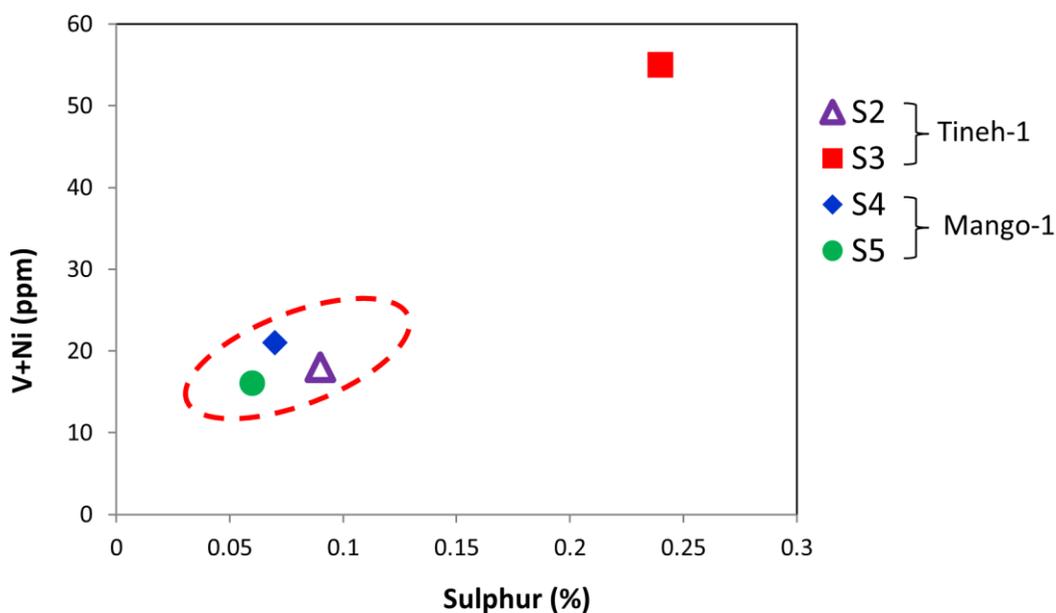


Fig. 3 Cross plotting of S Vs V+Ni of the Tineh-1 and Mango-1 oil samples, North Sinai offshore, Egypt (after El Sabagh et al., 2017).

3.3 Gross composition

The investigated crude oil samples showed that the saturate fraction is greater than the aromatic fraction (> 40%) as expressed by saturates/aromatic ratio (Table 1, Fig. 4). Accordingly, these oils are predominately paraffinic to paraffinic–naphthenic, which confirms the API gravity and sulphur content results.

3.4 Stable carbon isotope composition

An important application of stable carbon isotopes is to differentiate between algal and land plants organic matter sources, which distinguish marine from continental depositional environments (Younes et al., 2017). Accordingly, crude oils can be subdivided into marine (non-waxy) or terrigenous (waxy) based on the Canonical variable (CV). The terrestrial oils show CV values greater than 0.47, and marine oils show CV less than 0.47 (Sofer, 1984). In the present study, S2, S4, and S5 exhibits CV values more than 0.47 (5.89, 5.1 and 3.9, respectively) (Table 1), reflecting more contribution from terrigenous source rocks, while S3 shows CV value less than 0.47, which

refers to higher contribution from a marine organic source (Table 1). Such results show that the Tineh-1 oils are related to two different genetic families, whereas Mango-1 oil belonged to the same genetic oil family. That result was confirmed by Sofer's (1984) plot (Fig. 5). In addition, the nickel and vanadium content supports the same suggestion.

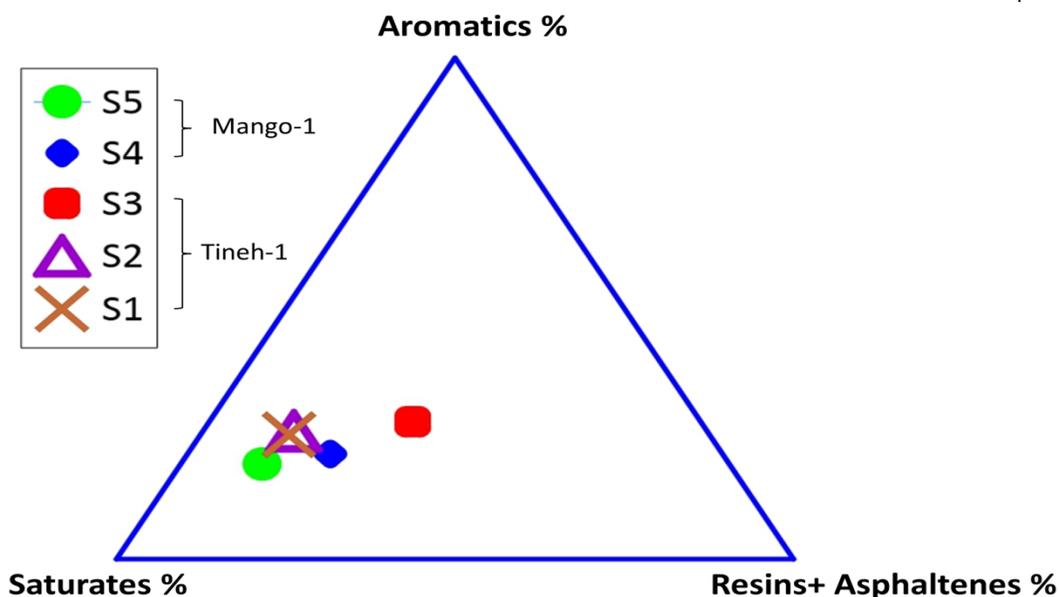


Fig. 4 Triangular diagram of Saturate-Aromatic-Resin and Asphaltene compounds (SARA diagram) showing the gross composition of oil samples, recovered from the Mango-1 and Tineh-1 wells, North Sinai offshore (after Peters et al., 2005).

3.5 Alkanes and acyclic isoprenoids

An integral part of identifying the source of organic matter is the configuration of n-alkanes and isoprenoids (Moldowan et al., 1985; El Nady and Mohamed, 2016; Fig. 6). The pristane/phytane (Pr/Ph) ratio can be used for predicting redox conditions during sediment accumulation but with some limitations because of the interference between thermal maturity and source inputs (Peters et al., 2005). High Pr / Ph (> 3.0) suggests oxic conditions, whereas low ratios (< 1) are consistent with anoxic, usually hypersaline, or carbonate environments (Peters et al., 1995, 2005). Cross-plot isoprenoid

pristane / n-C17 versus isoprenoid phytane / n- C18 is used to evaluate the depositional environments, type of organic matter, in addition to maturation and biodegradation (Wever, 2000). In the present study, the cross-plotting n-C17/n-C18 shows that samples S1, S2, from the Tineh-1 well and S5 from the Mango-1 well are originated from mixed organic matter (kerogen type II/III) with higher terrestrial input in S1. AMONG samples S3 showed highest contribution from marine organic matter, whereas sample S4 is originated in oxic environments from terrestrial organic matter (type III kerogen) (Fig. 7, Table 1).

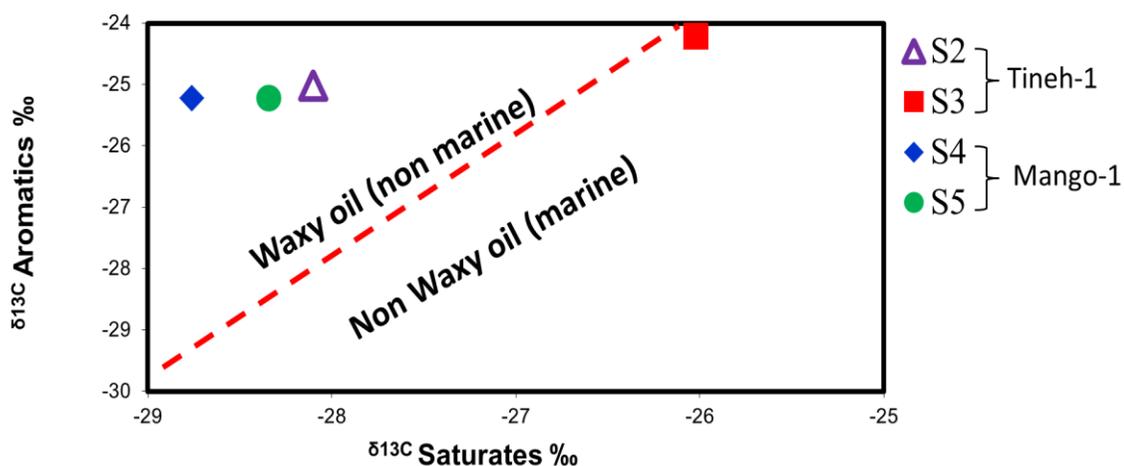


Fig. 5 Stable carbon isotope values of saturate versus aromatic hydrocarbons from the oil samples of the Mango-1 and Tineh-1 wells, North Sinai offshore. Dashed line divides waxy from non-waxy oils (after Sofer, 1984).

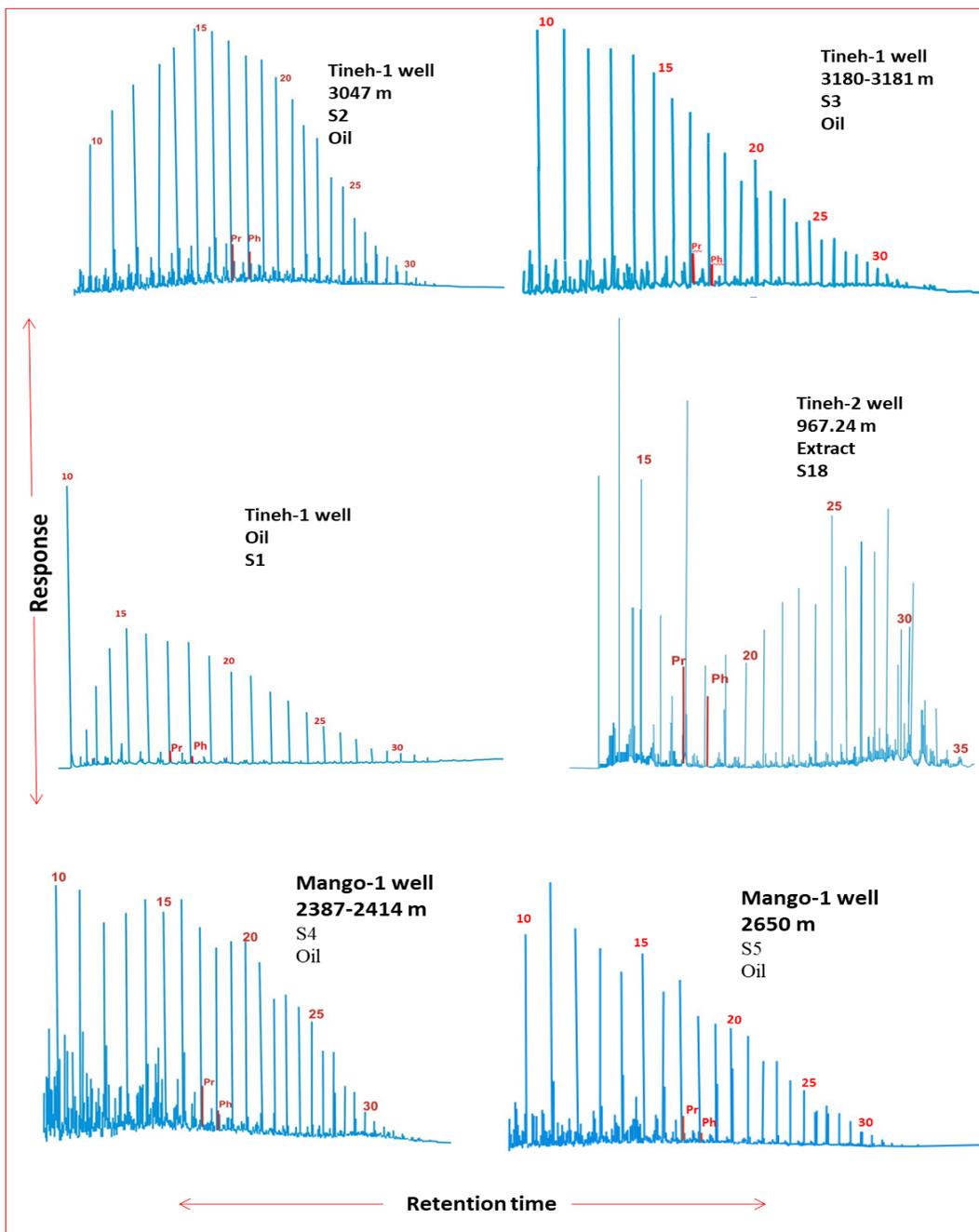


Fig. 6 Gas chromatograms of saturated hydrocarbon fractions of oil samples from the Tineh-1 and Mango-1 wells, North Sinai offshore, Egypt.

3.6 Waxiness index

Degree of waxiness is a standard method for categorizing the amount of land-

derived organic material (Hedberg, 1968; El Naggar and El Nady, 2016). This approach assumes terrigenous material contribution with normal paraffin of high molecular weight components to the oil (Köket et al., 1997). The degree of waxiness is calculated according to the formula $SC_{21}-C_{31}/SC_{15}-C_{20}$ ratios as expressed by (Scalan and Smith, 1970). The degree of waxiness in the studied oil samples ranges between 0.65 and 0.82 (Table 1) indicating a low waxy nature for the samples. Cross plotting of waxiness index with pristane/phytane revealed that the studied oils were originated from mixed organic matter under different conditions. The Mango-1 organic matter (S4 and S5) were deposited under more oxic conditions than that deposited in the Tineh-1 area (S1, S2, and S3) (Fig. 8).

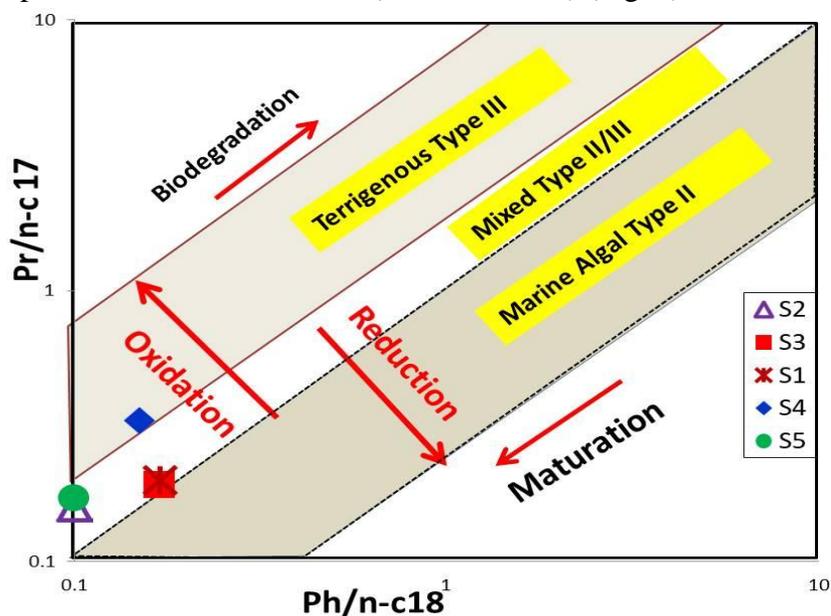


Fig. 7 Pristane/n-C₁₇ versus phytane/n-C₁₈ for the studied oil samples from the Mango-1 and Tineh-1 wells, North Sinai offshore (after Peters et al., 2005).

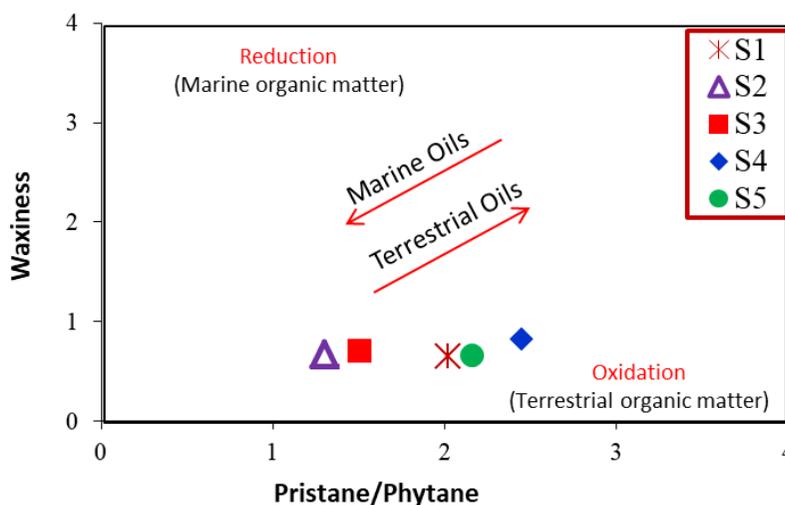


Fig. 8 Cross-plots of Waxiness index $\Sigma (n-C_{21}-C_{31}) / \Sigma (n-C_{15}-C_{20})$ versus pristane/phytane ratios the studied oil samples from the Mango-1 and Tineh-1 wells, North Sinai offshore (after Al Areeq and Meky, 2015).

3.7 Thermal maturity

The thermal maturity level of the investigated oil samples was detected using a range of oil characteristics. The relative proportions of saturated, aromatic hydrocarbons, resins and asphalt were used as markers of thermal maturity according to El Bassoussi et al. (2018). The studied oils show a high content of saturates and aromatic with respect to other constituents (Table 1) indicating mature source rocks. The studied crude oils have CPI values ranging between 1.03 and 1.22 (Bray and Evans, 1961; Table 1) close to unity indicating that the studied oils are related to a mature source rock, and oils expelled from source facies having wax-rich terrestrial components (Peters et al., 2005). In addition, odd-even predominance (OEP) can be calculated from the relation used by Scalan and Smith (1970). The OEP values for the studied oil samples vary from 1.04 to 1.22 (Table 1) reflecting mature source rocks. The Pristane/*n*-C₁₇ and phytane/*n*-C₁₈ cross plot (Fig. 7) indicates that the oil generated from the studied source rocks reached high thermal maturity levels. Equivalent vitrinite reflectance (Req%; Table 1) gives the same indication reflecting increased level of thermal maturity in the

source rocks, where S1, S2 and S3 samples from the Tineh-1 well show Req values 0.74, 0.57 and 0.67 respectively and the Mango-1 well has the values 1 % in S4 and 0.82 % in S5. These Req values suppose that the Tineh-1 oil types were generated from source rock at the early generation stage, while the Mango-1 oil types were derived from source rock that entered the main stage of hydrocarbon generation (Table 1).

4. CONCLUSIONS

The undertaken study of the geochemical characteristics of the crude oil in the southeastern margin of the Mediterranean basin, North Sinai, Egypt permits making three general conclusions:

1. The bulk composition of crude oil revealed that all the studied samples have low sulphur content, high API gravity and high paraffinic-naphthenic composition with two various types of oils from the Tineh-1 and Mango-1 wells. The Mango-1 samples 4 and 5 have the same genetic oil family, whereas Tineh-1 oils (samples 2 and 3) are dissimilar.
2. That variation in the depositional conditions indicates that oil samples S4 and S5 in the Mango-1 oils were derived from the same source rock, while the Tineh-1 oils are not related to one source rock (possibly two different organic matter types).
3. The studied offshore oil types were derived from source rocks reached the thermal maturity with varied levels from early mature in the Tineh-1 well to the oil-window in the Mango-1 well.

Well name	Tineh-1			Mango-1	
Sample no	S1	S2	S3	S4	S5
Depth (m)	ND	3047	3180.5	2403	2650
Sample Type.	Oil	Oil	Oil	Oil	Oil
Age	Oligocene			Cretaceous	
API (60°)	ND	28.4	ND	36	38
Sulfur (%)	ND	0.09	0.24	0.07	0.06
Vanadium (V)		9	28	12	8
Nichel (Ni)		9	27	9	8
Liquid chromatography					
Saturates (%)	69	57.9	42.7	61	62.1
Aromatics (%)	19	20.9	27.2	25.6	24.9
Asphaltenes	2	3	13.5	0	1.8
NSO	10	18.2	16.6	13.4	11.2
Saturates/ Aromatics	3.63	2.77	1.57	2.38	2.49
Gas chromatography					
Saturates	ND	-28.10	-26.02	-28.76	-28.86
Aromatics	ND	-25.02	-24.22	-25.23	-24.99
CV	ND	5.888	0.4122	5.1022	3.8986
Carbon isotopes $\delta^{13}C$					
Pristane/Phytane	1.5	2.02	1.3	2.45	2.16
Pristane/ n-C ₁₇	0.2	0.16	0.19	0.33	0.17
Phytane/ n-C ₁₈	0.1	0.1	0.17	0.15	0.1
C27/C17 index	ND	0.19	0.20	0.27	0.2
CPI	1.03	1.16	1.13	1.22	1.12
OEP	1.04	1.07	1.2	1.2	1.1
Waxiness Index	0.71	0.65	0.67	0.82	0.66
Equivalent Vitrinite reflectance (Req%)					
Req%	0.74	0.57	0.67	1	0.82

Table 1 Bulk composition, vitrinite reflectance measurements, carbon isotopes, and gas chromatography of the oil samples in the Tineh-1 and Mango-1 wells, North Sinai offshore area, Egypt.

ND: No data.

NSO: nitrogen, sulfur, and oxygen

$$CV = -2.53 \delta^{13}C_{Sat} + 2.22 \delta^{13}C_{Aro} - 11.65.$$

$$CPI = 1/2[(C_{25}+C_{27}+C_{29}+C_{31}+C_{33})/(C_{24}+C_{26}+C_{28}+C_{30}+C_{32})] + [(C_{25}+C_{27}+C_{29}+C_{31}+C_{33})/(C_{26}+C_{28}+C_{30}+C_{32}+C_{34})]$$

$$OEP = (C_{25}+6C_{27}+C_{29})/(4C_{26}+4C_{28}).$$

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الملخص العربي

اعتمد تقييم صخور المصدر في منطقة الجنوب الشرقي لساحل البحر المتوسط على دراسة خمس عينات للزيت، عينتان من الكريتاوي السفلي لمتكون الملح في حوض مانجو وثلاث عينات من الاوليغوسين العلوي لمتكون تينة في حوض تينة. وتعتبر الدراسات الجيوكيميائية التي اجريت على المادة العضوية في منطقة شمال سيناء البحرية قليلة. وقد اختبرت هذه العينات لقياس محتوى عنصر الكبريت والوزن النوعي وكذلك دراسة Gas Chromatography وذلك بغرض دراسة اصل المادة العضوية وبيئة الترسيب وكذلك تكوينها والنضج الحرارى لها. اوضحت النتائج ان كل عينات الزيت المستخدمة بها نسبة قليلة من الكبريت تتراوح ما بين 0.06 إلى 0.24. وتعتبر العينتان 4،5 من بئر مانجو-1 لهما وزن نوعي عالي 36،38 بينما العينة 3 من بئر تينة-1 لها وزن نوعي متوسط 28.4. وتوضح هذه النتائج وجود نوعين مختلفين من الزيت. واوضحت نسبة عنصرى النيكل والفانديوم ان العينتان 4،5 من بئر مانجو-1 وعينة-2 من بئر تينة-1 انهم يحتوا على نفس نوع الزيت بينما العينة رقم 3 من بئر تينة-1 بها نوع نوع زيت اخر. كما بينت دراسة النظائر المشعة للكربون وجود نوعين زيت مختلفين، نوع ثقيل من العينة رقم 2 من بئر تينة-1 و العينات 4،5 من بئر مانجو-1 ونوع خفيف من عينة 3 من بئر تينة-1. كشفت نسبة Alkanes/Isoprenoids عن وجود نسبة عالية من المادة العضوية القارية لصخر المصدر في العينة رقم 4 من بئر مانجو-1 بينما يحتوي صخر المصدر للعينات 4،5 من بئر مانجو-1 والعينة 4 من بئر تينة-1 على مادة عضوية مختلطة. وتوجد اعلى نسبة المادة العضوية البحرية في العينة رقم 3. وتشير هذه النتائج إلى ان المادة العضوية لصخر المصدر في بئر مانجو-1 قد ترسبت في بيئة مؤكسدة او تحت مؤكسدة بينما في بئر تينة-1 ترسبت المادة العضوية في بيئة غير مؤكسدة إلى تحت مؤكسدة مما يشير إلى وجود نوعين من صخور المصدر في منطقة شمال سيناء البحرية والتي وصلت إلى درجة جيدة من النضج الحرارى تزيد في عينات 4،5 من بئر مانجو-1 لتصل إلى نطاق الزيت.