Organic Geochemical Assessment of Jurassic Source Rock from Duhok, North Iraq

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Abstract: In the Northern Iraq, the Jurassic oil shales are widely distributed. The studied samples were collected from Sehkanyian, Sargelu and Naokelekan formations in Banik area, about 25 km due NE of Zakho town, Duhok Governorate, North Iraq. The hydrocarbon potentiality parameters, including type and amount of bitumen and kerogen, and thermal maturity of 72 samples as well as two oil samples were assessed by using Rock-Eval/TOC pyrolysis and GC/GCMS techniques. In general, Sargelu Formation samples have PI values below 0.1 and Tmax values varying between 437 and 449, suggesting immature to early mature stage of thermal maturity of oil. They can be considered as good to excellent source, where TOC content ranges from 0.06 to 28.57 with an average of 4.69%. The Naokelekan Formation samples can also be considered as an excellent source rock with TOC content ranges from 5.39% to 25.55%, averaging 17.77%, while the Sehkanyian Formation has no potential (TOC below 0.09%). The samples of Sargelu and Naokelekan formations contain both kerogen types I and II indicating marine organic matter mainly derived from algae and phytoplanktonic organisms proposing typical oil source kerogen. The extracted bitumen samples from Sargelu Formation show diagnostic ratios of Pr/Ph, Pr/nC17 and Ph/nC18 for generation of oil at an early thermal maturity stage. The low Pr/Ph ratio of the analyzed two oil samples from Jeribe and Sargelu formations together with the low Pr/n-C17 and Ph/n-C18 suggest that the oils was generated from a source rock containing mainly marine organic matter deposited under reducing conditions.

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Introduction

The Jurassic strata in northern Iraq contains very significant source rocks owing to the high total organic carbon (TOC) content of the Sargelu and Naokelekan formations that were deposited throughout the Jurassic basin that exists in these areas [1-3]. As far as the authors are aware, the published data on the stratigraphy and sedimentology of these source rocks are insufficient. The available information concerning the TOC content, thermal maturation, and burial history are seriously required for better understanding of the existence of other hydrocarbon resources [4]. The dominant organic matter in oil shales is derived from one or more primary sources, the terrestrial plants, lacustrine algae and marine organisms. These include large lake basins of tectonic origin, bogs, small lakes, lagoons and shallow seas [5].

To advance the existing database, Banik area in Duhok Governorate, Northern Iraq has been elected to perform the present assessment on the hydrocarbon potential. Banik section is situated in far north of Iraqi territory, about 25 km to the NE of Zakho town, (Duhok Governorate). The studied section lies on latitude 37°13′ 33.4" N and longitude 42° 58′ 2.6" E, nearly 100 meters to the west of the Banik village. The village is also known among resident farmers as Banik Haji Ghazi. Several mountains surround the area such as; Kokharash to the north, Khamtur to the west and Shaban to the south Fig. 1.

This paper reports the results of hydrocarbon source rock characterization of two formations of interest within the Banik area namely, Sargelu and Naokelekan formations and their hydrocarbon potential of the oil shale to be used economically.

Stratigraphy

The Jurassic rocks are commonly exposed as isolated patches at some eroded cores and limbs of anticlines in the highly folded, imbricated and thrust zones of Northern Iraq Fig. 2 a-b.

Banik area belongs to the highly folded zone of the unstable shelf of Northern Iraq [6]. The Banik section lies in a rugged terrain where the Jurassic rocks crop out in the southern limb of Kokharash anticline (mountain). The collected section is located about 100 meters to the west of Banik village near the famous and primitive open coal mine (dug in rocks of Naokelekan Formation). The general trend of strata follows that of Gara, which is E-W.

Banik section includes stratigraphic succession starting with Sarki and Sehkanyian formations (Early Jurassic) which underlies Sargelu Formation (Middle

Jurassic). The latter is, in turn, overlain by the younger Naokelekan and Barsarin formations (Late Jurassic), with Chia Gara Formation topping them Fig. 2a.

Sargelu Formation

The Sargelu Formation was first recognized and described by Wetzel in 1948 [7] at the Surdash anticline of the highly folded zone of Northeastern Iraq. The lithologic composition of the formation is rather uniform; it consists of thin bedded, black bituminous limestone, dolomitic limestone, and black papery shale with streaks of thin black chert in the upper parts [7].

The thickness of Sargelu Formation in Banik section is about 30 meters of which the limestone rocks are mostly dolomitized. In the subsurface sections on the foothill and

Mesopotamian zone of the unstable shelf, the thickness is considerably higher and varies between 250 and 500 m [8]. The contacts of the formation are both apparently conformable and gradational with underlying Sehkanyian and overlying Naokelekan formations. The Banik section can be subdivided in to three different lithofacies [9], which are, in ascending order:

Dolomitized Lithofacies (7 m thick).

Posidonia–bearing dolomitic limestone Lithofacies (5.5 m thick).

Cherty dolomitic limestone Lithofacies (17.5 m thick).

The depositional history offers indication that the environment of Sargelu had initiated and terminated as deep, quiet marine, interrupted by some relative shallow intervals but still remaining in the basinal realm. The tectonic subsidence during the Late Toarcian-Bathonian, when Sargelu was deposited, was fast and high over eastern area of Arabian Platform [10].

The organic–enriched black shale and limestone of Sargelu Formation indicate anaerobic to dysaerobic conditions [11]. Such conditions are suitable for preservation of organic matter that gets thermally mature to generate hydrocarbon in subsequent geologic times. In some other subsurface sections of Sargelu Formation, the potential source rock behavior is provided by being seated at great depths [12].

Naokelekan Formation

This formation was described by Buday [6] from top to bottom as follows:

1. Laminated shaly bituminous limestone alternated with bituminous shale and fine-grained limestone.
2. Fine grained hard mostly thin bedded, fossiliferous dolomitic limestone (motteled bed).
3. Thin bedded, highly bituminous dolomitie and limestone with intercalated black bituminous shale (coal horizon).

The age of Naokelekan Formation had determined as Upper Oxfordian–Lower Kimmeridgian according to the study of ammonites of the Upper Jurassic in that area [7]. The depositional environment of the formation was interpreted as euxinic and deposited in a slow subsiding basin [6]. The boundaries of the formation are agreed by many researchers to be conformable and gradational with underlying Sargelu Formation and overlying Barsarin Formation.

2. Methodology

About 72 samples were collected from the Banik area as well as two oil samples from Tawke oil field. The samples are collected along a traverse perpendicular to the bedding plane with spacing range of 0.5 to 1m. The collected samples represent the different lithologies of the Sargelu, Naokelekan and Sehkanyian formations. These lithologies are commonly of limestone, shale and chert. Analyses were conducted at Strato Chem Services Company in Cairo, Egypt. The representative 72 samples were prepared and submitted for Total Organic Carbon (TOC) determination, while forty one samples with TOC of 0.7% were analyzed by Rock Eval pyrolysis. The measured parameters include S1 (mg HC/g rock), S2 (mg HC/g rock), S3 (mg CO2/g rock), Tmax (°C), and TOC (wt. %) are quoted in Table 1. Several additional parameters such as Hydrogen Index (HI = S2/TOC*100), Oxygen Index (OI = S3/TOC*100) and Production Index (PI = S1/(S1+S2)) are calculated from these measured values and are shown in Tables 1-2. Gas chromatography (GC) and Gas chromatographymass spectrometry (GC-MS) were carried out to identify different hydrocarbon compound classes and biomarkers of the rock extracts and oil samples.

3. Results and Discussion

Pyrolysis techniques were used to establish the hydrocarbon source and the likely hydrocarbon products or source type.

Quantity of organic matter

The TOC content of the study sediments can be considered as straight expression of kerogen and bitumen abundance. The TOC (weight %) ranges in Sargelu Formation from 0.06% to 28.57 %, averaging 4.63% Table 1. The Rock-Eval Pyrolysis data are summarized in Table 3.

According to Peters [13], the sediments from the upper part of Sargelu Formation were rated from TOC contents as having excellent hydrocarbon potential (0.031 to 19.52%, averaging 4.12), the middle part rated as good hydrocarbon potential (0.06 to 18.12%, averaging 2.40), while the lower part can be rated as excellent hydrocarbon potential (0.12 to 28.57%, averaging 8.82). The TOC% values of the Naokelekan Formation range from 5.39 to 25.55% (17.77% in average) and can be considered as an excellent source rock. The Sehkanyian Formation has no source potential (0.03% to 0.09% wt).

Types of organic matter

It is necessary to recognize, that the quantitative aspects of kerogen evolution vary from one type to the other as a result of differences in the original composition of kerogen and that these generalizations may not reflect the true nature of the chemistry that occurs during the maturation process [14]. So, the types of organic matter must be distinguished and identified because different types of organic matter have different hydrocarbon generation potential and products [5, 15, 16]. The data of Rock-Eval Pyrolysis of potential source rock are presented in Tables 1-3. Most of the samples of Sargelu Formation plot in the field of kerogen types I and II Fig. 3 which suggest common lacustrine depositional environment. The organic matter is mostly derived from marine algae and phytoplanktonic organisms which can be considered as a typical "oil source" kerogen [17]. Only one sample lies in the field of kerogen Type III which is derived mainly from terrestrial organic matter. Naokelekan Formation belongs to kerogen type I and is considered as excellent oil prone.

The upper part of Sargelu Formation is characterized by localized intervals with capacity for both oil and mixed oil/gas generation. The oil-prone source rock intervals are characterized by very high TOC (5.37-19.52%) and excellent potential to generate oil as indicated by their high Hydrogen Indices (pyrolysis S2 yields from 32 to 127mg HC/g Rock; and HI mostly >500 mg HC /g TOC). The mixed-prone source rock intervals are characterized by low to very high TOC (0.70-8.13%) and fair to very good potential to generate oil and gas as indicated by their high Hydrogen Indices (pyrolysis S2 yields 2.60-19.03.16 mg HC/g Rock; and HI mostly >200 mg HC /g TOC).

The middle part of the Sargelu Formation is characterized by having capacity for oil, mixed oil/gas and gas generation. The oil generating rocks are characterized by very high TOC (4.66.37-18.12%) and very good to excellent potential to generate oil as indicated by their high Hydrogen Indices (pyrolysis S2 yields from 17 to 95mg HC/g rock; HI 365-525 mg HC/g TOC). The mixed oil/gas generating rocks are characterized by high TOC (1.71%) and fair potential to generate oil and gas as indicated by their high Hydrogen Indices (pyrolysis S2 yields 4.15 mg HC/g rock; HI 242 mg HC/g TOC). In addition the gas generating rocks are characterized by relatively high TOC (1.81%) and fair potential to generate oil and gas as indicated by their high Hydrogen Indices (pyrolysis S2 yields 2.95 mg HC/g rock; HI 163 mg HC /g TOC).

Lower part of Sargelu Formation is characterized by having capacity for oil, mixed oil/gas and gas generation. The oil-prone source rocks are characterized by their very high TOC (2.34-28.57%) and very good to excellent potential for oil generation as indicated by their high Hydrogen Indices (pyrolysis S2 yields 14.75-192.89 mg HC/g rock; HI >400 mg HC /g TOC). The mixed oil/gas prone source rocks are characterized by very high TOC (4.29%) and good potential to generate oil and gas as indicated by their high Hydrogen Indices (pyrolysis S2 yields 8.83 mg HC/g rock; HI 206 mg HC /g TOC). In addition the gas-prone source rocks are characterized by very high TOC (3.28%) and fair potential to generate oil and gas as indicated by their high HI (pyrolysis S2 yields 3.50 mg HC/g Rock; HI 107 mg HC /g TOC).

Naokelekan Formation is characterized by having the capacity for both oil and gas generation. The oil-prone source rocks are characterized by very high TOC (5.39-25.55%) and very good to excellent potential to generate oil as indicated by their high Hydrogen Indices (pyrolysis S2 yields 18.05-149.14 mg HC/g rock; HI >500 mg HC /g TOC) Fig. 4. The plotting of S1 versus TOC can differentiate between the migrated and non-migrated hydrocarbons [18]. The dividing line on the plot is where S1/TOC = 1.5. Values belonging to non-indigenous hydrocarbons plot above this line while indigenous hydrocarbon values emerge below it [18]. Thus all the samples analyzed of Sargelu and Naokelekan formations indicate indigenous hydrocarbons Fig. 5.

Thermal Maturity

The maturity levels for the oil window depend on the type of organic matter[19], and encompass a vitrinite reflectance (Ro) ranges from 0.5 to 1.3% and temperature at maximum rate of hydrocarbon generation during S2 evolution (Tmax) from 435 to 470°C. The PI parameter is another measure of maturity, with values ranging from 0.15 to 0.4 normally associated with oil generation. HI versus Tmax is commonly used to avoid influence of the OI for determining kerogen type [18]. Determining kerogen type using Tmax versus HI appears to be more accurate than OI versus HI. The difference between the results is expected because the Sargelu and Naokelekan formations are dominated by carbonates which affect the OI. The cross plot of HI versus Tmax indicates dominance of kerogen type II and mixed types II – III in both Sargelu and Naokelekan formations Fig. 6.

The level of thermal maturity can roughly be estimated from the HI versus Tmax plot Fig. 6. The figure shows that the majority of the samples are thermally immature to early stage of maturation, where pyrolysis Tmax ranges from 437 - 449°C and PI is than 0.15 [20].

Table 1: Total Organic Carbon and Rock Eval Pyrolysis data of the Sargelu Formation samples.

No	TOC wt. %	S1 mgHC/ g rock	S2 mgHC/ g rock	S3 mgHC/ g rock	Tmax (°C)	HI (mgHC/ g TOC)	OI (mgCO$_2$/ g TOC)	(S1/TOC) *100	PI S1/ (S1+S2)
1	8.13	0.24	19.03	2.68	442	234	33	3	0.01
2	6.41	1.05	44.69	1.43	437	697	22	16	0.02
3	5.37	0.33	32.34	1.38	442	564	24	6	0.01
4	1.22	0.03	2.6	0.63	439	213	52	2	0.01
5	16.55	0.32	106.83	2.62	444	645	16	2	0
6	2.41	0.15	6.36	0.08	442	264	3	6	0.02
7	14.6	0.34	84.76	2.09	444	581	14	2	0
8	11.07	0.25	62.51	0.88	443	565	8	2	0
9	1.87	0.08	5.23	0.84	440	280	45	4	0.02
10	2.29	0.11	6.4	1.33	442	279	58	5	0.02
11	0.7	0.06	1.51	1.16	441	215	165	9	0.04
12	19.52	0.24	127.16	2.45	443	652	13	1	0
13	0.86	0.03	1.62	0.86	444	189	100	3	0.02
14	16.53	0.24	91.63	4.58	444	554	28	1	0
16	18.96	0.32	109.65	4.89	440	578	26	2	0
19	0.96	0.03	1.03	0.46	446	107	48	3	0.03
21	0.71	0.03	0.8	0.18	444	113	25	4	0.04
25	0.96	0.03	1.19	0.2	444	124	21	3	0.02
27	0.99	0.06	0.95	0.4	449	96	41	6	0.06
34	1.81	0.43	2.95	1.2	445	163	66	24	0.13
35	18.12	0.37	95.07	3.96	444	525	22	2	0
36	5.88	0.19	29.54	0.48	444	502	8	3	0.01
37	1.71	0.34	4.15	0.37	439	242	22	20	0.08
43	0.76	0.04	0.78	0.2	447	103	26	5	0.05
46	4.66	0.08	17	1.32	440	365	28	2	0
49	2.62	0.04	1.49	2.41	443	57	92	2	0.03
50	3.28	0.07	3.5	2.68	442	107	82	2	0.02
51	4.29	0.13	8.83	1.87	443	206	44	3	0.01
54	20.4	2.45	132.66	1.54	444	650	8	12	0.02
55	19.86	2.39	131.18	1.81	444	660	9	12	0.02
56	13.15	0.67	90.78	2.26	445	690	17	5	0.01
57	3.53	0.35	14.75	0.82	441	418	23	10	0.02
58	7.43	0.21	39.05	0.45	443	526	6	3	0.01
59	28.57	0.67	192.85	3.78	446	675	13	2	0
60	2.34	0.18	11.04	0.67	441	472	29	8	0.02

Table 2: Total Organic Carbon and Rock Eval Pyrolysis data of the Naokelekan and Sehkaniyan formations samples.

Formtion	Sample No.	TOC wt. %	S1 mgHC/ g rock	S2 mgHC/ g rock	S3 mgHC/ g rock	Tmax (°C)	HI (mgHC/ g TOC)	OI (mgCO$_2$/ g TOC)	(S1/TOC) *100	S1+S2	PI S1/ (S1+S2)
Naokelekan	1	10.82	1.85	52.24	1.41	443	483	13	17	54.09	0.03
	2	25.07	3.15	132.37	3.70	445	528	15	13	135.52	0.02
	3	5.39	0.78	18.05	1.71	438	335	32	14	18.83	0.04
	4	21.28	4.42	135.25	2.46	444	635	12	21	139.67	0.03
	5	25.55	6.42	149.14	2.03	434	584	8	25	156.56	0.04
	6	18.49	3.39	106.58	1.87	442	576	10	18	109.97	0.03
Sehkaniyan	1	0.06									
	2	0.09									
	3	0.03									
	4	0.04									
	5	0.06									
	6	0.06									

Table 3: The ranges of TOC values for the studied samples are summarized in the following table.

Fm.	TOC Average (Min:Max)	S1 Average (Min:Max)	S2 Average (Min:Max)	S3 Average (Min:Max)	Tmax Average (Min:Max)	HI Average (Min:Max)	OI Average (Min:Max)	S1/ TOC Average (Min:Max)	PI Average (Min:Max)
U. SER	4.12 (0.06 : 19.52)	0.21 (0.03 : 1.05)	37.17 (0.80 : 127.16)	1.53 (0.08 : 4.89)	443 (437 : 449)	366 (96 : 697)	39 (3:165)	4 (1:16)	0.02 (0.00:0.06)
M. SER	2.40 (0.16:18.12)	0.24 (0.04:0.43)	24.92 (0.78:95.07)	1.26 (0.20:3.96)	443 (439:447)	317 (103:525)	29 (8:66)	9 (2:24)	0.04 (0.00:0.13)
L. SER	8.82 (0.12:28.57)	0.72 (0.04:2.45)	62.62 (1.49:192.89)	1.83 (0.45:3.78)	443 (441:446)	446 (57:690)	32 (6:92)	6 (2:12)	0.02 (0.00:0.03)
NAO	17.77 (5.39:25.55)	3.34 (0.78:6.42)	98.94 (18.05:149.14)	2.20 (1.41:3.70)	441 (434:445)	524 (335:635)	15 (8:32)	18 (13:25)	0.03 (0.02:0.04)
SEK	0.06 (0.03:0.09)								

Fig. 1: Location map of the studies Banik area, Northern Iraq.

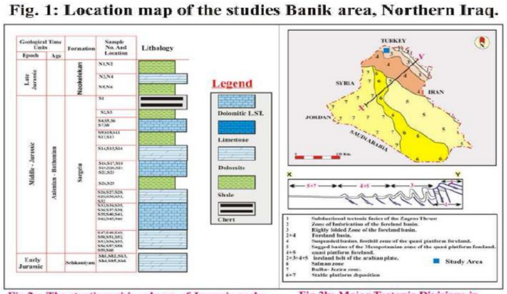

Fig.2a: The stratigraphic column of Jurassic rocks crop out in the Banic section, Northern Iraq indicating the collected samples.

Fig.2b: Major Tectonic Divisions in Iraq (after Numan, 2000)

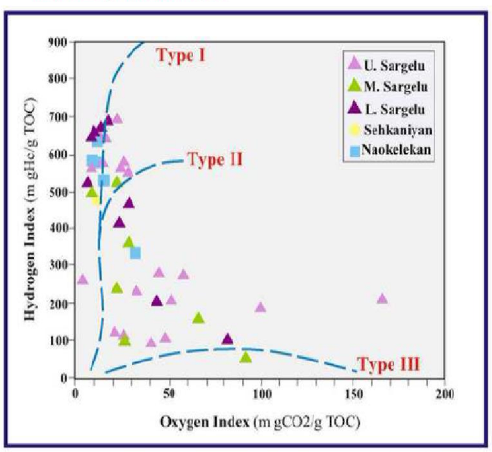

Fig. 3: Van Krevelen diagram, showing Rock-Eval Hydrogen Index vs. Oxygen Index for the analyzed samples of the study area (adapted from Espitalie et al.,1977).

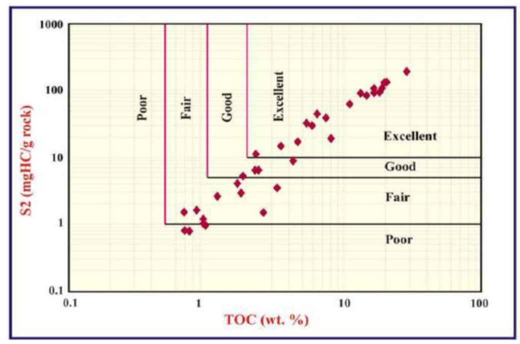

Fig. 4: The hydrocarbon potentiality of Sargelu and Neokelekan formations by using their hydrocarbon potentiality using TOC wt% versus S2.

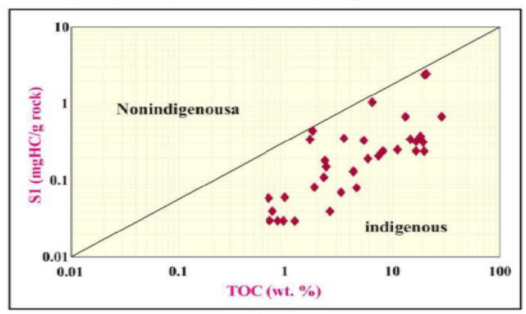

Fig. 5: S1 versus TOC for Sargelu and Naokelekan formations in Banik section (modified after [17] Hunt, 1996).

The Rock Extracts and Oil

Gas-chromatography is performed for the extract of 6 rock samples in addition to 2 oil samples. The gas chromatograms of the extracted samples from the Sargelu Formation show unimodal distribution pattern of the normal alkane with maximum intensity of the light normal hydrocarbon less than n-C20 peaks indicating autochthones source of the organic matter Fig. 7. The low content of hydrocarbon and the hump of the heavy hydrocarbon indicate that the samples are thermally immature. This is also certified by the low ratio of isoprenoid to normal alkane and the low PI parameter (< 0.10). The ratios of the Pristane/Phytane (Pr/Ph) in the 6 extracts of the present work are low (0.70 to 1.38) Table 4, indicating anoxic reduced marine carbonate depositional environment. The organic-rich anoxic carbonate rocks generally generate oils with Pr/Ph ratios less than 2 [18, 21-24].

CPI is the ratio of odd carbon number to even carbon number n-alkanes in extractable organics. CPI may be used as an indicator of maturation, where immature rocks often had CPI values more than 1.2 or less than 0.8. Some marine sponges, fresh water aquatic plants, ferns, fungi, yeasts and bacteria have small odd carbon preferences [25]. The average of CPI of the extract samples of Sargelu Formation is less than 0.9 indicating marine source at early stage of maturation.

The six extracted samples from Sargelu Formation have Pr/nC17 (0.23-0.39) and Ph/nC18 (0.20-0.44) reflecting deposition under reducing marine condition rather than effect of maturity or biodegradation Fig. 8. The whole oil gas chromatograms of two oil samples recovered from North Iraq at 353- 416 m of Jeribe Formation and 2840 m of Sargelu Formation reveal n-alkane distribution pattern in the n-C4 to n-C41 range Fig. 7 with API values of 23.18º and 29.85º; respectively.

The both oils samples of Jeribe and Sargelu formations have low Pr/Ph ratio (0.87 and 0.73, respectively), which together with the Pr/n-C17 (0. 37 and 0.26, respectively) and Ph/n-C18 (0.52 and 0.44, respectively) suggest that they are generated from a source rock containing mainly marine organic matter deposited under reducing depositional conditions Fig. 8.

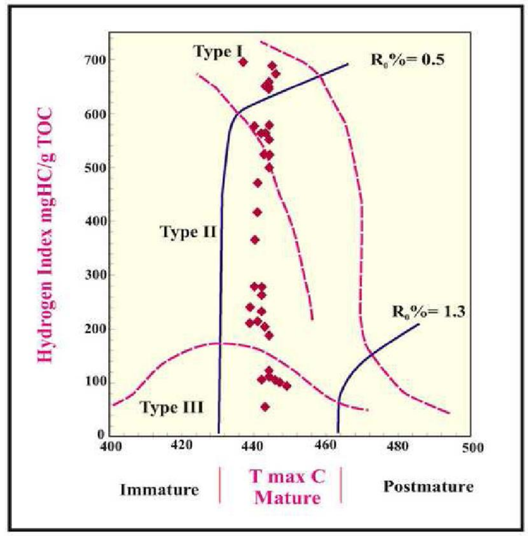

Fig. 6: Kerogon plots for HI vs. Tmax of the samples of Sargelu and Naokelekan formations of the Banik section

Table 4: Gas Chromatograph data of the 6 extract samples of Sargelu Formation as well as the 2 oil samples

S. No.	Pri/Phy	Pri/nC_{17}	Phy/nC_{18}	CPI Marzi[4]	N Paraffins	Isoprenoids	Resolved unknowns
1	1.14	0.26	0.37	0.89	11.9	1.5	86.6
12	1.33	0.39	0.39	0.91	12.7	2.0	85.3
18	1.15	0.28	0.25	0.97	23.6	2.2	74.2
21	0.91	0.24	0.29	0.87	25.3	2.9	71.7
26	1.38	0.23	0.20	0.78	20.9	3.1	76.0
52	0.70	0.28	0.44	0.95	23.5	3.6	72.9
Jeribe Fm	0.87	0.37	0.52	0.99	26.1	4.5	60.9
Sargelu Fm	0.73	0.26	0.44	0.98	30.8	3.8	56.5

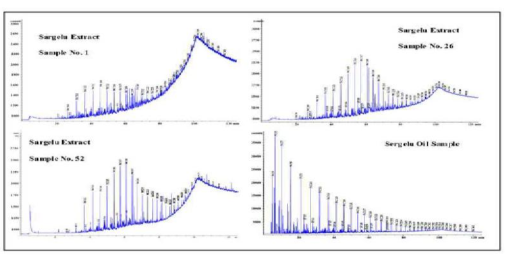

Fig. 7: Gas Chromatograms of the rock extract of the Sargelu Formation And its oil sample from North Iraq.

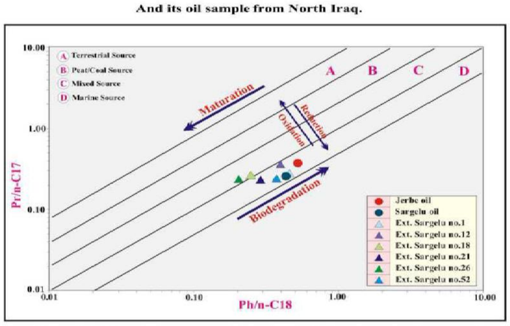

Fig. 8: Pr/n-C17 versus Ph/n-C18 of the Sargelu Formation and the two oil samples.

Conclusions

In this study the rock units of Jurassic age of the Duhok area have been assessed for their hydrocarbon source potential. A total of 72 samples from Sehkanyian, Sargelu and Naokelekan formations were analyzed by Rock-Eval /TOC pyrolysis as well as 6 rock extract and two oil samples were analyzed by GC/GCMS techniques.

In general, The Sargelu and Naokelekan formations have kerogen types I and II in immature to early mature stage of thermal maturity of oil. They can be considered as good to excellent source, where the average of TOC content is more than 4.69%, while the Sehkanyian Formation has no potential (TOC below 0.09%).

The extracted bitumen samples from Sargelu Formation show diagnostic ratios of Pr/Ph, Pr/nC17 and Ph/nC18 for generation of oil at an early thermal maturity stage. The low Pr/Ph ratio of the analyzed two oil samples from Jeribe and Sargelu formations together with the low Pr/n-C17 and Ph/n-C18 suggest that the oils was generated from a source rock containing mainly marine organic matter deposited under reducing conditions.

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