Geochemical Characteristics of Six Formations Based on Organic Geochemical Parameters, Murzuq Basin, Libya

ABSTRACT

Cutting samples (n = 150) and Core samples (n = 6) from the Taouratine, Dembaba, Assedjefar Marar, Awaynat Wanin, Tanezzuft and Mamuniyat Formations (Jurassic to Ordovician), derived from wells (A-3, B-1, H-1 and H-15), locate in the A, B and H fields, present in Murzuq Basin were analysed. Rock-Eval Pyrolysis, Total Organic Carbon and specific Aromatic Molecular Biomarker (by the use of chromatography-mass spectrometry GC-MS) as geochemical parameters implemented to investigate their Lithology, kerogen type, organic matter (OM) richness and maturity evaluation. Such Formations are fair to very good quantity of organic matter passing in the course of excellent source rocks, have average of organic carbon richness (TOC) value ranged between 0.2% to 16.7% with one anomalously rich sample at 666m (well H-1) where a dark grey shale has a TOC content of 46.1% and high potential yield over 90000 ppm. The studied rocks are ranged from immature to mature (oil-prone organic matter) of hydrogen index (HI) ranged between 24 - 302 mg HC/g TOC, related with oxygen index (OI): 3 - 161 mg CO2/gTOC indicate that organic matter is dominated by Type II/III kerogen. T_{max} , spore colouration (SCI) and Vitrinite Reflectance (% R_o) as maturity parameters ranged among 425 - 445, 5 - 8.5 and 0.35 - 3 respectively. Aromatic hydrocarbon ratios by use of gas chromatography - mass spectrometry pointed to two levels of thermal maturity, where the high level of thermal maturity recorded in lower Silurian , whereas the less maturity was from other formations.

Keywords: Murzuq Basin; Libya; Tanezzuft Formation; Aromatic biomarkers; Potential yields; Kerogen type; Maturity source rocks, kerogen type, Rock-Eval pyrolysis, maturation, aromatic biomarkers

1. INTRODUCTION

The Murzuq Basin, located in southwestern Libya, is one of Libyan productive petroleum basins, extend southward into Niger. The Libyan regain covers an area of approximately 58000 km^2 (Figure 1). Exploration operations started in this region already in 1957 and exploratory wells have been drilled, resulting in over tens discoveries with around 4,000 million barrels of oil even now. Most discoveries have been completed in Ordovician sandstone reservoirs where the Lower Silurian Tanezzuft Formation sourced [1]. In the basin center, the very slight amounts of early organic matter were probably expelled from Devonian rich shale [2]. The hot shale (Tanezzuft formation) described as the only effective hydrocarbon source of significance in the Basin [3], even as [1] reported single proven mature source rock in the Murzuq Basin with at least two other potential oil source rocks. Petroleum system in the basin is sourced by Rhuddanian hot shall were deposited on Ordovician sandstone surface with gravities API ranges 36 – 45^0, prior to main Hot Shale Member (Tanezzuft Formation) with thickness of 350 to 475 m in Silurian marine transgression [1]. The API by gamma ray from C1 in concession 174 recorded 800 units with Tanezzuft thickness more than 15m [1]. TOC values in the hot shale (Lower Silurian) in concession 174 reach more than 16% in contrast with normal shale who have TOC values ranging from 0.5 to 1.3%. The richness TOC from concession 58 showed 3.45% reported by [2], as well an average 1.8% TOC in center basin published by [4]. there many Organic geochemistry have been published, concerned with the petroleum geochemistry of oil-Libyan basins including studies on the kerogen type, source rock potential, oil-oil and oil-source rock correlation [5, 6, 7, 8, 9]. The kerogen type and thermal maturity of organic matter can be categorized according to hydrogen index (HI) and Tmax values [10, 11]. The purpose of this paper is to describe lithology of studied formations, TOC analysis, rock pyrolysis, organic matter reflectivity, spore colouration and aromatic molecular biomarker as geochemical parameters to establish kerogen type, potential yield and thermal maturity.

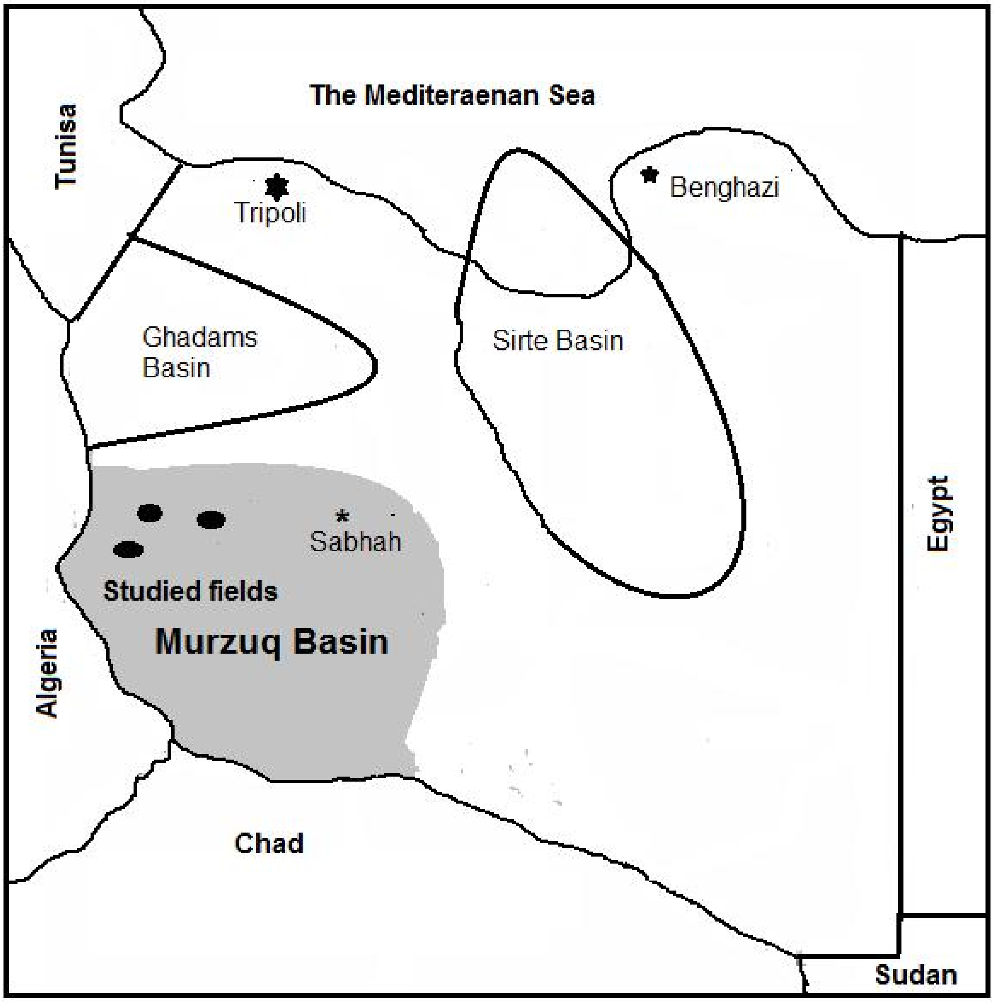

Figure 1. Map shows the location of the Murzuq Basin and studied fields. Modified from Hallett D. 2002 and Aboglila et al. 2010.

1.1. Geological Setting

The basin is to the southeast of the Ghadames Basin, which is separated by the Atshan Saddle, an anticlinal structure trending approximately east north east to west south west which leads into the western end of the Al Qarqaf High. Three major defining tectonic elements are surrounding the Libyan part of Murzuq Basin, the Al Qarqaf High to the north, and the Tibisti Uplift to the east and the Hoggar Uplift to the west. The contained sediments are ranging in age from Precambrian basement to Quaternary [4]. Thicknesses of formations show a broad similarity over the three fields (A, B and H) from which selected group of samples that were supplied and analysed. The deposition environment of Quaternary is between 20 and 55m thick. Across the studied area, the absence of the Miocene, Eocene and Oligocene formations with overlied Mesozoic sediments (Fig 2).

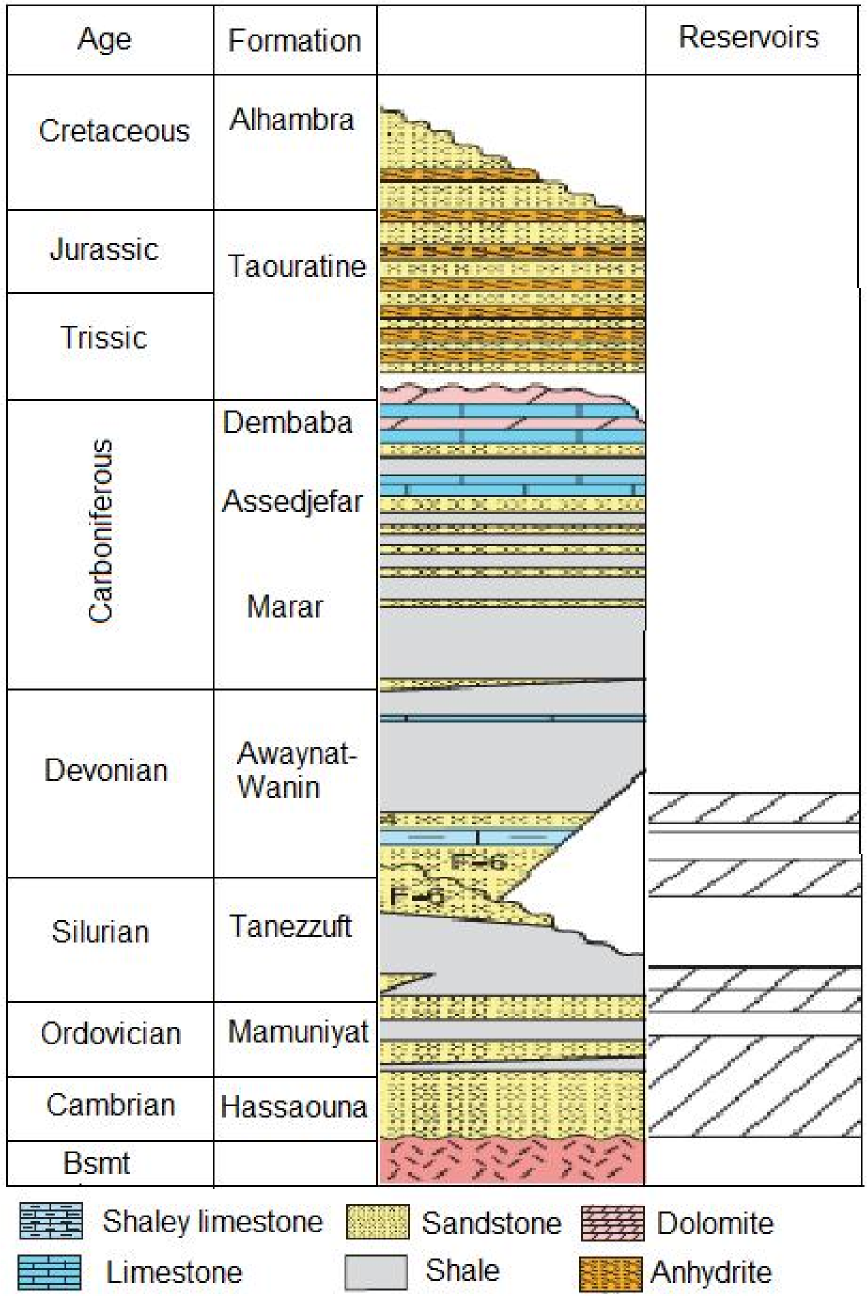

Figure 2. Stratigraphic column of the Murzuq Basin highlighting the lithology of the formations and the reservoir units. Modified from Rusk, D. C., 2001.

The sediments of Upper Cretaceous and Lower Cretaceous are destroyed and truncated by an escarpment to the southeast of the basin. The Jurassic Taouratine Formation nearly 200m thickness is preserved in the three fields from which samples were taken. Depth of Jurassic deposition is greatest to the south and southeast of the basin and diminishing towards northwest. Under the Jurassic, the Triassic Formation is between 250 and 300m thick, with absent of Permian sediments leaving the Triassic deposition resting on the Upper Carboniferous in the fields sampled. The Greater thicknesses of Triassic sediments also deposited to the northwest of studied zone [1]. The Carboniferous sediments in the studied area were of four formations, the Upper Carboniferous consists Dembaba and Assedjefar Formations, while the Lower Carboniferous consists Marar and Lower Marar Formations. Several differences in the thicknesses of individual formations across the zone, even though the general thickness of the Carboniferous preserved is roughly similar, ranging from 501m to 548m .The Carboniferous sediments overlies the Upper Devonian Awaynat Wanin Formation with range in thickness from 115m to 148m, thickening from South West to North East. Below this, the Tanezzuft Formation of Llandovery age (Silurian) was a preserved thickness of between 200 and 250m. It contains the Hot Shale Member which is present over approximately half of the search area in thicknesses of up to 30m. The Tanezzuft Formation is thinner in the eastern part of the zoon and is eventually truncated in the subsurface to the east of the aimed section. The Ordovician Mamuniyat Formation is present across the basin with depth up to 240m in well B1 [2].

2. MATERIAL AND METHODS

2.1. Samples

The study is geochemical analysis of a wide variety of samples from wells A-3, B-1, H-1 and H-15, locate in the A, B and H fields, present in Murzuq Basin to provide a better understanding of a number of aspects of the source rock geochemistry. One hundred and fifty six samples of drill cuttings and core were collected by the National Oil Corporation (NOC) in Tripoli from 4 different wells, locate in the A, B and H fields (Fig. 1). The units sampled were the Taouratine, Dembaba, Assedjefar Marar, Awaynat Wanin, Tanezzuft and Mamuniyat Formations consists of a diversity of interbedded lithofacies, shales and conglomerates (Fig. 2).

2.2. Samples Lithology

The physical characteristics of rock unit such as colour and texture were described to define sediment types and show their differences.

2.3. Preparation of Samples

The rock samples were washed twice by distilled water and dehydrated at room temperature prior to analysis, Trailing by Rock-Eval pyrolysis and TOC measurement. The representative samples were chosen for additional comprehensive organic geochemical assessment. These samples were ground to a fine powder (particle size) using a ring-mill.

2.4. Rock-Eval Pyrolysis and Total Organic Carbon (TOC) Measurements

Rock-Eval pyrolysis of powdered rock (10 - 50 mg) was carried out on a Girdel Rock-Eval instrument, while the TOC was measured on a Leco instrument. Samples with low TOC values (<0.3%) were considered unsuitable for Rock-Eval pyrolysis such as the samples from Taouratine Formation and therefore not subjected to further analyses.

2.5. Solvent Extraction and Isolation of the Maltenes

Extraction data for group of selected source rock samples is presented and good source rock is considered to have a total extract of over 1000 part per million as petroleum potential yield. The samples from representative formations were selected for solvent extraction based on TOC values (>0.3%). About 10 - 20 g of ground sediment were extracted into an ultrasonic bath for two hours using a 9:1 mixture of dichloromethane (DCM) and methanol (MeOH). The solvent extract was then filtered and excess solvent removed by careful heating on a sand bath (60°C) to obtain the bitumen. In brief, the sample extraction (maltenes, about 10 - 20 mg) was applied to the top of a small column (5.5 cm × 0.5 cm i.e.) of activated silica gel (120°C, 8 h). The aliphatic hydrocarbon (saturated) fraction was eluted with n-pentane (2mL); the aromatic hydrocarbon fraction with a mixture of n-pentane and DCM (2mL, 7:3 v/v,); and the polar (NSO) fraction with a mixture of DCM and MeOH (2mL, 1:1 v/v).

2.6. Gas Chromatography-Mass Spectrometry (GC-MS)

Aromatic fractions were analysed by GC- MS using a Hewlett Packard (HP) 5890 mass-selective detector (MSD) interfaced to a AMS 92 gas chromatograph (GC). The GC oven was programmed from 30°C to 310°C at 3°C/min, after which it was held isothermal for 30 min. Samples were dissolved in n-hexane and introduced by the HP 5890 sampler into a split-split less injector operated in the pulsed-split less mode and biomarker data was acquired in a full-scan mode m/z 50 to m/z 500. Selected aromatic compounds were identified using m/z 178 (phenanthrene), m/z 156 (dimethylnaphthalenes) and m/z 184 (dibenzothiophene) ions.

3. Results and Discussion

A variety of organic geochemical analyses were applied to reveal petrogenic characteristics of rock hydrocarbons from three fields located in the Murzuq Basin. On the basis of the purpose of this paper, the results of lithology description, TOC analysis, rock pyrolysis, organic matter reflectivity, spore colouration and aromatic molecular biomarker have carried out to discuss potential yield, kerogen type and thermal maturity for sediments collected from three discovered fields in the Murzuq Basin.

3.1. lithology of studied formations

The lithology of the studied formations show diverse physical characteristics such as colour and texture (Table 1). The samples from Taouratine and Dembaba Formations demonstrate texture of the majority mudstone mixed with low sand ratio with varied color. Medium dark to grey shale with trace Mudstone samples from Assedjefar Fm and continues this lithology with low to minor sandstone in samples of Marar Fm. Mostly multicolor shale with minor mudstone and sandstone described in samples of Awaynat Wanin Fm, except samples obtained from Well B1 where appeared Multicolor mudstone and sandstone with low dark-grey shale. The lithology of the Tanezzuft Fm recorded a great dominance of Dark-grey shale with low to minor mudstone and sandstone while low Tanezzuft Fm (Hot shale, Table 1) illustrated full Dark-grey shale. Below this, the sample from Ordovician Mamuniyat Formation showed multicolor sandstone and mudstone with low dark-grey shale (Table 1).

Table 1. The lithology of the studied formations show diverse physical characteristics.

Formation	Well A1	Well B1	Well H1	Well H15
Taouratine	Mudstone multicolor with sand	Red claystone with multicolor mudstone	Mudstone multicolor mostly dark	non
Dembaba	Multicolor mudstone mostly dark to dusky	Very pale mudstone with multicolor mudstone	Mostly multicolor mudstone with minor sandstone	non
Assedjefar	Dark-grey shale with red-brown mudstone	Medium dark-grey shale with trace mudstone	none	non
Marar	Dark-grey shale with multicolor mudstone	Mostly multicolor shale with low mudstone and sandston	Dark-grey shale with low to minor mudstone and sandstone	non
Awaynat Wanin	Dark-grey shale with low mudstone	Multicolor mudstone and sandstone with low dark-grey shale	Mostly multicolor shale with minor mudstone and sandstone	non
Tanezzuft	Dark-grey shale with low to minor mudstone	Medium dark-grey shale with trace mudstone	Dark-grey shale with very low mudstone	dark-grey shale with trace mudstone

Hot shale	non	non	Dark-grey shale	Dark shale
Mamuniyat	Multicolor sandstone	Multicolor sandstone with low dark-grey shale	Dark-grey shale with low mudstone	non

3.2. organic matter richness

Organic matter richness in source rocks can be investigated by several parameters. TOC (%) and potential yields measurements for the studied samples show quite diverse values (Table 2). The Lower Silurian Hot Shale Member at the base of the Tanezzuft Formation is a high overall average of TOC content with a maximum recorded value of 16.7% with the exception of one anomalously rich sample from Lower Carboniferous (Marar Formation, well H1) where dark grey shale has a TOC content of 46.1%. Potential hydrocarbon yields determined by Rock-Eval pyrolysis demonstrated fairly varied values between formations (Table 2). Well A3, the Potential yields, however, are low throughout the formations reaching a maximum of 1750ppm and increased in samples from Well B1 reaching a maximum of 3110ppm (Tanezzuft Fm). Potential hydrocarbon yields are fair to good in H1 formations, yielding a maximum of 4220ppm with the exception of the dark grey shale sample (TOC = 46.11%) from Marar Formation, recorded high Potential hydrocarbon yields reaching 90850 ppm. The Potential yield of Hot Shale member (low Tanezzuft Fm) from Well H15 is fair to excellent, ranging from 630 to 37419 ppm (Table 2). The organic matter richness of sediments is estimated typically using the TOC wt% and potential yield (ppm). The data of TOC (wt %) and potential yield (ppm) measurements for the studied formations showed quite diverse values. The studied source rock quality is generally poor due to low TOC and potential yield values. Even though, the results of the representative samples as description study are showing that the major source rock in the sequence is the Hot Shale Member (Lower Silurian, Llandovery) which consistent with published search about petroleum provenance. From the other side, their samples have fair potential with exception of one anomalously rich sample from Marar Fm (well H1) which has a TOC content of 46.1% and potential yield 90850ppm (Table 2). This could be pushed to think around a possible contribution from another source rock in the remaining formations.

Table 2. Results of bulk geochemical data for selected formations from the Basin

Well	Sa	Fm	TOC%	Pot. (ppm)	HI	OI	Tmax	SCI	Ro%
		1	0.20 - .21	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
		2	0.63 - 0.65	n.d.	n.d.	n.d.	n.d.	7	0.55
A3	46	3	057 - 1.97	210- 1740	24-88	18 - 68	427- 436	7	0.63 - 1.70
		4	0.59 - 0.92	260- 570	33-62	28 - 40	431- 435	7	0.53 - 1.73
		5	0.66 - 1.11	790- 1750	108-179	11 - 40	431- 437	8	1.14 -1.75
		6	0.16 - 0.24	n.d.	n.d.	n.d.	n.d.	6	n.d.
		1	0.21 - 0.35	n.d.	n.d.	n.d.	n.d.	5	n.d.
		2	0.53 - 1.40	220- 230	26-30	40- 43	427- 428	6	0.57 - 105
B1	54	3	0.25 - 4.20	640- 2550	26-86	19- 46	425 -431	7	0.35 -1.35
		4	0.60 - 1.10	760- 1510	75-176	30- 42	427- 434	6	0.54 - 1.55
		5	0.50 - 2.30	550-3110	30-133	30-161	427- 439	7	0.59 - 1.00
		6	0.70 - 1.0	3020	302	22	433 -434	5	1.28 - 1.90
		1	0.17	n.d.	n.d.	n.d.	n.d.	7	n.d.
		2	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
H1	33	3	0.37- 46.12	1041-90850	63-197	16- 26	427- 430	6	0.44 - 1.52
		4	0.65- 1.50	1040 - 1330	107-121	5- 7	428- 430	7	0.56 -1.07
		5	0.50- 0.95	1360-1960	166-210	50-55	436- 438	7	0.40 - 3.
		6	0.40- 2.35	1190- 4220	138-214	9- 28	431- 435	7	1.37 - 2.0
H15	23	5	0.40- 0.83	1030- 1880	166-237	19-68	430- 437	7	0.57 - 0.85
		7	0.80- 16.70	630- 37419	90-260	3-44	433- 445	8.5	0.68 - .71

Sa: Sample number; Fm: Formation; Fm 1= Dembaba; Fm 2: Assedjefar; Fm 3: Marar; Fm 4:Awaynat Wanin; Fm 5:Tanezzuft; Fm 6:Mamuniyat, 7:Hot Shale, TOC (wt%): total organic carbon; Pot. (ppm): potential yields; HI: Hydrogen Index; OI: Oxygen Index; Tmax (C°): maximum pyrolysis temperature yield; SCI: spore colour index; %Ro: Vitrinite Reflectance; n.d.: not determined.

3.3. kerogen type

Kerogen type data measured based on parameters of OI and HI (Table 2) and Plot of hydrogen index (HI) versus oxygen index (OI) showed in Fig 3, point to the organic matter categorized as kerogen type II and kerogen type III, with absence of kerogen type I in studied Samples. Sediments characterized by HI >300 mg HC per gram TOC, can generate oil [12, 13]. The organic matter within studied formations is identified as predominant mixed types II - III kerogens (oil prone) as indicated by HI values (Fig. 3). Kerogen Type II is dominated and characteristically derived from a mixture of amorphous organic matter desmocollinite (vitrinite) and exinite (cutinite or suberinite) with minor lamalginite [14].

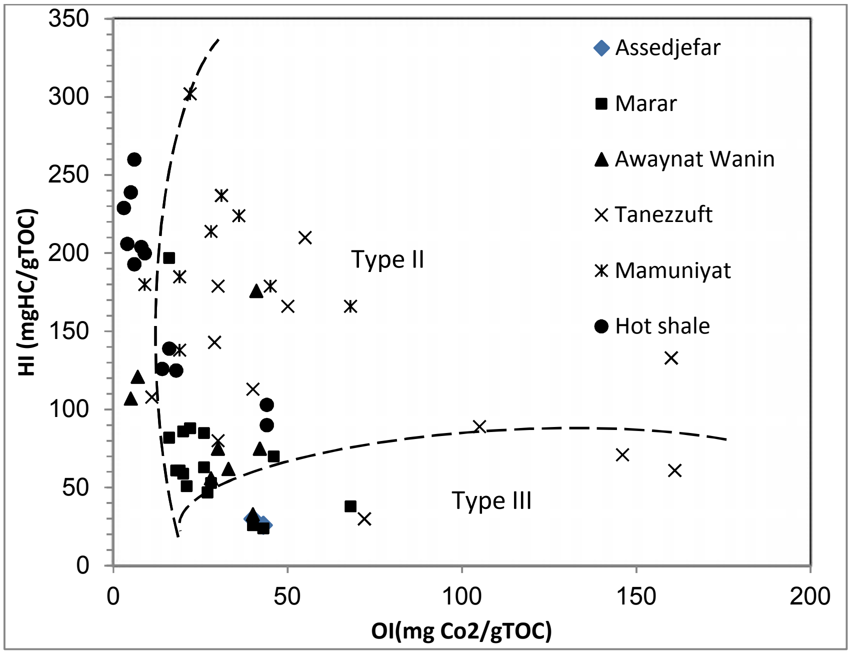

Figure 3. Plot of hydrogen index (HI) versus oxygen index (OI) illustrating the variation of kerogen type (II and III) in source rocks of the studied fields.

3.4. Thermal maturity

3.4.1. T_{max} , spore colouration (SCI) and Vitrinite Reflectance (% R_o)

T_{max}, spore colour index and Vitrinite reflectance are the most widely used thermal maturity parameters of kerogen [14], were used to estimate maturity of the organic matter. Data of representative samples from selected formations show quite diverse values. Hot shale member (lower Tanezzuft, the maximum value of Tmax (445), spore colour index range from 5 – 8 and difference values of Vitrinite reflectance was estimated (Table 2). The maturity determined by Tmax, illustrated that most of the samples lie inside the mature region of hydrocarbon generation >430 [15]. The thermal maturity of organic matter classified according to hydrogen index (HI) and Tmax values [10]. The data of HI and T_{max} values confirmed that the analysed rock samples contain immature, marginal mature and mature organic matter with a significant amount of oil-prone possible to generate, particularly from older sediments and closer to part of rifting such Tanezzuft, Hot Shale and Mamuniyat, (Silurian and Ordovician) as it is extremely obvious in Fig. 4.

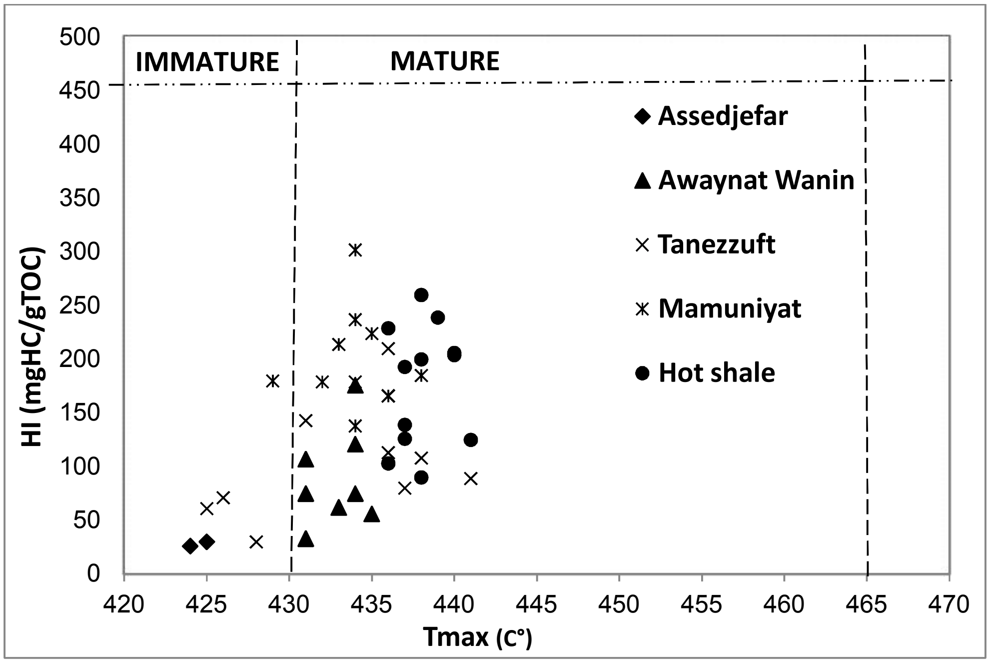

Figure. 4. A plot of HI vs Tmax from Rock-Eval pyrolysis showing position of various samples from three fields, Murzuq Basin.

4.2.1. Aromatic molecular biomarker

Results of aromatic fraction gas chromatography - mass spectrometry used as geochemical parameters of thermal maturity, determined from the distribution and abundance of aromatic biomarkers (Table 3 and Fig 5). In representative samples, aromatic molecular biomarker such naphthalene , methylnaphthalene, dimethylnaphthalene, trimethylnaphthalene, phenanthrene, methylphenanthrene and methyldibenzothiophene compounds are presented in Table 3 and used to discuss the level of thermal maturity of studied rocks. Results of aromatic fraction via gas chromatography - mass spectrometry and plot (Fig. 5) of distribution and abundance of aromatic hydrocarbon ratios revealed two levels of thermal maturity . The aromatic hydrocarbon extracted from Hot shale member are significantly different (characterized as high aromatic content and indicated high level of thermal maturity) furthermore could not be correlated with any of the aromatic hydrocarbon extracted from other formations which low aromatic content and levels of thermal maturity clear in table 3. Maturation level with respect to oil generation is an interpretation based upon all available primarily spore colour and Vitrinite reflectance. Samples from the Silurian and Devonian showed spore colouration indicate a diversity between the maturity of the formations as the data mentioned table 3.

Table. 3. Aromatic hydrocarbon ratios based on peak areas gas chromatography - mass spectrometry

Aromatic compounds		A1	H15	H1	B1
1: 2MN/1MN	m/z 142	*	0.85	1.75	1.54
2: 26,27DMN/15DMN	m/z 156	*	2.4	7.79	3.53
3: 236TMN/146,135TMN	m/z 170	1.2	1.13	1.19	1.04
4: 125TMN/136TMN	m/z 170	0.43	0.19	0.06	0.1
5: 3MBP/2MBP	m/z 168	*	*	*	*
6: MPI-1: 1.5*(3MP+2MP)/(P+9MP+1MP)	m/z 192,178	0.5	0.75	0.87	0.87
7: MPI-2: 3*2MP/(P+9MP+1MP)	m/z 192,178	0.59	0.82	0.94	0.96

8: (3MP+2MP)/(3MP+2MP+9MP+1MP)	m/z 192	0.38	0.44	0.47	0.46
9: 2MP/(3MP+2MP+9MP+1MP)	m/z 192	0.23	0.24	0.26	0.25
10:(TA20+TA21)/(TA20+TA21+TA26+TA27+TA28)	m/z 231	*	*	*	0.83
11: TA21/(TA21+TA28R)	m/z 231	*	*	*	0.92
12: TA26S/TA28S	m/z 231	*	*	*	3.22
13: TA27R/TA28R	m/z 231	*	*	*	0.52
14: 4MDBT/1MDBT	m/z 198	1.82	8.11	9.4	6.34
15: 4MDBT/DBT	m/z 198,184	0.96	3.91	0.6	0.56
16: DBT/P	m/z 184,178	0.1	0.08	0.55	0.56

MN=Methylnaphthalene, DMN= Dimethylnaphthalene, TMN=Trimethylnaphthalene, MBP= methylbiphenyl, MP= methylphenanthrenes, TA= triaromatic steroids, MDBT= methyldibenzothiophene, DBT= dibenzothiophene, P= phenanthrene

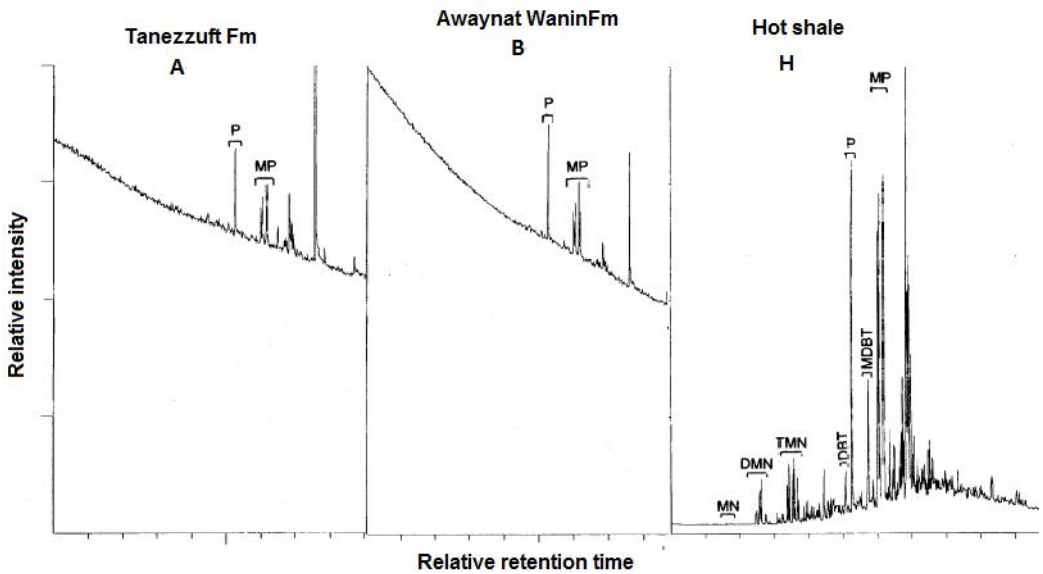

Figure. 5. Aromatic fraction gas chromatography - mass spectrometry of three selected formation from three fields, peak identification in table 3.

5. Conclusion

The main conclusions of this study are:

1- Most rock Kerogen is type II, good possibility to generate and expel liquid hydrocarbons in the mature case more than Kerogen Type III.

2- Although, T_{max} values indicating fair source rock potential and potential yields exceed 2000 ppm point to mature organic matter in the Carboniferous and Devonian Formations.

3- A regional study of the three fields has been carried out that the Hot Shale Member (Lower Silurian) at the base of the Tanezzuft Formation is a high quality, oil level source rock which is currently at oil maturity and has not yet gone in the gas window.

4- Overall average TOC content of the Hot Shale Member is high with a maximum recorded value of 16.7%.

5- Vitrinite Reflectance (% R_o) and Spore colouration (SCI) of the Hot Shale Member ranges from 0.57% to 0.85% and 7.0-8.5 respectively, indicted higher than others.

6- Aromatic hydrocarbon ratios revealed two levels of thermal maturity, the Hot shale member showed high aromatic content, revealed stage of thermal maturity while rest sediments have low aromatic hydrocarbon reflected marginal thermal maturity.

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