

OIL-OIL CORRELATION OF ASMARI AND BANGESTAN RESERVOIRS USING GAS CHROMATOGRAPHY AND STABLE ISOTOPES IN MARUN OILFIELD, SW IRAN*

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Abstract – For this study, Asmari (Oligo-Miocene) and Bangestan (Cretaceous) reservoir oils from the Marun oilfield were studied geochemically. Gas chromatograms and stable isotopes of carbon and sulfur in different oil fractions were studied. Normal alkanes nC_{15}^+ are as high as 93% with a saturate percentage up to 53.9% which reveal a high maturity of Asmari and Bangestan reservoir paraffinic oils. Carbon Preference Index of both reservoir oils are around one, indicating mature oil samples. Pr/nC_{17} and Ph/nC_{18} ratios have confirmed this conclusion. The Pr/Ph ratio is less than one and the plot of $\delta^{13}C_{Aro.}(\text{‰})$ versus $\delta^{13}C_{Sat.}(\text{‰})$ both indicate a marine reducing environment during the deposition of their source rocks. The organic matter deposited in these sediments is of kerogen Type II (Algal). Stable carbon isotope results versus Pr/Ph ratio indicate that both oils originate from the same shaley limestone of the Mesozoic age. This study also proves that H_2S gas polluted Asmari oils have a similar isotopic range as Bangestan reservoir oil, hence the source of contamination must have originated from the Bangestan reservoir. Isotopic and geochemical results, for the first time, introduce three oil families; two H_2S polluted families and one non- H_2S polluted oil family; in the entire Marun oilfield.

Keywords – Marun, Asmari, Bangestan, Oil-Oil Correlation, GC, Carbon and Sulfur stable isotopes

1. INTRODUCTION

Southwest Iran constitutes one of the most prolific hydrocarbon producing habitats [1] with cumulative recoverable reserves of 136 billion (10^9) barrels of oil, including recent discoveries in the Kushk and Hosseinieh fields of Khuzestan Province, including 940 trillion (10^{12}) cubic feet of associated and non-associated gas [2]. Although the geological framework of this area is well defined [3-6], considerable uncertainty exists as to the origin of these hydrocarbons.

The history of geochemical investigation in Iran goes back to the years 1932-1941, when Lees [7] fractionated some crude oil as well as bitumen in Fars. The first comprehensive geochemical project in Iran was conducted by Bordenave and his coworkers [8-11]. It was conducted to determine the characters of source rocks and extent of their distribution. They also correlated the oils with probable source rocks and with tectonics of Fars, Khuzestan and Lorestan regions.

Bordenave and Burwood [1] carried out the most important and comprehensive research work on the distribution and maturation of oils in the Zagros basin using stable carbon and sulfur isotopes, as well as on biomarkers. They studied biomarkers and stable isotopes on oils from Asmari and Bangestan reservoirs, as well as the probable source rocks in the Dezful Embayment. Due to the extended area of

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study, only two oil samples from the Asmari and Bangestan reservoirs of the Marun oilfield were studied. Their results reveal that, despite the similarity of their source rock, different isotopical characters are shown.

The probable source rocks (Pabdeh, Gurpi, Kazhdumi and Garau Formations) in the Marun oilfield have been pyrolyzed by Rock-Eval6 and reported by Alizadeh and coworkers [12]. The Kazhdumi Formation (Albian), with its high TOC, HI and PI, is reported to be the best source rock of the Marun oilfield [12].

In the Marun oilfield H₂S gas pollution in the Asmari reservoir is rapidly spreading. Hence, in this study oil samples from polluted and non-polluted oil from Asmari, as well as oil from the Bangestan Reservoir have been correlated to determine the source of H₂S gas in the Asmari reservoir. For this reason the normal alkane distribution and stable isotopes of carbon and sulfur have been studied.

Stable carbon ($\delta^{13}\text{C}$) and sulfur ($\delta^{34}\text{S}$) isotopes are a very powerful tool for oil-oil correlation [13]. For high sulfur oils, a stable sulfur isotope study is suggested [14]. Carbon isotope ratio of whole oils is generally not used alone for correlation because of the narrow range of $\delta^{13}\text{C}$ values for all oils, which are about 15‰, ranging from -18 to -33‰ [15]. Consequently, for oil-oil correlation in this study oil fractions were used. In geochemical investigations multiple tools are recommended; thus, to achieve better and more reliable results, normal alkane distribution, determined by gas chromatography, were used to support stable isotope results.

2. GEOLOGICAL SETTING

The Zagros Fold-Thrust Belt lies on the northeastern margin of the Arabian plate. Like other fold-thrust belts, it shows intensive shortening close to the suture which becomes less intense toward the foreland. It is dominated by NW-SE trending folds and thrusts. The mountain belt has been divided into NW-SE trending structural zones (imbricated and simply folded Belt) parallel to the plate margin, separated by major fault zones such as the High Zagros and mountain Front Faults (Fig. 1). The imbricated belt situated between the high Zagros and Zagros main Reverse Faults and Simply Folded Belt lies to the southwest of the High Zagros Fault [6].

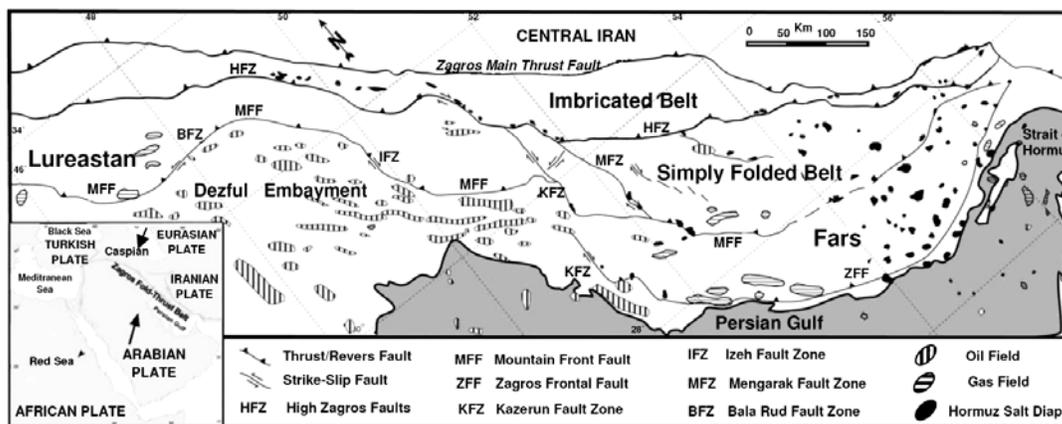


Fig. 1. Marun Oilfield in the Zagros Fold-Thrust-Belt with the structural setting of the region (modified from Sepehr and Cosgrove [6])

Marun oilfield is situated at the end of a simply folded zone, very close to the border of the folded zone (southwest Iran) and unfolded zone (Arabian plate). During Upper Cretaceous to early Tertiary periods, stratigraphy and structural geology in the Marun area were severely influenced by tectonic activities. In the Upper Cretaceous Bangestan Group sediments have been deposited in a shallow sea in

the Zagros basin and on the Arabian plate. In the southwest of Iran, Bangestan Group sediments consist of organic matter, rich limestone, and a thick pile of shales. It is believed that Bangestan Group sediments are source rocks for all oils in Arabian reservoirs in the region [16].

Tectonic activities during the deposition of the Bangestan Group caused various structural features in the Arabian Plate. During Tertiary, these activities caused severe effects on distribution as well as the formation of Gurpi, Pabdeh and Asmari Formations. The beginning of the Tertiary Era coincides with sea transgression, and hence the deposition of the Pabdeh Formation on the marls of the Gurpi Formation. The contact between Pabdeh and Gurpi indicates the Cretaceous-Tertiary boundary. The second phase of tectonic activities at the end of Cretaceous caused the deposition of ophiolites along with the north, as well as the eastern Arabian Plate. A succession of erosion and deposition of these ophiolites in the deep sea created clastic sediments in southwest Iran [16].

The Asmari reservoir (Oligo-Miocene) in the studied oilfield comprises sequences of limestone, sandstone and shale, while in the Bangestan reservoir (Upper Cretaceous) the lithologies consist of only limestone and shale (Fig. 2). Until now 305 oil wells have been drilled, but only 4 wells were completed in the Khami reservoir, 17 wells in the Bangestan reservoir and the rest were completed in the Asmari reservoir [17].

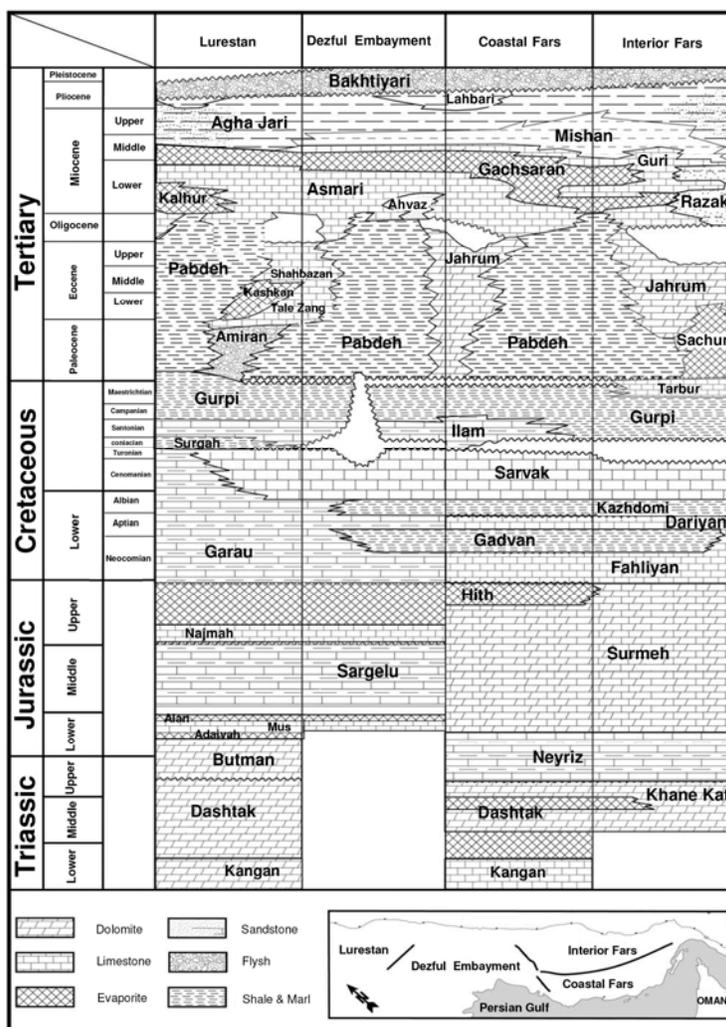


Fig. 2. Mesozoic-Cenozoic stratigraphic correlation chart of the Iranian Sector of the Zagros basin showing the lateral lithology and facies changes along and across the folded Belt [6]

3. METHODS

For this study 10 oil samples from producing wells of the Marun Oilfield were selected and analyzed using Gas Chromatography as well as stable isotopes of carbon and sulfur.

3.1. Sample Selection

National Iranian South Oil Company (NISOC) collected the oil samples for the present study from the selected wells all over the oilfield. Geological information, as well as chemical composition of the samples are given in Table 1. For better correlation of Asmari and Bangestan oils, two samples were taken from the Bangestan reservoir, three from non-contaminated and five from H₂S contaminated Asmari oils (Table 1).

Table 1. Geochemical data and gross chemical composition of oils

Well No.	Position (in oilfield)	FM.	Depth (m)	% C ₁₅ ⁺	% Sat.	% Aro.	% NSO	% Asph.
# 62	Center	Ban.	3428-3450	93	40.1	25.8	11.8	7.6
#237	SE	Ban.	3316.5-3382	88	53.9	20.7	12.9	2
#35	SW	Asm. (n- cont.)		90	38.1	29.5	16.4	1.3
#173	SW	Asm. (n- cont.)		89	45.0	26.8	13.6	1.4
#200	N	Asm. (n- cont.)		89	35.7	22.9	13.5	0.2
#208	S	Asm. (Cont.)	3407-3480	90	36.0	27.9	14.7	1.3
#233	S	Asm. (Cont.)	2964-3225	91	35.6	26.3	14.4	2.6
#162	N	Asm. (Cont.)	3501-3722	91	38.4	27.9	13.1	2.5
#69	S	Asm. (Cont.)		92	38.3	27.8	14.7	2.1
#41	S	Asm. (Cont.)	2882-2972	93	46.5	29.3	11.8	1.9

FM- Formation; Ban. – Bangestan Reservoir; Asm. (n-cont.)- Asmari Reservoir (non-contaminated with H₂S gas); Asm. (Cont.)- Asmari Reservoir (contaminated with H₂S gas).

3.2. Gas Chromatography

To perform Gas Chromatography (GC), asphaltene from the sample was removed with n-hexane. The asphaltene free sample was injected into Gas Chromatography model 2010 of Vinci Technologies in the petroleum laboratory of Shahid Chamran University. Malten (Asphaltene free oil) was injected in a heated port connected to a fused-silica crosslinked-methylsilicone gum capillary column (25 m×0.22 mm i.d.×0.25 µm film thickness). The oven temperature was ramped from 50 to 320°C. The biomarker peaks were identified by Flame Ionization Detector (FID).

3.3. Isotope Ratio Mass Spectrometry (IRMS)

3.3.1. Sample preparation

Total oil extracts were separated into their saturate and aromatic fractions by preparative silica gel chromatography. Hexane and toluene were used as elute solutions for saturate and aromatic separation, respectively.

3.3.2. Carbon-13 analysis

Stable carbon isotope ratios were determined on whole oil and on oil fractions using EA-IRMS (Elemental Analyzer Isotope Ratio Mass Spectrometry) of Iso-Analytical Laboratories (Sandbach, Cheshire, UK). Carbon dioxide peaks separated by a packed column gas chromatograph, held at an isothermal temperature of 110°C, were entered as the ion source of Europa Scientific GEO 20-20 IRMS to be ionized and accelerated. Gas species of different masses were separated in a magnetic field and simultaneously determined using a Faraday cup collector array to measure the isotopomers of CO₂ at m/z 44, 45, and 46.

The reference material used for the present analyses was IA-R002 oil standard with a δ¹³C value of -28.06‰ vs. V-PDB of Iso-Analytical Lab.

Carbon isotopic compositions (in per mil, ‰) are reported relative to the Pee Dee Belemnite standard (PDB) using the following equation [18]:

$$\delta^{13}\text{C} \text{ ‰} = [(R_{\text{sample}}/R_{\text{standard}}) - 1] \times 10^3$$

where R is the ratio of ¹³C to ¹²C.

3.3.3. Sulfur-34 analysis

Sulfur-34 analysis was undertaken by EA-IRMS. Sulfur dioxide is resolved on a packed GC- column at a temperature of 30°C. The resultant SO₂ peak enters the ion source of the IRMS where upon it is ionized and accelerated. Gas species of different masses are separated in a magnetic field then simultaneously measured on a Faraday cup universal collector array. Analysis was based on monitoring of m/z 48, 49 and 50 of SO⁺ produced from SO₂ in the ion source.

The reference materials for all samples were measured against IAEA-S-1, Silver sulfide, δ³⁴S_{V-CDT} = -0.3 ‰, NBS-127 (Barium sulphate, δ³⁴S_{V-CDT} = +20.3 ‰) and Iso-Analytical IA-R025 (Barium sulphate, δ³⁴S_{V-CDT} = +8.53 ‰). Use of three reference materials allows for calibration and correction of the oxygen-18 contribution to m/z 50. NBS-127 and IAEA-S-1 are distributed as isotope reference standards by the International Atomic Energy Agency (IAEA), Vienna. IA-R025 is calibrated against and traceable to NBS-127.

Sulfur isotopic compositions (in per mil, ‰) are reported relative to the standard Troilite sulfur (FeS) from the Canon Diablo meteorite using the following equation [19]:

$$\delta^{34}\text{S} \text{ ‰} = [(R_{\text{sample}}/R_{\text{standard}}) - 1] \times 10^3$$

where R is the ratio of ³⁴S to ³²S.

4. RESULTS AND DISCUSSION

4.1. Gas Chromatography

In the studied samples n-alkanes and nC₁₅⁺ are high (Table 1), revealing a high maturity [20] of the Asmari and Bangestan reservoirs of the Marun oilfield. For a compositional classification of the studied reservoirs, a ternary diagram of Tissot and Welte [21] was used (Fig. 3). On the basis of whole oil minor cuts percentages, the studied oils show paraffinic composition with relatively higher maturity in both Asmari and Bangestan reservoir oils. Here, well No. 237 shows maximum maturity and is well expected since it's of the deep reservoir Bangestan oil sample.

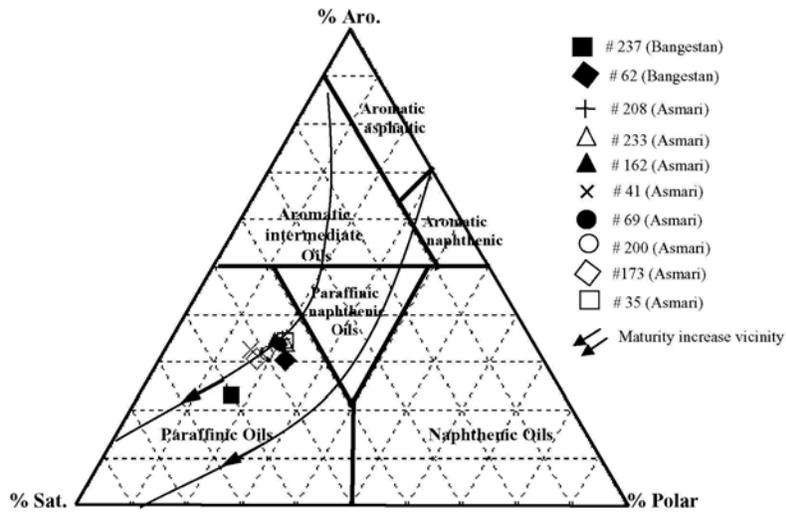


Fig. 3. Ternary diagram showing praffinic composition of Bangestan and Asmari crude oils from the Marun Field

In Gas Chromatograms of all 10 oil samples (Fig. 4), the carbon preference index (CPI) is around 1 (Table 2). This further indicates higher maturity of the source rocks generating the studied oils [15]. All these results are in accordance with the Rock–Eval 6 pyrolysis results reported by Alizadeh and his coworkers [12] on the probable source rocks of the Marun oilfield.

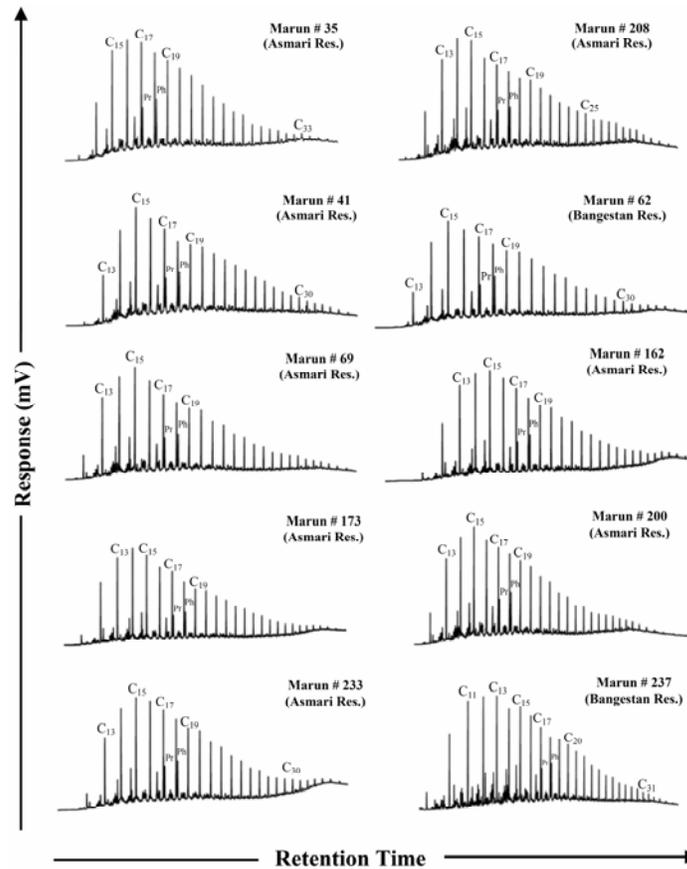


Fig. 4. Gas Chromatography of the whole oil of 10 oil samples in the Marun oilfield

Table 2. Isotopic and Geochemical data of Asmari and Bangestan reservoir oils in Marun oilfield

Well No.	FM.	Depth (m)	$\delta^{13}\text{C}_{\text{sat.}}$ (‰)	$\delta^{13}\text{C}_{\text{aro.}}$ (‰)	$\delta^{13}\text{C}_{\text{oil}}$ (‰)	$\delta^{34}\text{S}_{\text{oil}}$ (‰)	Pr/Ph	Pr/n-C ₁₇	Ph/ n-C ₁₈	CPI	CV
# 62	Ban.	3428-3450	-27.27	-26.61	-26.92	-2.57	0.78	0.37	0.53	0.94	-1.73
#237	Ban.	3316.5-3382	-27.07	-26.33	-26.77	-0.6	0.91	0.42	0.54	0.95	-1.61
#35	Asm. (n- cont.)		-27.23	-26.75	-27.02	-3.61	0.83	0.36	0.5	0.94	-2.14
#173	Asm. (n- cont.)		-27.33	-26.78	-27.10	-4.09	0.89	0.3	0.42	0.96	-1.95
#200	Asm. (n- cont.)		-27.27	-26.52	-26.95	-2.44	0.83	0.37	0.5	0.91	-1.53
#208	Asm. (Cont.)	3407-3480	-27.26	-26.24	-26.82	-0.31	0.85	0.42	0.51	0.99	-0.93
#233	Asm. (Cont.)	2964-3225	-27.26	-26.44	-26.88	-1.82	0.89	0.35	0.45	0.95	-1.37
#162	Asm. (Cont.)	3501-3722	-27.29	-26.27	-26.91	-2.31	0.8	0.32	0.46	0.96	-0.92
#69	Asm. (Cont.)		-27.29	-26.34	-26.95	-2.19	0.91	0.4	0.5	0.9	-1.08
#41	Asm. (Cont.)	2882-2972	-27.26	-26.5	-26.93	-2.6	0.85	0.38	0.54	0.95	-1.51

FM- Formation; Ban. – Bangestan Reservoir; Asm. (n-cont.)- Asmari Reservoir (non-contaminated with H₂S gas); Asm. (Cont.)- Asmari Reservoir (contaminated with H₂S gas).

Organic rich anoxic carbonate sequences in a very high reducing environment generally form oils with a ratio of Pristane to Phytane (Pr/Ph) less than 2 [22, 23]. For the studied oil samples this ratio is less than 1 (Table 2). To determine the source of the studied oils, a plot of Pr/nC₁₇ versus Ph/nC₁₈ was used [24]. The ratios demonstrate the algal source and kerogen type II with medium high maturity of the original organic matter. Again these data are in accordance with our previous results. In all Asmari and Bangestan oil samples the biodegradation phenomena is inactive since paraffin is high in oil samples and also the isoprenoids (Pristane and Phytane) are less than their adjacent normal alkanes (Fig. 5).

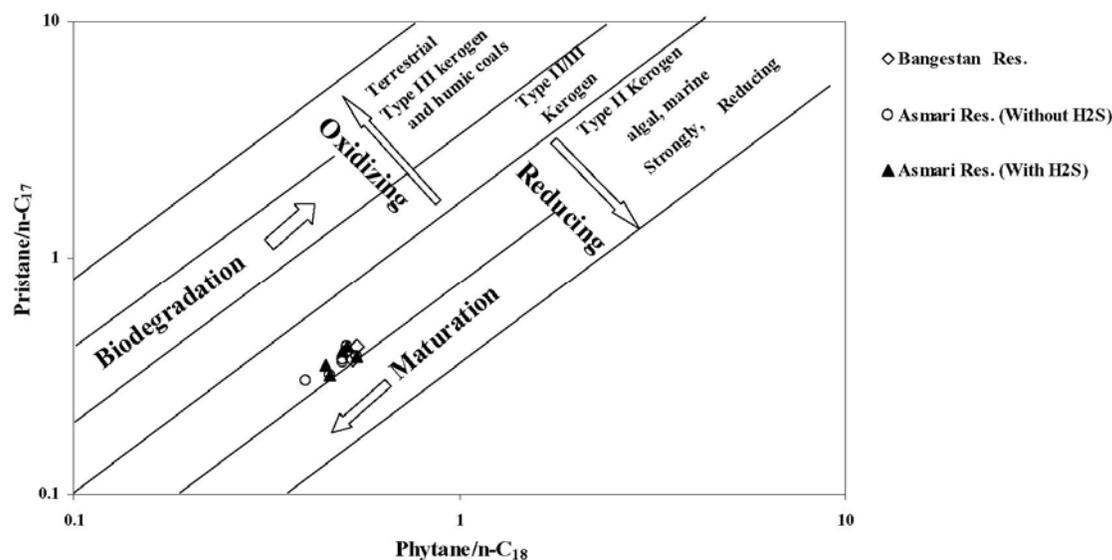


Fig. 5. Pristane/n-C₁₇ versus Phytane/n-C₁₈ showing the marine source (type II Kerogen with strongly reducing environment) for Asmari and Bangestan Reservoir oils in Marun Oilfield (After Connan & Cassou [24])

To correlate oil fingerprinting of both the reservoirs and evaluate their origin, a sensitive star diagram was used [25]. This correlation accurately shows their unique source, from which they have been generated (Fig. 6).

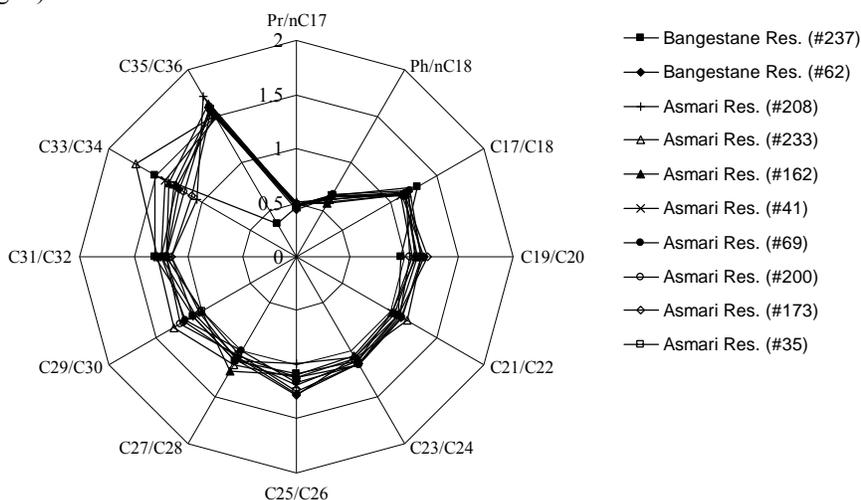


Fig. 6. Star diagram of biomarker ratios in Asmari and Bangestan Reservoirs, showing significant similarity in all the oil samples studied from Marun oilfield

4.2. Stable Carbon and Sulfur Isotopes

The plot of $\delta^{13}\text{C}$ values of aliphatic and aromatic hydrocarbon fractions of the oils [26] examined in this study show their sources have originated from a similar marine organic matter (Fig. 7). The difference between the two equations of terrigenous and marine oils was evaluated statistically and a statistical parameter, CV (the canonical variable),

$$CV = -2.53 \delta^{13}\text{C}_{\text{sat.}} + 2.22 \delta^{13}\text{C}_{\text{aro.}} - 11.65,$$

was used to distinguish between marine and terrigenous oils. CV values larger than 0.47 indicate a predominantly terrigenous organic source for the oil, whereas CV values smaller than 0.47 indicate mostly a marine organic source [26].

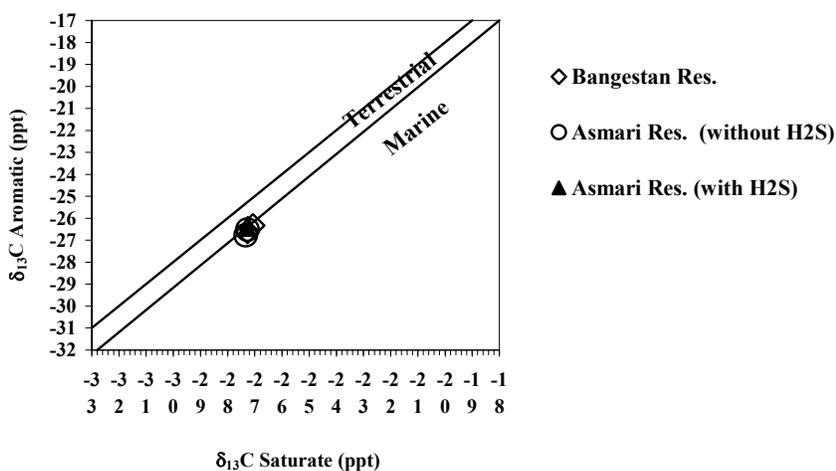


Fig. 7. A Sofer plot of $\delta^{13}\text{C}$ values for the aromatic and saturate hydrocarbons in Asmari and Bangestan Reservoirs. All $\delta^{13}\text{C}$ values are in ppt(%) relative to PDB

Ratio of Pr/Ph can show a correlation with the isotopic composition of the oil, namely with the CV. The plot of the CV data versus the log of Pr/Ph (Fig. 8) show a positive trend between the two parameters: high Pr/Ph ratios are associated with terrigenous oils having high values of CV. Low Pr/Ph ratios are associated with marine oils along with low values of CV, indicating that terrigenous sources are more commonly oxidized in comparison to marine sources before they are incorporated into thick sedimentary sections [26].

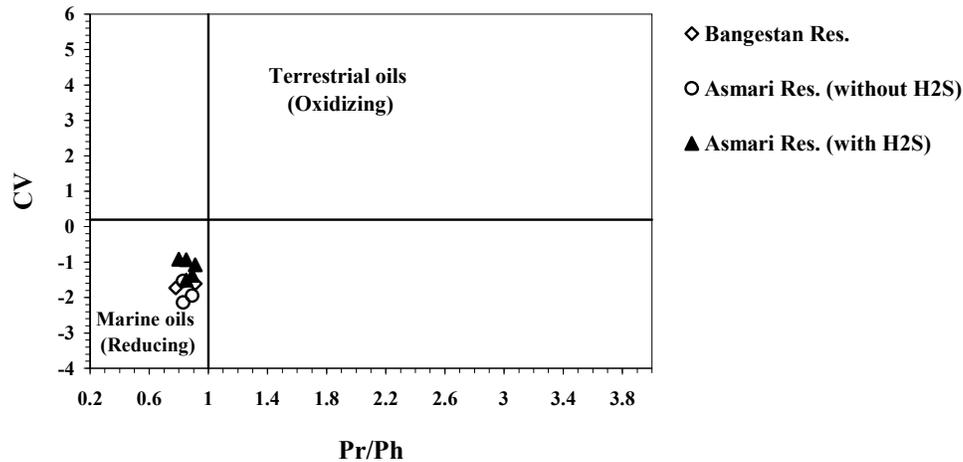


Fig. 8. Canonical variable (CV) versus Pristane/Phytane (Pr/Ph) ratio

Pristane/Phytane versus stable carbon isotope ratio of whole oil can be used to support genetic relationships among oils and infer depositional environments [27]. The plot of Pr/Ph versus whole oil stable carbon isotope ratio indicates Mesozoic argillaceous carbonates (Fig. 9). Interestingly, all samples from Asmari and Bangestan reservoirs show similar source rock. These results are supported by the Rock-Eval 6 results reported by Alizadeh and his coworkers [12].

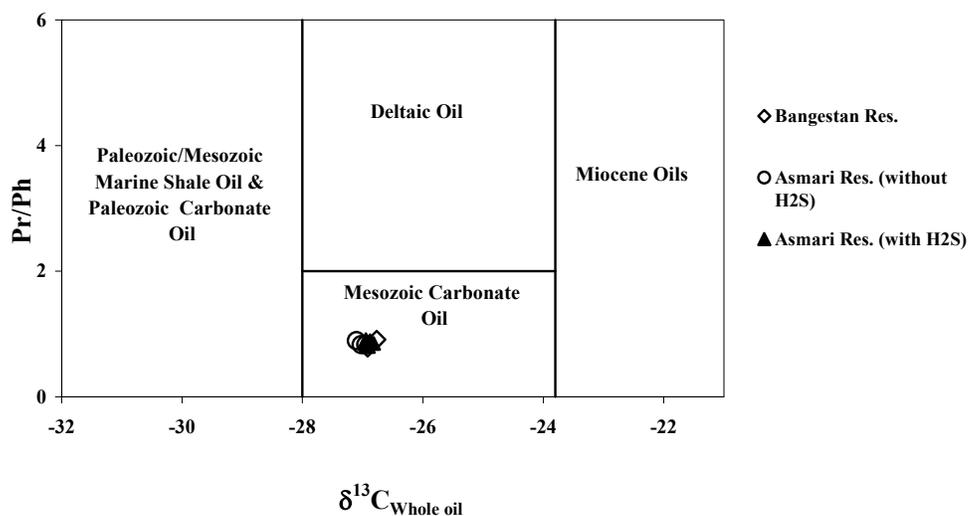


Fig. 9. Bivariate plot of $\delta^{13}\text{C}$ ratio of oil versus Pr/Ph ratio

The shapes and trends of stable carbon isotope type curves [28, 29] can be used to identify the relationship between different crude oils. The curves of the studied saturate, whole oil and aromatic fractions demonstrate three different oil families (Fig. 10). Oils from the Asmari reservoir unpolluted with H_2S gas are depleted with $\delta^{13}C$ in all type of curves belonging to whole oil saturate and aromatic fractions. Polluted Asmari oils with H_2S gas along with one of the Bangestan reservoir demonstrate a similar oil family. The third family comprises only one oil sample from the Bangestan reservoir producing well (Well No. 237). These oil families show the following characters:

1. Asmari reservoir oils not contaminated with H_2S gas comprise the least stable carbon isotope in whole oil and fractions as well.
2. Asmari reservoir oils contaminated with H_2S gas have a similar trend along with stable carbon isotope of well no.62 Bangestan reservoir. This shows mixing of oils from both reservoirs in that area.
3. In well no.237 of Bangestan reservoir the stable carbon 13 is showing enrichment in whole oil as well as saturate and aromatic fractions.

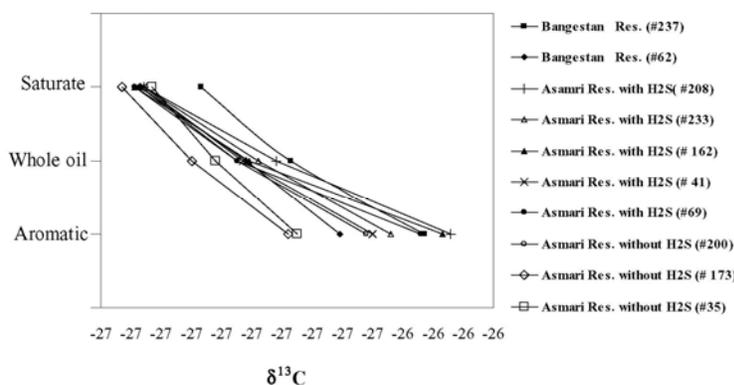


Fig. 10. Galimov curves of Asmari and Bangestan Reservoir oils

To correlate Asmari and Bangestan reservoir oils and the possibility of their mixing, carbon isotope ($\delta^{13}C$) versus sulfur isotope ($\delta^{34}S$) values were used (Fig. 11). The amounts of stable sulfur isotope in oils containing H_2S gas, produced by thermal sulfate reduction (TSR) or by bacterial sulfate reduction processes, illustrate more enrichment in $\delta^{34}S$ compared to H_2S gas generated by organic sulfur [30].

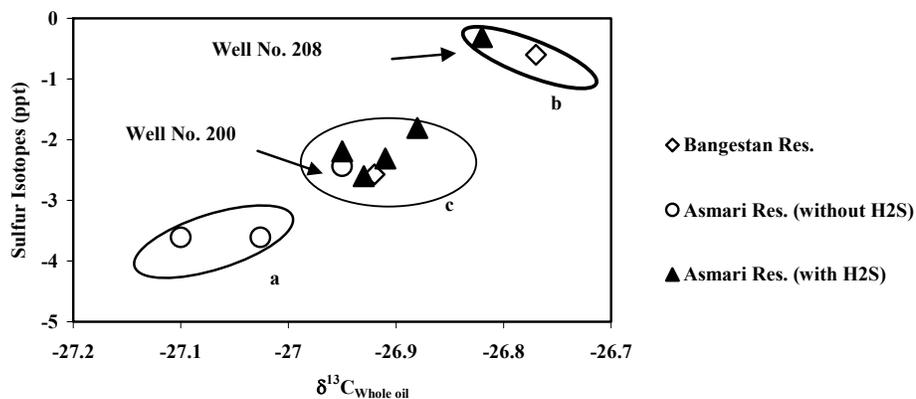


Fig. 11. Bivariate carbon and sulfur isotopes of Asmari and Bangestan Reservoir oils. Here, clearly three oil families are demonstrated. Asmari non-contaminated oils along with Bangestan oils (a, b), Asmari H_2S contaminated oils is in an intermediate position (c)

In this correlation non polluted Asmari reservoir oils to H₂S gas show depletion in $\delta^{34}\text{S}$ compared to polluted Asmari oils, as well as Bangestan reservoir oils (Fig. 11). The geothermal gradient of the Bangestan reservoir in the Marun oilfield is around 110-130°C [17]. This high reservoir temperature practically indicates the absence of bacterial activity. GC results also support this interpretation. It is concluded that the H₂S gas source in Bangestan reservoir oils could be from Gotnia anhydrite formation which is located in the deeper part of the studied reservoirs and the gas geneses could be related to thermal sulfate reduction phenomena. Similar conclusions are reported by Orr [31] and Vredenburg and Cheney [32]. Contaminated Asmari oils to H₂S gas fall within the vicinity of Bangestan oils (Fig. 11). Structural observations from the Marun oilfield reveal the existence of fissures and fractures in Asmari and Bangestan reservoirs. These structural features are responsible for the interrelation of the two reservoirs [17]. In Fig. 11 well no. 208 is producing from the deepest Asmari pay zone; amazingly its oil is falling within the Bangestan reservoir oil (well no. 237), which is also located in the deeper part. Finally, well no. 200, which was reported as non-contaminated Asmari oil, shows signs of pollution with similar isotopic results as contaminated Asmari and Bangestan oils. The location of this well draws undivided attention to the spread of H₂S gas pollution from the southeast to the northwest of the studied oilfield, and this must be carefully considered in future investigations.

5. CONCLUSIONS

The Asmari and Bangestan reservoir oils in the Marun oilfield have similar argillaceous carbonate source rock, deposited under severe reducing environment. Geochemical parameters from the studied oils in both reservoirs show middle to high maturity and inactivity of biodegradation with high paraffin content. Isotopic investigations also show a reducing environment of both reservoir oils. Stable carbon isotope in whole oil and saturate as well as aromatic fractions, for the first time, demonstrated three distinct oil families. Also, contaminated oil with H₂S gas has similar isotopic characters as Bangestan oils, indicating a similar source of pollution to be from the Bangestan reservoir or the common source rock of both Bangestan and Asmari reservoirs. It is worth noting that pollution to H₂S gas is spreading from the southeast to the northwest of the Marun oilfield.

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