

Biological marker geochemistry of selected oils and possible source rocks from central Alborz Basin

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Abstract

Surface oil seeps from Alasht, Lapur and Jenesem share many compositional affinities including distributions and concentrations of normal alkanes, terpanes and steranes. These oils also show many similarities with that of Jurassic sediments (Shemshak Group, Klariz Formation) systematically sampled from the Galandrud Coal Mine. Recent geochemical studies in this area indicated that the Shemshak Group is rich in organic matter and has potential to generate oil and chiefly gas. The Kalariz Formation here is composed of 350 m of mudstones, siltstones and fine-grained sandstones with over 30 coal seams. This formation is thought to have sourced Alasht and Lapur oil seeps. GC and GC-MS chromatograms of Jenesem oil are unique and show no any similarities with that of above mentioned oils or the Shemshak Formation. This indicates that the Jenesem oil has been generated from a different source rock but its origin is still unknown.

Keywords: Alborz Range, Shemshak Group, Oil seep, Source rock, Correlation, Biomarker.

Introduction

Southern regions of the Caspian Basin accommodate several significant hydrocarbon reservoirs; however, the source rocks of these hydrocarbons have not been adequately investigated. Since debut of the petroleum explorations in west of the Caspian Basin in 1947, several major oil and gas reserves have been discovered (Lebedev, 1991). The middle Pliocene sediments in this basin had been considered as the potential source rocks for the hydrocarbon reservoirs in this area. A detailed regional geological and geochemical study performed by Narimanove (1994) showed that Middle Pliocene sediments formerly thought as probable oil and gas source rocks in the southern Caspian Basin contain only about 0.3-0.6 kg/tonne of organic matter (OM). His investigations also revealed that in the southern Caspian, sediments up to 9 km depth could be important source rocks for hydrocarbon reserves of this area, and hydrocarbon generation is not entirely limited to deep central parts of this basin. In addition, geochemical studies supported by isotope and trace elements suggests vertical hydrocarbon migration. In fact, concurrent geochemical studies conducted by Kamali (1997) on Cheleken Series at exploration well (Khazar-1) drilled in offshore led to the similar results and indicated that the Cheleken Series of sediments are lean in organic matter and have not reached sufficient maturity for the petroleum generation. Subsequently, Schoellkopf and his co-workers

(1997) on the basis of geochemical modeling provided significant results suggesting that the southern Caspian Basin is abnormally cool (low geothermal gradient) and has a rapid heat flow with very high rates of sedimentation and subsidence.

Thus, the uppermost oil window lies between the depth intervals of 6 to 8 km. The corresponding depth intervals for light oil, condensate and wet gas is 8 to 13 km and for dry gas is estimated to be 10 to 15 km below sea level respectively. As stated by Berberian (1983) given a thickness of 1.5 to 2 km to quaternary sediments, 5 to 8 km for middle Miocene-Pliocene and 8 to 12 for Mesozoic-Paleocene sediments on the southern Caspian Basin, the Shemshak Group must have already reached oil and gas generation window particularly at the basin's margins.

Petrographical and geochemical studies including detailed organic petrography by Zamani (1999), Shalika *et al.*, (2011) and Shekarifard *et al.*, (2009, 2012) on thick organic-rich strata of the Shemshak Group, reveals that this Group ranging from upper Triassic to Middle Jurassic in age is extended over the southern districts of the Caspian Basin, in the Alborz mountain range. The unit is chiefly composed of coal deposits and Total Organic Carbon (TOC) reaches up to 70 % in coaly intervals and maturation indices are indicative of oil generation window.

In the coastal areas of South Caspian Sea, several oil seeps have been reported for decades, and National Iranian Oil Company (NIOC) has shown

interest in the exploration of these areas. Mud-volcanoes have long been reported from this region that is closely associated with oil and gas reservoirs but have no any relationship with magmatic volcanoes. Other significant elements from petroleum exploration point of view are the existence of oil seeps found in Azerbaijan and Mazandaran provinces. Such seepages containing gas bubbles were reported from Hassan Abad, Kejur, Tough Kuh, Naftchal and Dasht-e Kala (Aminsobhani, 1968). Objectives of this study included assessment of petroleum source potential of the Shemshak Group and establishing a genetic relationship among oil seeps with the candidate source rocks using their characteristic biomarkers. This study also intends to reveal the origin of oils and classifies them into different oil families.

Geological setting

In this study, selected samples have been taken from Shemshak Group. The Upper Triassic–lower Middle Jurassic Shemshak Group is a siliciclastic unit, up to 4000 m thick, which is common and widespread across the Iran Plate generally situated

in northern and central Iran and has been long known as a source of commercial coal. The group is sandwiched between two major unconformities: the contact with the underlying platform carbonates of the Elika and Shotori formations is characterized by karstification and bauxite–laterite deposits; the top represents a sharp change from siliciclastic rocks to rocks of a Middle–Upper Jurassic carbonate platform–basin system.

Selected samples including shallow drilling cores, coal mining tunnel and freshly cut outcrop, were collected from Galandrud Coal Mine which is situated 20 Km south of Alamdeh town in the Alborz structural-geological unit (Fig. 1). Shemshak Group in this area has been divided into four formations including Ekrasar, Laleband, Kalariz and Javaherdeh (Fig. 2). Most samples have been taken from Kalariz Formation, which comprises 350 m alterations of claystone, mudstone, siltstone to fine grained sandstone, with thirty-two workable coal seams and horizons of nodular ironstone that were deposited in the fluvio-deltaic environments (Zamani, 1999).

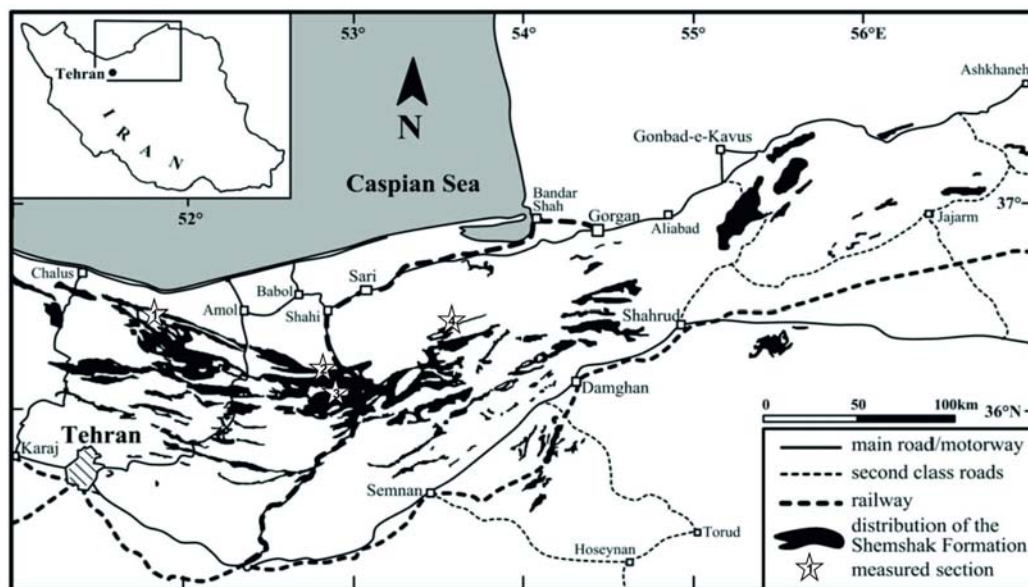


Figure 1: Location map showing the distribution of Shemshak Group outcrops (Fürsich *et al.*, 2009) with studied localities in the central and eastern Alborz Range. 1. Galandrud area; 2. Lapur oil seep; 3. Alasht oil seep; 4. Jenesem oil seep.

According to Fürsich *et al.*, (2005, 2009) studies, in the Alborz Mountains, Shemshak Group consists of a Triassic and a Jurassic unit, separated by an unconformity, which is in part angular in the northern part of the mountain range and less conspicuous towards the south. Two major facies belts, a northern and a southern belt running more

or less parallel to the strike of the mountain chain, can be distinguished. In the north (studied area), the Triassic part of the group is composed of the comparatively deep-marine Ekrasar Formation followed by the Laleband Formation, which represents prodelta–delta front environments. Up-section, the latter is replaced by the fluvial–

lacustrine, coal-bearing Kalariz Formation (Fig. 2). This lithostratigraphic scheme reflects the tectono-sedimentary evolution of the Shemshak Foreland Basin of the Alborz Mountains where, during the Late Triassic, a relict marine basin in the north became gradually infilled (Fürsich *et al.*, 2009). The Jurassic part of the group consists exclusively of the Javaherdeh Formation, coarse conglomerates of alluvial fan–braided river origin (Fig. 2).

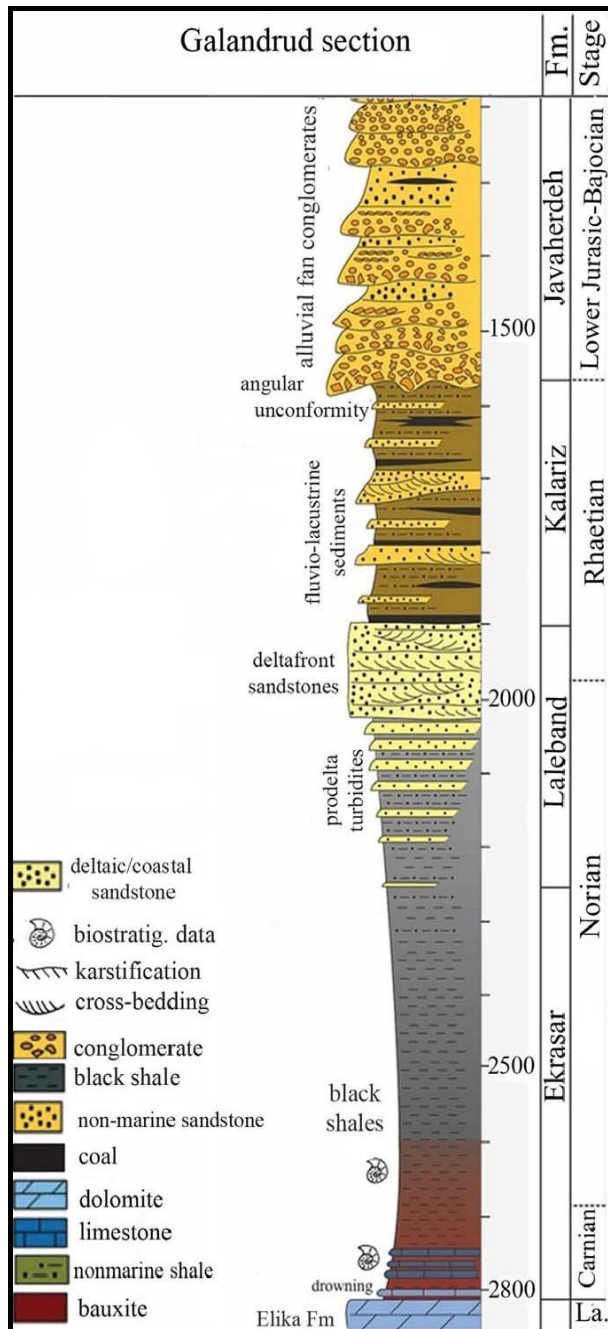


Figure 2: Stratigraphy column for the Shemshak Group and its formations (Ekraसर, Lalebанд, Kalariz and Javaherdeh) in the Galandrud area (Fürsich *et al.*, 2009).

The organic-geochemical characteristics of oils from three surface seeps in the Central Alborz Basin were also analyzed using standard techniques. These oils were collected from Lapur, Alasht and Jenesem in Mazandaran Province (Fig. 1). The oils were compared with each other (oil to oil correlation) and were correlated against the Kalariz Formation oil extract (oil to source correlation).

Material and Methods

A total of eighty samples from shallow drilling cores, coal mining tunnel and freshly cut outcrop have been investigated for evaluation of petroleum generation potential and thermal maturity of the Kalariz Formation. In order to evaluate source rock, these samples were subjected to oil show analyzer (Rock-Eval II) instrument. Twenty samples out of eighty, which were analyzed by Rock-Eval pyrolysis and showed higher potential for hydrocarbon generation, have been selected for detail organic petrography. The petrography observations have been performed on kerogen concentrates and coals using transmitted, reflected white and UV light microscopy. Measurements of vitrinite reflectance were made on polished blocks using a Leitz-MPM SP microscope.

In order to establish oil to oil and oil to source correlation, selected rock extracts (n=3) from the Kalariz Formation and oil from Lapur, Alasht and Jenesem seepages (n=3) were subjected to soxhlet extraction, thin layer chromatography, gas chromatography (GC) and gas chromatography-mass spectrometry (GCMS). The Chrompack-CP-9000 gas chromatograph was used to analyze the saturated fraction. The instrument was fitted with a 25m x 0.22mm column. The oven temperature was held at 60°C for 5 minutes then programmed from 60 to 260°C at 4°C/min steps. Helium was used as the carrier gas. The GC-MS analyses were undertaken using a Varian 3400 Gas chromatograph interfaced with a Finigann 8430 mass spectrometer.

Discussion:

The results of Rock-Eval pyrolysis indicate that Kalariz Formation in the Galandrud Coal Mine is composed of kerogen type III and IV (Figs. 3, 4). Total organic carbon (TOC) content in organic-rich beds is generally greater than 0.5% (Fig. 4) and reaches up to 72% in beds associated with coal veins. These source beds are thermally mature and represent oil generation window (or beginning of

catagenesis). Their Tmax values are often between 435 and 450°C (Fig. 3). Most samples have potential to generate wet gas, which is reflected by their hydrogen index (HI ≤ 150). However, few samples with relatively higher HI are capable of producing commercial quantity of liquid hydrocarbons (Zamani, 1999; Shekarifard *et al.*, 20011).

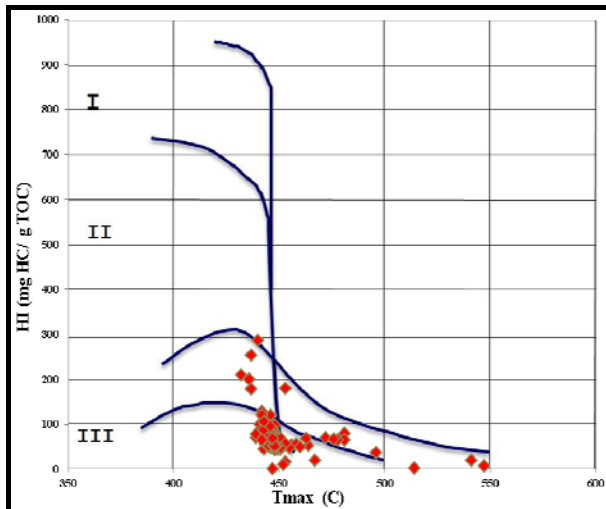


Figure 3: HI versus Tmax diagram for samples from Galandrud area. Most samples are representing catagenesis stage and kerogen type III-IV.

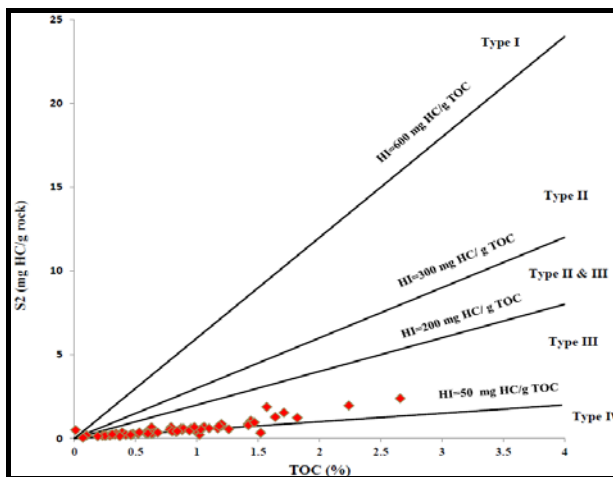


Figure 4: S2 versus TOC diagram for samples from Galandrud area. Most samples are representing kerogen type III-IV.

Petrographic observation of organic matter under transmitted and reflected lights indicates that the organic matter is mainly composed of vitrinite and inertinite and to a lesser extent liptinite. The amount of liptinite group of macerals in some samples is considerable and this view is true particularly where the resinite occurs as dominant maceral. The measured vitrinite reflectance (Ro%)

varies between 0.6 to 0.8% and corresponding thermal alteration index (TAI) values are in the range 2+ to 3- (Zamani, 1999). Under ultra violet excitation, the liptinite group of macerals shows pale yellow to dark yellow colors. The results of TAI, Ro and fluorescence study are all consistent and suggest that these sediments have reached early stages of oil generation window (Zamani, 1999; Shekarifard *et al.*, 20011).

Bitumen extracts from selected organic-rich shales are generally rich in resin and aromatic fractions compare to saturate and asphaltene fractions (Table 1). Gas chromatograms of saturated hydrocarbons display typical normal alkane distribution in a sense, that relatively high concentration of normal alkanes occurs at range C₁₉ and C₂₅ with odd number preference. In addition, noticeable concentration of an unknown compound shown by asterisk (Fig. 5) elutes just before C₂₅ in all Galandrud samples.

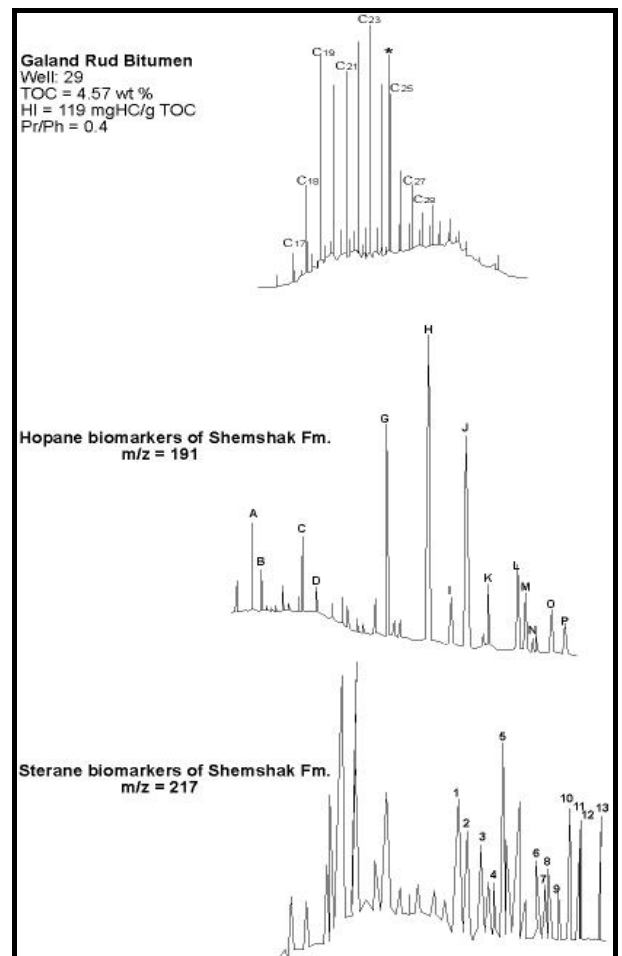


Figure 5: Gas chromatography and mass fragmentograms of steranes (m/z=217) and triterpanes (m/z=191) for a selected sample from Shemshak Formation.

Pristane to phytane ratio is about 0.33 suggesting highly reducing condition existed during deposition of source rock, which is somewhat unusual for deltaic sediments. However, strong odd to even

predominance particularly at range C₂₁ to C₂₇ is clear, which suggests that the bulk of organic matter was derived from land plants.

Table 1: Summary of rock extracts for The Shemshak Formation from Galandrud Coal Mine.

Borehole No.	Depth (m)	EOM (%)	Paraffin (%)	Aromatic (%)	Resin (%)	Asphaltene (%)	Total (%)
6	484	0.042	21.67	35	39.18	2	97.85
20	138	0.048	20	34	37.3	6	99.3
29	187.25	0.052	25	35	32	5	97

Oil to oil and oil to source correlation studies

In conjunction with detailed source rock analyses, few oil seeps from Lapur, Alasht and Jenesem were subjected to GC and GC-MS analyses in order to identify their geochemical fossils and establish oil-oil and oil-source correlation study. Alasht and Lapur oil seeps share many similarities from the point of view of terpane and sterane biomarker distribution. Representative mass fragmentograms are shown in Figures 6 and 7 (see also Table 2). In m/z 191 mass fragmentogram, 17 α (H), 21 β (H)-30-norhopane is the highest peak for Lapur but 17 α (H), 21 β (H)-hopane predominates over 30-norhopane in Alasht oil seep. A smooth decrease in the homohopane (C₃₁-C₃₅) profile with increasing carbon number is also typical for all three samples. In these samples 18 α (H), 22, 29,30-trisnorneohopane (Ts) occurs at higher concentration relative to 17 α (H), 22, 29,30-trisnorhopane (Tm) and their Tm/Ts ratios vary between 0.5 and 0.6. It is known that Ts is more stable to thermal maturation than Tm (Seifert & Moldowan, 1978). Peters and Moldowan (1993) believes that the Tm/Ts ratio (sometimes reported as Ts/(Ts+Tm)) is most reliable as a maturity indicator when evaluating oils from a common source of consistent reliable maturity indicator when evaluating oils from a common source of consistent organic facies. Therefore, this ratio has to be used with caution because it is often source and facies dependent (McKirdy *et al.*, 1984) and decreases in an anoxic carbonate section (Peters & Moldowan, 1993) and increases above 0.9 % Ro (Van Grass, 1990). An appreciable concentration of an unknown compound (possibly gammacerane) elutes after C₃₁ homohopane. In addition, these oil seeps are characterized by having high amounts of tricyclic terpanes (Fig. 6).

The C₂₀-C₂₆+ tricyclic terpanes are molecular fossils of bacterial lipids. Their abundance relative to each other (e.g. C₂₃/C₂₀-C₂₄ = 0.32-0.45) and to

tetracyclic terpanes (e.g. C₂₃ tricyclic/C₂₄ tetracyclic = 1.5-2.2) are known to be source-related and therefore potentially useful in future oil-oil and oil-source correlations (McKirdy, 1994).

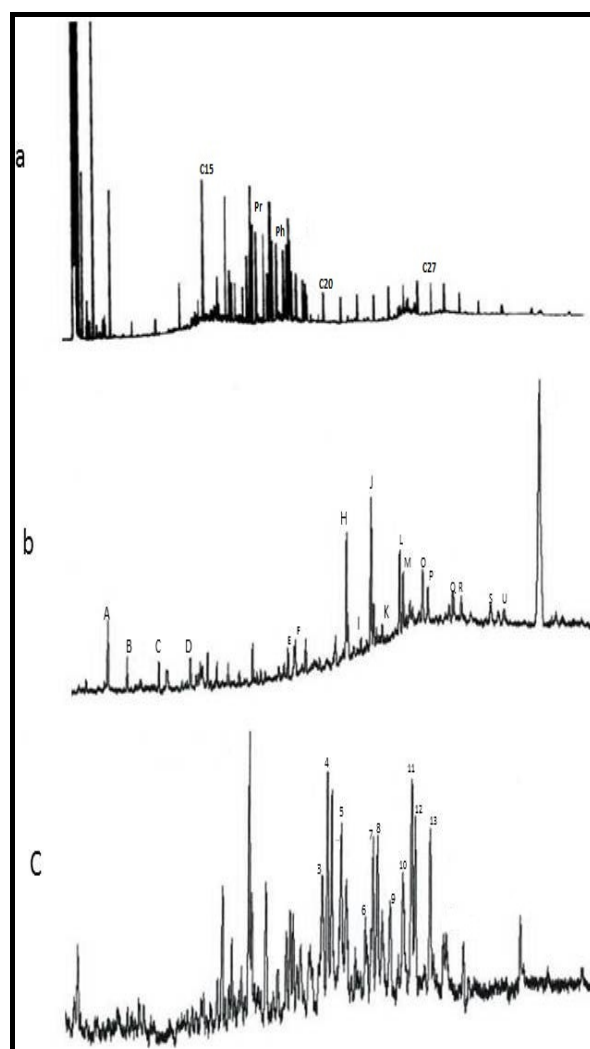


Figure 6: Gas chromatogram (a) and mass fragmentograms of steranes (b, m/z=217) and triterpanes (c, m/z=191) for a selected oil seep (Alasht). Pr: Prystane, Ph: Phytane.

In fact, Seifert and his co-workers (1980) found a strong genetic relationship between the Sag River

oil and Shublik shale (Jurassic source rock) on the basis of their relative amounts of tricyclic terpanes.

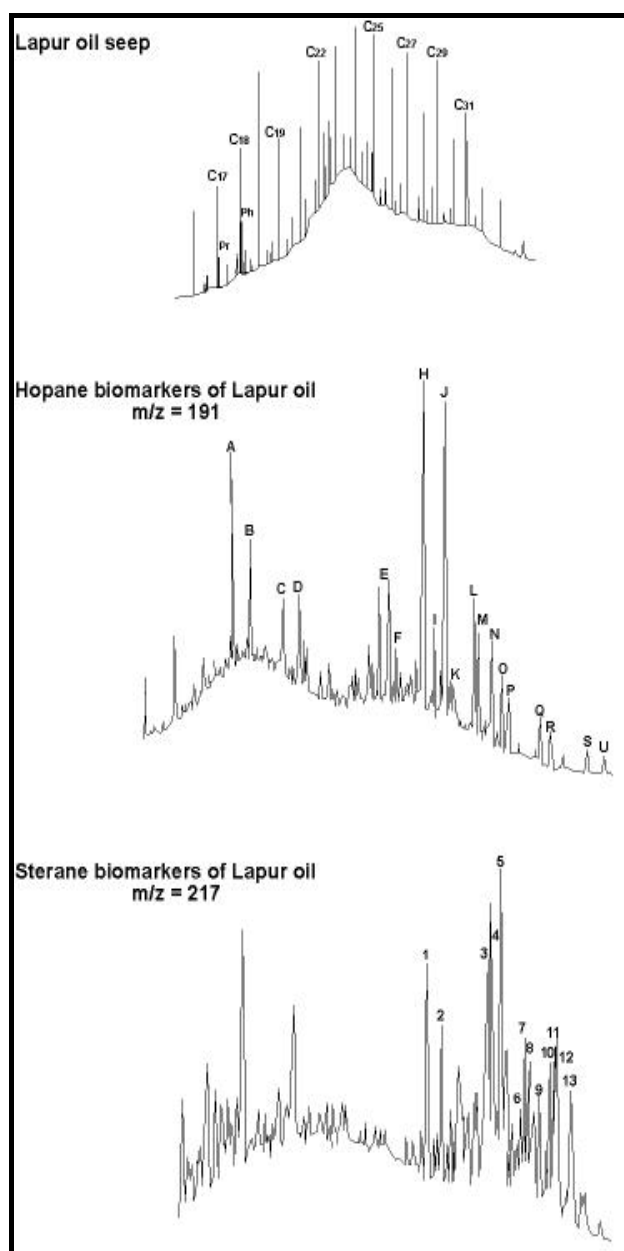


Figure 7: Gas chromatography and mass fragmentograms of steranes ($m/z=217$) and triterpanes ($m/z=191$) for a selected oil seep (Lapur).

In addition, many workers have reported that C_{19} - C_{45} terpanes are highly resistant to biodegradation, surviving even when hopanes are removed (Reed, 1977; Seifert & Moldowan, 1979; Palacas *et al.*, 1986; Connan *et al.*, 1980). Also, Zhusheng *et al.*, (1990) compared related unaltered and severely biodegraded oils from the Kelamayi oil field in northwestern China chiefly on the basis of tricyclic

and tetracyclic terpanes, gammacerane and 8, 14-secohopanes. These compounds were found in high concentration in biodegraded oil, but were absent in the unaltered oil.

As indicated by almost identical $C_{32}S/(S+R)$ ratios (0.5-0.6) the isomerization of homohopanes have reached equilibrium values in all the studied samples. In addition, C_{24} tetracyclic/ C_{30} hopane ratios range from 0.2 to 0.4 suggesting that restricted and anoxic conditions were prevailed during deposition of their source rocks. Ratios of tetracyclic terpanes/hopanes increase in more mature source rocks and oils, indicating greater stability of the tetracyclic terpanes (Peters & Moldowan, 1993; Aquino Neto *et al.*, 1983).

Although, the higher concentration of C_{24} tetracyclic terpane has been reported as a marker for carbonate and evaporate depositional environments (Palacas *et al.*, 1984; Connan *et al.*, 1986; Connan *et al.*, 1987; Mann *et al.*, 1987; Clark & Philip, 1989) this compound was also reported from Australian oils believed to be generated from terrigenous organic matter (Philip & Gilbert, 1986). A striking feature associated with Kalariz Formation (bitumen extract) is the occurrence of noticeable quantities of 28, 30-bisnorhopanes. This compound is typical of organic matter from highly reducing to anoxic environment (Peters & Moldowan, 1993). This view is entirely in good agreement with low values of pristane/phytane ratio (<0.6) which is often used to indicate as redox potential of the source sediments.

All m/z 217 mass fragmentograms are dominated by C_{29} and C_{27} diasterane peaks (Fig. 6). The proportion of $C_{27}:C_{28}:C_{29}$ regular steranes is similar for all samples ($C_{27}>C_{28}<C_{29}$). The $S/(S+R)$ ratios for the C_{29} steranes are close to equilibrium values (0.45-0.47). The [C_{29}/C_{27} $5\alpha(H)$, $14\alpha(H)$, $17\alpha(H)$ (20R)-sterane] ratios range between 1 and 1.3 suggesting that algal and land plants were almost equally available during source rock deposition (Haung & Meinshein, 1979). On the other hand, the Jenesem oil has entirely different geochemical fossils including higher pristane/phytane ratio (~ 2) and considerable concentrations of 28,30-bisnorhopane, $18\alpha(H)$ -30-norneohopane ($C_{29}Ts$), $18\alpha(H)$ -oleanane and C_{29} regular steranes (Fig. 8). These features are unique to the Jenesem oil and there seems to be no any positive correlation between this oil and other previously mentioned oils and the Kalariz Formation either.

Table 2: Selected biomarker ratios for oil seepages (Alasht, Sangnisht, Lapur) with a common source.

Steranes	Percentage and Ranges	m/z	Specificity
C ₂₇ :C ₂₇ :C ₂₈ :C ₂₉ αββ sterane	40:24:36	217	
	36:28:36	217	Source
	37:31:32	217	
C ₂₉ /C ₂₇ sterane (20R)	1-1.3	217	Source
C ₂₉ sterane 20S/20S+20R	0.45-0.47	217	Source, maturity
Triterpanes (hopanes)	Ranges	m/z	Specificity
C ₂₇ Tm/Ts	0.5-0.6	191	Maturity, source
C ₃₂ 22S/22R bishomohopane	1.1-1.2	191	Maturity
C ₂₃ /C ₂₀ -C ₂₄ tricyclic	0.45	191	Source
C ₂₃ tricyclic/C ₂₄ tetracyclic	2.1-2.2	191	Source
C ₂₄ tetracyclic/C ₃₀ hopane	0.27-0.4	191	Source

Conclusions

The biomarker fingerprints of oil seeps (Alasht and Lapur) from the Mazandaran Province are all consistent and share many compositional affinities with each other. These include distribution and concentration of normal alkanes, terpanes and steranes. Therefore, it is safe to assume that the above mentioned oils have a common origin and have been generated from a single potential source rock. As already discussed, Kalariz Formation (Shemshak Group) in the Galandrud Coal-Mine area showed to be a potential source rock. The biomarker characteristics of this formation including distribution of normal alkanes, isoalkanes, steranes and terpanes are all similar with

those of previously mentioned oil seeps. Interestingly, noticeable concentration of an unknown compound which occurs just before C₂₅ normal alkane is considered as a characteristic feature of all rock extracts and oil seeps. Moreover, lower Pr/Ph ratios (0.4-0.6) and high concentration of C₁₉ normal alkane are other diagnostic features associated with the above oil and rock samples. Finally, the Jenesem oil showed no any compositional affinity with other group of oils and Kalariz Formation either. This suggests the presence of at least two groups of oil families generated from two different source rocks at the Mazandaran Province located in Central Alborz.

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APPENDIX

Identities of common triterpanes (hopanes and moretanes) in the m/z 191 mass chromatograms (GC-MS data).

Peak No.	Identification	Carbon No.
A	C ₂₃ Tricyclic terpane	23
B	C ₂₄ Tricyclic terpane	24
C	C ₂₅ Tricyclic terpane	25
D	C ₂₄ Tetracyclic terpane	24
E	18 α -22,29,30-trisnorhopane (T _s)	27
F	17 α -22,29,30-trisnorhopane (T _m)	27
G	17 α -28,30-bisnorhopane	28
H	17 α -norhopane	29
I	17 β -normoretane	29
J	17 α -hopane	30
K	17 β -moretane	30
L & M	17 α -homohopane (22S + 22R)	31
N	Gammacerane	30
O & P	17 α -homohopane (22S + 22R)	32
Q & R	17 α -homohopane (22S + 22R)	33
S & U	17 α -homohopane (22S + 22R)	34

Identities of common steranes and diasteranes in the m/z 217 mass chromatograms (GC-MS data).

Peak No.	Identification	Ring stereochemistry	Carbon No.
	Regular steranes		
3	5 α (H)14 α (H)17 α (H) 20S	$\alpha\alpha$	27
4	5 α (H)14 β (H)17 β (H) 20R + 20S	$\beta\beta$	27
5	5 α (H)14 α (H)17 α (H) 20R	$\alpha\alpha$	27
6	5 α (H)14 α (H)17 α (H) 20S	$\alpha\alpha$	28
7	5 α (H)14 β (H)17 β (H) 20R	$\beta\beta$	28
8	5 α (H)14 β (H)17 β (H) 20S	$\beta\beta$	28
9	5 α (H)14 α (H)17 α (H) 20R	$\alpha\alpha$	28
10	5 α (H)14 α (H)17 α (H) 20S	$\alpha\alpha$	29
11	5 α (H)14 β (H)17 β (H) 20R	$\beta\beta$	29
12	5 α (H)14 β (H)17 β (H) 20S	$\beta\beta$	29
13	5 α (H)14 α (H)17 α (H) 20R	$\alpha\alpha$	29
	Diasteranes		
1	20S diasteranes	$\beta\alpha$	27
2	20R diasteranes	$\beta\alpha$	27