

Original article

Biological marker indicators of paleo-depositional environment for the Eocene formations around Zara-Bolucan region (central anatolia, Turkey)

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Abstract:

Biological marker (biomarker) characteristics and paleo-depositional conditions of Eocene sediments around Zara-Bolucan northeast of Sivas Basin have been determined by means of detail organic geochemical studies such as gas chromatography (GC) and gas chromatography-mass spectrometry (GC-MS) analysis. In the investigated area, northeast section of the Sivas Basin, the Eocene Bozbel and Kozluca Formations consist of a organic matter rich sedimentary rocks deposited within transgressive-regressive cycles. During the evaluation of paleoenvironment characteristics with biomarker data, data related to n-alkanes, isoprenoids, terpenoids and steroids were used. Especially, the marine environment features indicated by C_{23} tricyclic/ C_{30} hopane, the C_{26}/C_{25} tricyclic terpanes, homohopane/ C_{30} hopane and diasterane/regular sterane data. The ratios of Pr/Ph, Pr/n- C_{17} and Ph/n- C_{18} indicate that there is marine organic matter predominantly precipitated under reducing environmental conditions. In the studied samples, the algal-bacterial organic matter contribution was interpreted to be due to the superiority of the C_{23} tricyclic terpene to C_{24} , as well as larger than the C_{24} tricyclic terpene. However, the gammacerane index suggests that there is initially a low salinity environment. The significant terrestrial organic matter contribution is indicated by the isoprenoid and biomarker ratios, dominance of higher n-alkanes such as n- C_{29} , n- C_{31} , CPI, TAR values, n- $C_{19}/n-C_{31}$, relative abundance of $C_{27}-C_{28}-C_{29}$ regular steranes. Suboxic to anoxic paleo-depositional conditions are confired by the isoprenoid and n-alkane ratios and T_m/T_s , $C_{29}T_s/C_{29}$ hopane ratios.

1. Introduction

There is a constantly growing need for fossil fuels in the world, which is often provided by sources such as oil, natural gas and coal. Many different methods are used to exploration these types of sources, and organic geochemical methods are vital for hydrocarbon investigation. Today's exploitation and production can be especially helpful in increasing the prediction efficiency of organic geochemistry as decreasing petroleum reserves and the lack of potential fields become increasingly difficult. Turkey also is one of the world's countries with the limited amount of fossil fuel reserves as many countries. In particular, oil reserves are limited to Southeast Anatolia and gas reserves are limited to those in the Thrace Basin. For this reason, the search for new resources of the country and the studies related to it continue with different perspectives. The study area is located in the Sivas Basin in the northeast section, it is one of the major sedimentary basins in Turkey (Fig. 1a) (Görür et al., 1998). In this basin, the

sediments range from Upper Cretaceous to Pliocene in age, their thickness is very variable, and may reach up to 8000 m (Kurtman, 1973) and the basin fill is mostly composed of fine-grained sediments such as shales and limestones. The basement is composed of metamorphics which outcrop in the northwestern part of the basin and an ophiolitic complex to the south (Kurtman, 1973; Görür et al., 1998; Özçelik, 2000).

The basin developed unconformably over the Paleozoic-Mesozoic basement and Eocene-Oligocene sediments (Fig. 1b and 1c). It is one of the sedimentological, tectonic and hydrocarbons-related basins that have been used for various geological (stratigraphic, paleontological, tectonic) researches (Artan and Sestini, 1971; Gökçen, 1981; İlleez et al., 1990; Korkmaz, 1990; Özçelik and Altunsoy, 1992; Altunsoy and Özçelik, 1995; Özçelik and Altunsoy, 1996; Ocakoğlu, 2001; Çiner et al., 2002; Yılmaz and Yılmaz, 2006; Yalçın Erik et al., 2015).

In particular, the main reason for the investigation of the Eocene units is to determine the levels of medium to



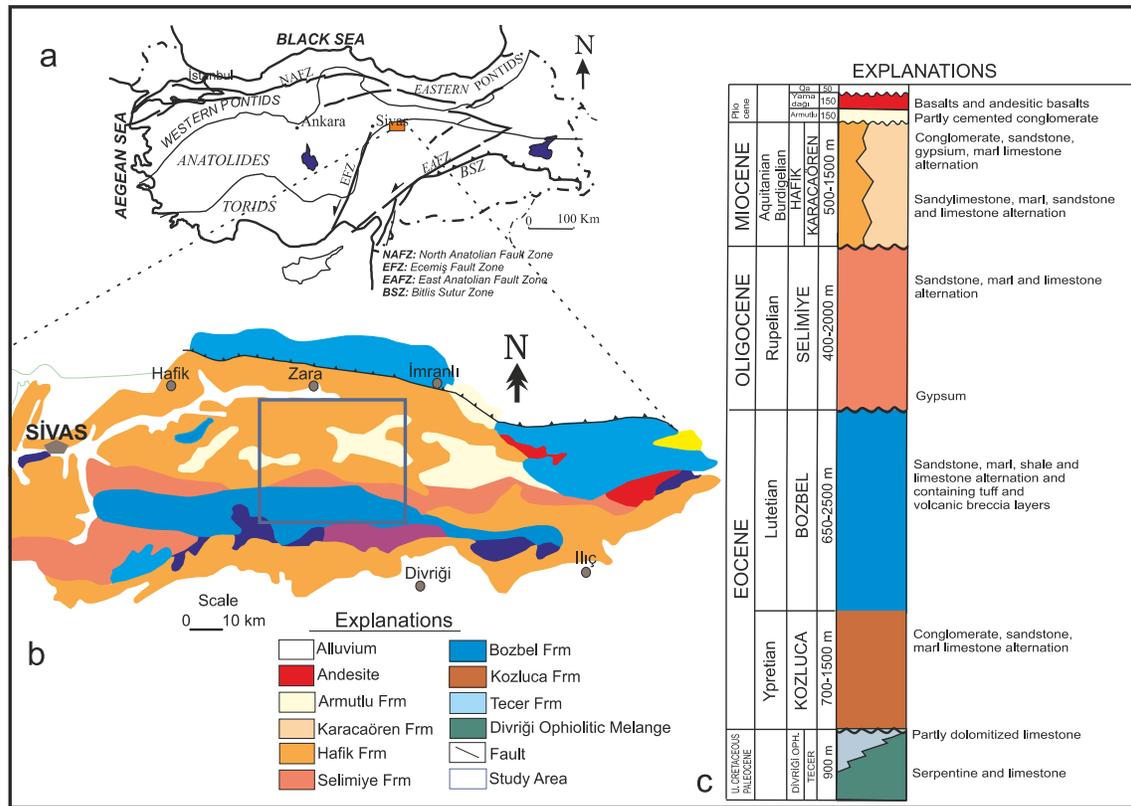


Fig. 1. Location map, simplified geological map and stratigraphic section of the study area Özçelik (2000).

high organic matter content (wt % TOC > 1) and suitable type of kerogen for hydrocarbon generation (Type II/III mixture) within the basin, especially in Bozbel formation (İlleez et al., 1990; Korkmaz, 1990; Altunsoy and Özçelik, 1995; Özçelik and Altunsoy, 1996; Özçelik, 2000; Yalçın Erik et al., 2015). For this reason, it was aimed to determine the paleo depositional environment characteristics of the Eocene units related to hydrocarbon exploration by using detailed organic geochemical evaluations around Zara-Bolucan region.

2. Geological setting

The development on the northern margin of the Sivas basin started with the ophiolitic complex of the Upper Paleocene-Middle Eocene units and it was stated that these include fan-delta conglomerates, volcanics, nummulite-bearing limestones and marls. The Eocene aged rocks of Sivas Basin are represented by thick olistostromes, turbidites and volcanoclastics (Artan and Sestini, 1971; Kurtman, 1973; Gökten, 1993; İnan, 1993; Guezou et al., 1996; Poisson et al., 1996; Özçelik, 2000). Oligocene units overlying the Eocene sediments consist of thick gypsum, siltstones and sandstones (Temiz, 1996). A widespread marine sedimentation process (except the western part of the Sivas-Tecer Mountain line) was observed in the basin at the beginning of Upper Oligocene (?) - Lower Miocene (Dizer, 1962; Kurtman, 1973; Poisson et al., 1996; Temiz, 1996). The Lower Miocene shallow sea sediments represented by reefal limestones and marls pass upward to

the Middle Miocene red clastics and unconformably over the middle and eastern parts of the basin (Temiz, 1996). The last sedimentary process of the basin and the elevation of the region are represented by Middle Miocene terrestrial-lagoonal sediments. The Pliocene aged conglomerate and sandstone strata were unconformably overlain by Miocene gypsiferous series. Also, quaternary alluviums and travertines are found in some parts of the basin (Kurtman, 1973) (Fig. 1b). The general stratigraphic section of the basin is given in Fig. 1c. The Kozluca formation, which is one of the Eocene aged units, has been formed by alternation of fine shales, marls, sandstones and clayey limestones, and it is İpresian aged. The formation is overlain by tuffite layers belonging to Bozbel Formation. Lithological features and fossil content indicate that this formation has been deposited in the deep sea conditions (Kurtman, 1973). The Bozbel formation, composed of marl, sandstone, shale, limestone and marly limestone alternations, indicates the marine sedimentary environment and Ypresian age.

3. Material and methods

Thirty-five organic rich shale, mudstone and carbonaceous shale outcrop samples of Eocene units were collected from around Zara-Bolucan region. A systematic and detailed sampling guideline was carried out based on lithological change in vertical and lateral of the Eocene sediments on two measured columnar sections in the study area. Some shale and carbona-

Table 1. Some n-alkane and isoprenoid ratios and calculated parameters for the studied samples.

Determined Parameters	ZB-1	ZB-2	ZB-3	ZB-4	ZB-5
Pr/Ph ratio	0.59	0.52	0.54	0.63	0.82
Pr/nC ₁₇	0.41	0.36	0.45	0.46	0.38
Ph/nC ₁₈	0.43	0.35	0.44	0.45	0.36
CPI ₂₅₋₃₃	1.07	1.07	1.09	1.02	1.14
OEP ₂₅₋₃₃	1.07	1.13	1.08	1.03	1.14
nC ₁₉ /nC ₃₁	0.95	1.06	0.85	0.14	1.01
nC ₂₄ +/nC ₂₄ -	1.37	1.64	1.39	4.05	1.33
TAR	1.75	2.44	1.86	9.02	1.28
Extract (mg/kg rock)	29	112	68	141	76

ceous shale samples (> 0.5 TOC, wt. %) were applied with solvent extraction liquid chromatography gas chromatography (GC) and gas chromatography-mass spectrometry (GC-MS) analyses were done on bulk extracts obtained from five samples. The aliphatic components obtained by chromatographic fractionation were done according to ASTM (D 5307-97, 2002) by Agilent 6850 instrument. Sterane and terpane ratios were compute by integration of peak highs from the m/z 217, m/z 191 mass fragmentograms, n-alkanes and acyclic isoprenoids were describe from fragmentation ion m/z 85 and the biomarker ratios were calculated by measuring peak heights in the fragmentograms. Biomarker analyses were done in the research laboratories of the Turkish Petroleum Corporation (TP Research Group, Ankara).

4. Results and discussions

4.1 Molecular geochemistry of organic matter

In this context, the quantity of extractable organic matter (EOM), saturated, aromatic fractions and hetero atomic (NSO) compound ratios of the samples have been determined. The extractable organic matter ratios of the investigated samples range from 29 to 141 mg/kg rock (Table 1). The aliphatic fractions and hetero atomic components are varies between 36% to 81% and 12% to 52%, respectively, while aromatic fractions range from 6% to 27%.

4.1.1 N-Alkanes, acyclic isoprenoids

The gas chromatography (GC) analysis has been used to determine paleo-environmental conditions of Eocene Bozbel and Kozluca Formation based on the distribution of n-alkanes and acyclic isoprenoids. These compound properties are very similar to each other in the investigated samples. n-alkane distribution of the studied samples comprise mainly from n-C₁₂ to n-C₃₅. Table 1 summarizes the results and some calculated parameters of isoprenoids and n-alkanes. Odd-even carbon number predominance (OEP) values and carbon preference index (CPI) values of the Eocene samples ranging from 1.03 to 1.14 and 1.02 to 1.14, respectively.

The long chain n-alkanes (n-C₂₅-n-C₃₅) in the compositions of the studied samples are more in proportion to

the mid-chain (C₂₁-C₂₅) n-alkanes. In this composition, C₂₇, C₂₉ and C₃₁ being the most important components. Also, the terrigenous/aquatic ratio (TAR) is the ratio between the concentration of long-chain n-alkanes mostly derived from terrestrial plants (> n-C₂₇) to short chain n-alkanes of algal origin (< n-C₁₅), and it is used to evaluate the importance of terrigenous versus aquatic inputs. The calculated n-C₁₉/n-C₃₁ ratios vary between 0.85 and 1.06, while, the n-C₂₄+/n-C₂₄- ratios are high (> 1) and TAR is more than 1 and range between 1.28 to 9.02. This wide range of values suggests various source organic matter inputs, from terrestrial to marine. This results support by predominance of C₂₇, C₂₉ and C₃₁ n-alkanes (Peters and Moldowan, 1993; Peters et al., 2005).

Furthermore, the determined samples have acyclic isoprenoids (pristane and phytane). Phytane (Ph) is higher than pristane (Pr), therefore the Pr/Ph ratios range from 0.52 to 0.82 (Table 1). Additionally, acyclic isoprenoid ratios were calculated as Pr/n-C₁₇ and Ph/n-C₁₈ ratios compared to n-alkanes and values range between 0.36 to 0.46 and 0.35-0.45, respectively (Table 1) and indicate to anaerobic environment conditions (Tissot and Welte, 1984; Peters et al., 2005). In this respect, all of the analyzed samples have Pr/Ph ratios of <1 and usually close to 1 CPI values (1.02-1.14) define algal/bacterial organic matter, which is typical of marine depositional conditions that indicate sedimentation under reducing conditions (Tissot and Welte, 1984).

4.1.2 Terpanes and steranes

The detailed biomarker properties and some calculated ratios as a result of organic geochemical evaluations are given in Table 2. C₂₄ tetracycliterpane, C₂₃ tricyclic terpanes, C₂₉ hopane, C₃₀ hopan, C₂₇ sterane, C₂₉ sterane are considerable components (Fig. 2a, b).

Especially, the terpanes are prominent saturated cyclic triterpenoid components of the Eocene sediments in the investigated area. The pentacyclic triterpanes and the homohopanes (Fig. 2a). In this study hopane (C₃₀17 α (H)-hopane), norhopane (C₂₉17 α (H)-hopane), C₃₁17 α (H)-hopane, moretane (C₃₀), normoretane (C₂₉), 18 α (H)-trisnorhopane (T_s), and 17 α (H)-trisnorhopane (T_m) components were determined and assess-

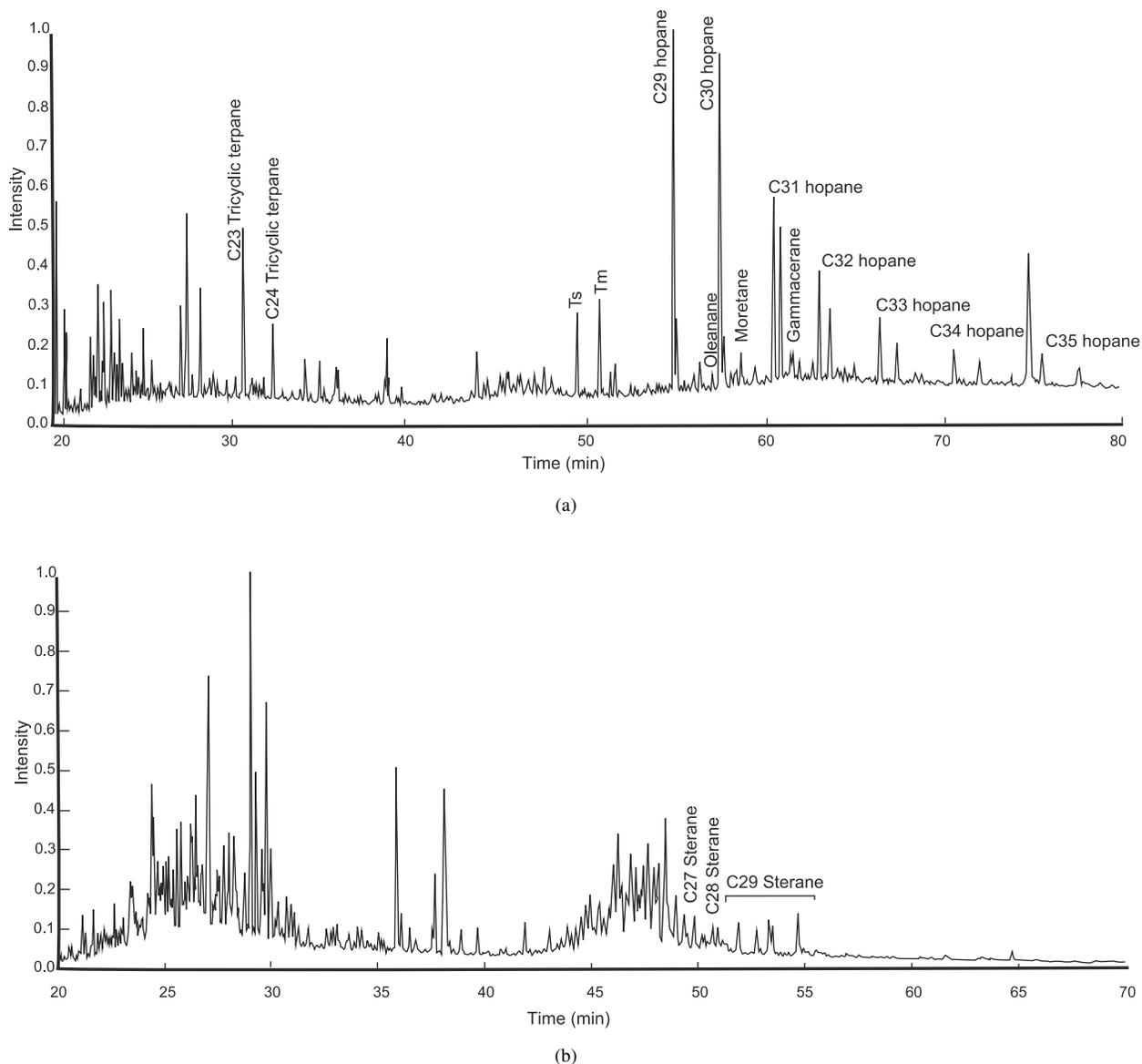


Fig. 2. a) m/z 191 and b) m/z 217 chromatograms of the investigated samples (ZB-2).

ments related to the paleo-environmental interpretation were made.

C_{23} tricyclic terpanes were determined in the investigated samples and (Fig. 2a) have relatively high C_{23} tricyclic terpane ratios (1.39-2.76) (Table 2). Also, the ratio of C_{24} tetracyclic terpane relative to C_{23} triterpanes vary according to organofacies characteristics, with comparatively high ratios in source rocks with terrestrial organic matter input (Peters and Moldowan, 1993). In the terpane distribution (m/z 191), the dominantly algal-bacterial organic matter contribution for the extracts can be suggested from the C_{19} - C_{25} tricyclic terpanes distribution by the dominance of C_{23} tricyclic terpane relative over the C_{24} tricyclic terpane greater than C_{24} tricyclic terpane (> 1). However, C_{26}/C_{25} tricyclic terpane ratios generally low (0.29-0.55).

In the pentacyclic terpanes, the high C_{24} tetracyclic/ C_{26} tricyclics ratio, the moderate-high norhopane/hopane ratio, and

the moderate to high C_{35}/C_{34} hopane ratio (Table 2) indicate marine carbonate facies and anoxic depositional condition for the Bozbel and Kozluca Formation samples.

The $T_s/(T_s+T_m)$ ratio is range between 0.44 to 0.49, with a mean value of 0.44 and indicating that T_m is the dominant component of investigated samples. The T_m/T_s ratio change between 1.14 and 1.41, in the meanwhile the $C_{32}S/(S+R)$ ratio is varies between 0.51 and 0.56. While, the diasterane index is ranging from 0.36 to 0.91, the tricyclic/hopane ratio is low, like the C_{24} tetracyclic terpane/ C_{26} tricyclic terpane and the C_{26}/C_{25} tricyclic terpane ratios (Table 2).

The relative abundances of the C_{27} -cholestane, C_{28} ergostane and C_{29} stigmastane have been evaluated from m/z 217 chromatograms (Fig. 2b). The stigmastane is more common in most of the samples than the other regular sterane components, with a contribution of 31.7%-41.1%, followed by cholestane (25.8%-45.2%), and ergostane (22.7%-33.9%).

Table 2. m/z 191 and 217 parameters of GC-MS data and some determined parameters for the investigated samples.

Determined Parameters	Sample Numbers				
	ZB-1	ZB-2	ZB-3	ZB-4	ZB-5
C ₂₃ Tri./C ₂₄ Tri.	2.76	2.69	2.71	2.19	1.39
Norhopan/Hopan	0.97	0.95	0.93	0.92	0.90
C ₃₁ R/C ₃₀	0.45	0.43	0.41	0.43	0.45
C ₂₆ Tri./C ₂₅ Tri.	0.29	0.49	0.41	0.53	0.55
C ₂₃ Tri./C ₃₀ Hopan	0.95	0.54	0.29	0.44	0.55
C ₂₄ Tet./C ₂₆ Tri.	5.59	2.21	3.37	3.28	2.24
Tri/Hop	0.45	0.06	0.18	0.25	0.37
C ₃₅ /C ₃₄ Homohopane	1.63	-	1.27	2.18	-
T _m /T _s	1.14	1.21	1.36	1.31	1.41
T _s /T _s + T _m	0.49	0.48	0.45	0.46	0.44
C ₃₂ 22S/(22S+22R) hopane	0.56	0.54	0.54	0.51	0.56
C ₂₉ ββ/(ββ + αα)	0.63	0.65	0.62	0.58	0.59
C ₂₇ /C ₂₉	0.93	0.79	0.73	0.65	0.89
C ₂₈ /C ₂₉	0.73	0.67	0.71	0.81	0.73
C ₂₇ /C ₂₇ +C ₂₉	0.45	0.41	0.40	0.39	0.47
Dia/Reg (Diasterane Index)	0.91	0.54	0.46	0.36	0.58

The compositional ratios of regular steranes ($\alpha\alpha\alpha$ 20R) in the examined samples is $C_{29} > C_{27} > C_{28}$. The predominance of the stigmastane component concerns an intense terrestrial organic matter additive, whereas a dominance of cholestane and ergostane indicate a dominance of marine phytoplanktons and lacustrine algae, respectively (Peters and Moldowan, 1993) (Fig. 3).

4.2 Paleodepositional conditions

Terpanes and steranes are the most important compounds that can give very specific information about the source and deposition environment of investigated organic matter rich levels (Peters et al., 2005). Biomarker ratios and indices of triterpanes and steranes of investigated samples are shown in Tables 1 and 2.

The C₃₅/C₃₄ homohopane and norhopane/C₃₀ hopanes (C₂₉H/C₃₀H) values can be used to identify paleodepositional facies characteristics (Peters et al., 2005) and the moderate to high C₃₅/C₃₄ homohopane ratios indicate the anoxic conditions of the paleo depositional environment of Eocene sediments. The diasteranes/regular steranes ratio (diasterane index) for the analyzed samples show generally low (0.36-0.91) (Table 2) and they indicate carbonate rocks and anoxic depositional environment. The same conclusion can be reached by the dominance of 17 α (H)-22, 29, 30-trisnorhopane (T_m) over 18 α (H)-22, 29, 30-trisnorhopane (T_s), as well as the high values of C₂₄ tetracyclic/C₂₆ tricyclic terpane ratios (Tissot and Welte, 1984; Peters et al., 2005). Especially, C₂₄/C₂₆ terpane ratios are related with a mixed organic matter of algal, bacterial and abundant terrestrial sourced. Also, the C₃₁R homohopane/C₃₀ hopane ratios (> 0.25) indicate that the Eocene sediments were deposited in a marine depositional

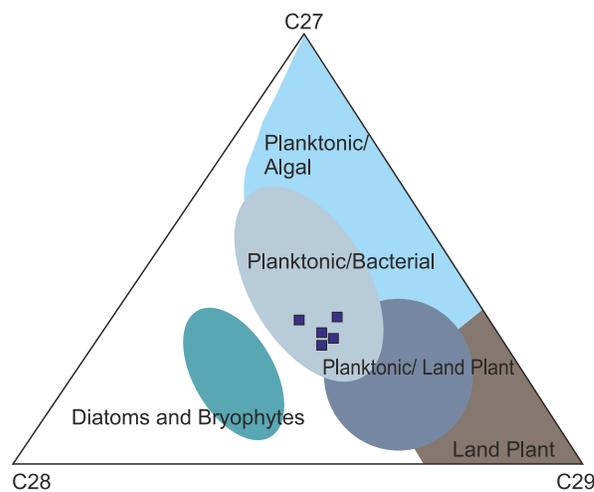


Fig. 3. Sterane ternary diagram of C₂₇%, C₂₈% and C₂₉% ($\alpha\alpha\alpha$ R) concerning the selected samples in Bozbel and Kozluca Formations.

environment (Fig. 4a and 4b). The C₂₇, C₂₈, and C₂₉ regular sterane distributions imply that there are greater contributions from terrestrial higher plants than marine algae in the Bozbel and Kozluca Formations samples.

The ratios of the Pr/Ph for the Eocene sediment extracts were generally low and ranged between 0.52 and 0.82 (Table 1) indicating anoxic, reduced marine carbonate depositional environment (Tissot and Welte, 1984; Peters et al., 2005). Also, the Pr/n-C₁₇ ratios ranged between 0.36 and 0.46 (< 0.5) indicating marine depositional environment. The cross plot of Pr/n-C₁₇ versus Ph/n-C₁₈ for the whole analyzed samples (Fig. 5a) shows mixed (marine and terrestrial) source organic matters deposited under reduced condition with a

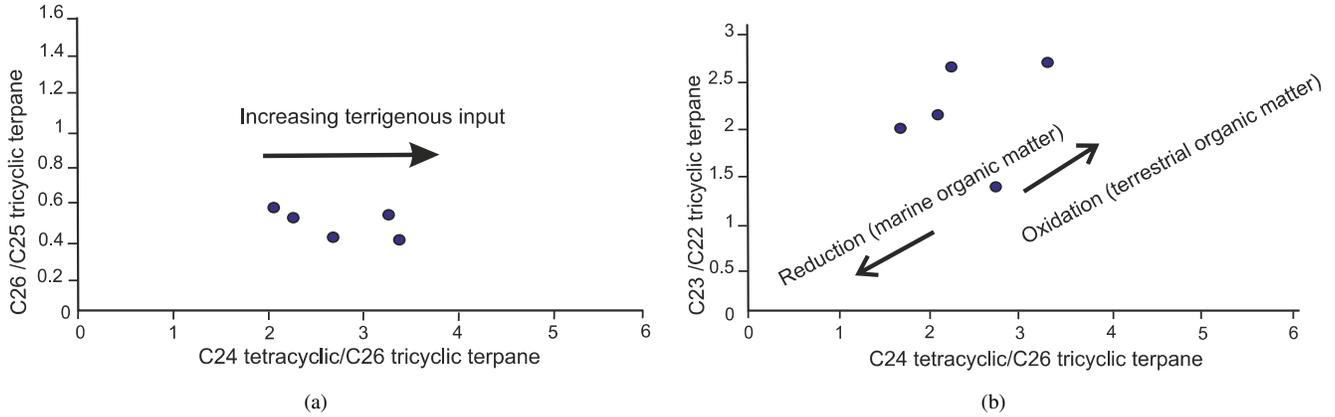


Fig. 4. a) Relationships between C_{26}/C_{25} tricyclic terpane vs C_{24} tetracyclic/ C_{26} tricyclic terpane ratios, b) C_{23}/C_{22} tricyclic terpane vs C_{24} tetracyclic/ C_{26} tricyclic terpane ratios for Eocene sediments.

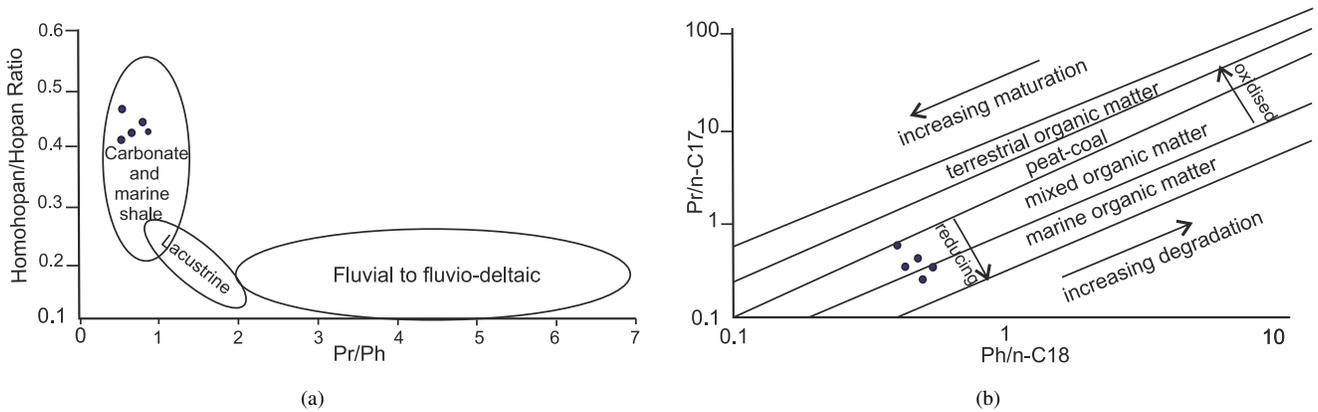


Fig. 5. Result of GC and GC-MS analysis data on diagrams; a) Homohopane/hopane ratio vs Pr/Ph ratio, b) Pr/nC_{17} vs Ph/nC_{18} ratio for Eocene sediments.

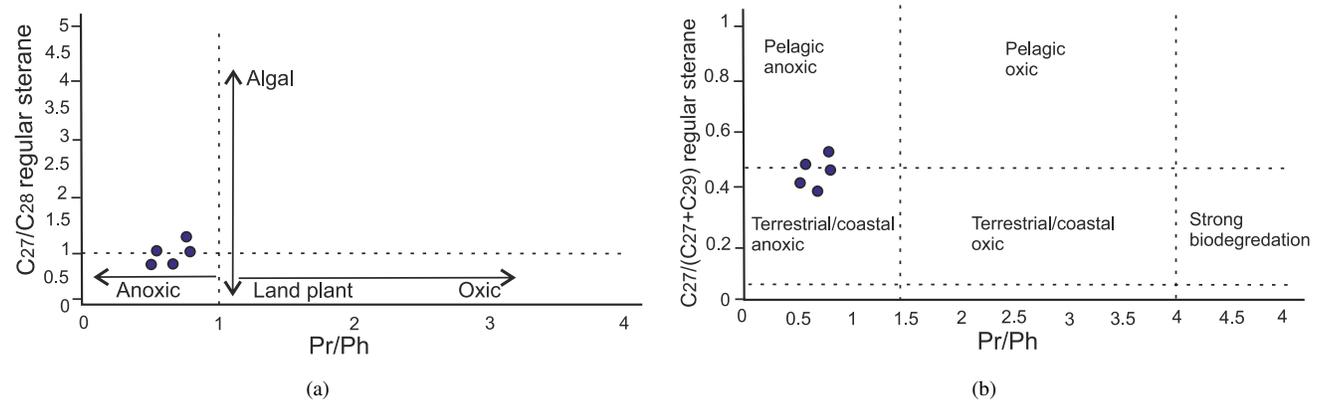


Fig. 6. Relationship between a) C_{27}/C_{28} regular sterane vs Pr/Ph ratios and b) $C_{27}/(C_{27}+C_{27}+C_{29})$ regular sterane vs Pr/Ph ratios for investigated samples.

less effect of biodegradation. Additionally, the Pr/Ph versus $C_{31}R$ homohopane/ C_{30} hopane diagram and C_{26}/C_{25} tricyclic terpanes ratios (< 0.9) indicate a carbonate and marine shale (Fig. 5b) (Peters et al., 2005). This result supports the data is provided by the $C_{27}/C_{29\alpha\alpha\alpha}$ (20R) sterane ratio which rises from 0.65 to 0.93 and the $C_{27}/C_{27+C_{29}}$ sterane ratio was 0.39-0.47.

The majority of the analyzed samples show relatively high concentrations of C_{29} peaks with high norhopane/ C_{30} hopane ratios ranging from 0.90 to 0.97 (> 0.6) and C_{35}/C_{34} values (> 0.8) indicate to a marine environment (Peters and Moldowan, 1993; Peters et al., 2005) with organic rich carbonates and evaporites (Peters et al., 2005). The extracts of the Eocene samples are characterized by the presence of a relatively low to moderate abundance (0.36-0.91) of the rearranged steranes (Table 2). This is a characteristic feature of an anoxic, clay poor, carbonate, marine depositional environment. Also, the identification of C_{30} steranes in all samples supports their derivation from marine source rocks (Peters and Moldowan, 1993) (Table 2). There is a positive correlation between Pr/(Pr + Ph) and C_{27} diasterane index that were controlled by the depositional environments. Plots of regular steranes $C_{27}/(C_{27} + C_{29})$ versus Pr/Ph ratio, and C_{27}/C_{29} versus Pr/Ph ratio provide important knowledge of organic matter rich rocks based on paleo depositional characteristics (Moldowan et al., 1991). According to these diagrams suggests a major contribution of terrigenous organic matter and a secondary input of aquatic microorganisms (Fig. 6a and 6b).

5. Conclusions

The Eocene shale, mudstone and carbonaceous shale samples of Bozbel and Kozluca Formations are exposed around Zara region and were analyzed for interpret the paleo depositional environment characteristics. Some of the remarkable features of the biomarker evaluations of the examined samples are; dominance of T_m over T_s and the ratios of diasterane/regular steranes, C_{24} tetracyclic terpane/ C_{23} tricyclic terpane, C_{24} tetracyclic terpane/ C_{26} tricyclic terpane and C_{29}/C_{30} hopane. The detail biomarker parameters such as Pr/Ph, $n-C_{19}/n-C_{31}$, $n-C_{24}+/n-C_{24}-$ ratios, OEP, CPI, T_m/T_s ratio and high ratio of C_{29} regular steranes indicate that the organic matter is mostly sourced from terrestrial with a minor contribution from marine-influenced origin. Generally, the paleo-redox features of depositional environment are supported by the T_m/T_s and Pr/ $n-C_{17}$ versus Ph/ $n-C_{18}$ diagram. The marine depositional environment conditions was supported by C_{26}/C_{25} tricyclic terpanes, C_{23} tricyclic/ C_{30} hopane ratios and $C_{31}R$ homohopane/ C_{30} hopane ratios. Based on biomarker properties, it was interpreted that Eocene Kozluca and Bozbel formation were deposited in a marine environment and contained mixed aquatic and terrestrial organic matter. Also, there is no significant difference between the shales in the Bolucan and Kozluca formations for the analyzed samples.

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