

Organic Geochemistry of Hydrothermal Petroleum in the Menderes-Gediz Graben System, Denizli-Saraykoy, Western Turkey: Preliminary Results

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ABSTRACT

Western Turkey is situated in the geologic province of Aegean region. It is seismically one of the most active regions of the world and includes prospective areas for hydrocarbons, coal beds, and geothermal sources. The KB-5 well, from which the petroleum sample was taken, has been located at the intersection of the geothermally active Menderes and Gediz grabens near the town of Saraykoy. During drilling, the petroleum erupted onto the surface with its associated thermal water (120°C) from a depth of 120-132 m (e.g., from Early to Middle Pliocene aged claystone and marl). Drilling was stopped at a depth of 253 m when it reached to reservoir limestones of the Sazak Formation. The purpose of this paper is twofold: (1) to understand the generation mechanism of the petroleum, either by geothermal-gradual or hydrothermal-rapid processes, and (2) to assess the nature of source from which it was derived. In order to achieve these purposes, the sample was analyzed by organic geochemistry using thin layer chromatographic separation followed by gas chromatography-mass spectrometry (GC-MS).

The KB-5 petroleum is composed primarily of polar NSO compounds+asphaltenes (77%), aromatic hydrocarbons (19.22%), and minor saturated hydrocarbons (3.78%). This feature is similar to hydrothermal petroleum of Calcite Springs from Yellowstone National Park, Wyoming, U.S.A. TIC traces of the KB-5 sample shows n-alkanes ranging from $n\text{-C}_{20}$ to $n\text{-C}_{38}$ but depleted $< n\text{-C}_{20}$ and maximum at $n\text{-C}_{30}$. An absence of acyclic isoprenoids (pristane, phytane) and considerably large UCM (unresolved complex mixture) point out that this oil has been subject to moderate degree of alteration possibly by hot geothermal waters. The parameters obtained from sterane and terpane biomarkers present in the oil (low $\text{C}_{29}\text{NH/C}_{30}\text{H}$ ratio; presence of gammacerane, high $\text{C}_{35}/\text{C}_{34}$ homohopane ratio; $18\alpha(\text{H})$ -oleanane, $n\text{-C}_{28}< n\text{-C}_{27}< n\text{-C}_{29}$, $\text{C}_{28}< \text{C}_{27}< \text{C}_{29}$ steranes, $\% \text{C}_{29} \alpha\alpha\text{R}> \% \text{C}_{27} \alpha\alpha\text{R}$) suggest that the petroleum sample was generated from a Tertiary source rock with a clay-rich lithology, terrestrial-rich organic matter deposited in relatively saline and anoxic environmental conditions. Maturity sensitive biomarker parameters ($\text{Tm}>\text{Ts}$, $\text{C}_{29} 20\text{S}/20\text{S}+20\text{R}=0.35$, $\text{C}_{32} 22\text{S}/22\text{S}+22\text{R}=0.57$, low diasterane/regular sterane ratio) correspond to a maturity level of about 0.50-0.60 %Ro. This level of maturation could not be reached by such a young and shallow source rock and/or oil in the absence of "instantaneous" hydrothermal activity

1. INTRODUCTION

Western Turkey is situated in the geologic province of Aegean region (Fig. 1a). It is seismically one of the most active regions of the world and includes prospective areas for hydrocarbons, coal beds, and geothermal sources. Although large amounts of geothermal resources have been already discovered (Şimşek and Yılmazer, 1977; Şimşek, 1985; Şimşek and Güleç, 1994; Şimşek, 2002), however, petroleum potential of the region appears to be limited regardless of the existing and promising structures. National oil company of the Turkey, TPAO, has drilled a few wells to explore any possible potential for commercial quantities of hydrocarbons in the area. One of the wells (Alaşehir-1; Fig.1) drilled in the Gediz Graben encountered petroleum at about 1991 m but it was in non-commercial quantities. The other well, (Nazilli-1; Figs.1b and 2) drilled again by TPAO was in the Büyük Menderes Graben where, no petroleum was encountered. Recently, the well drilled (KB-5 well; Figs. 1b and 2) by a private geothermal water supply company, in the Büyük Menderes Graben, approx. 30 km away from the Nazilli-1 well (Fig.1) erupted with petroleum like material from 120-132 m interval with accompanying hydrothermal solution at 120°C. Close association of hot geothermal fluids and petroleum in the KB-5 well made us to suspect that this petroleum might have been hydrothermally generated.

"The geothermal-gradual (conventional) petroleum is natural products of basin evaluation and are generated coterminously with sediment compaction and heating. In contrast, petroleum generated in high temperature and with high-fluid flow regimes are defined here as hydrothermal in origin because the agent of thermal alteration and mass transfer, hot circulating water, is responsible for petroleum generation and migration from the source rocks or unconsolidated sediments. Generation of hydrothermal oils and gases are relatively rapid, whereas geothermal oils are generated at a rate that is tied to basin subsidence" (Didyk and Simoneit, 1990).

The purpose of this paper is twofold: (1) to understand the generation mechanism of the petroleum collected from the KB-5 well, either by geothermal-gradual or hydrothermal-rapid processes, and (2) to assess the nature of source from which it was derived. In order to achieve these purposes, petroleum like material from the KB-5 well, was compared to the petroleum from the nearby Alaşehir-2 well in the Gediz Graben. Both, the KB-5 and Alaşehir-2 petroleum samples were analyzed by organic geochemistry using thin layer chromatographic separation followed by gas chromatography-mass spectrometry (GC-MS). The petroleum sample, which has been described to be

generated hydrothermally (Kvenvolden and Simoneit, 1990) is also used in this study for comparison purposes.

2. GEOLOGICAL SETTINGS

2.1 Tectonic Settings

Extensive geological investigations have been performed for understanding of western Anatolian tectonics (Dewey and Şengör, 1979; Şengör and Yılmaz, 1981), core complexes (Işık et al., 2004), and graben systems (Bozkurt and Sözbilir, 2004; Seyitoglu and Scott, 1992). Therefore, a very brief summary of these investigations are given here.

Tectonic activities in the Anatolian plate are controlled by four major structural zones: Bitlis Suture Zone, North Anatolian Fault Zone (NAFZ), East Anatolian Fault Zone (EAFZ), and western Anatolian Graben systems (Fig. 1a). Bitlis Suture Zone is the appearance of the collision between the Arabian and the Anatolian plates during about the middle Miocene. This activity is still in progress. NAF and EAFZ are strike-slip faults developed as a result of northward movement of the Arabian plate. The extension in western Anatolia (i.e., the study area) is closely related to the northward movement on the Arabian plate in the east, which pushed Anatolia westwards through the NAFZ and EAFZ (Dewey and Şengör, 1979). This creates E-W compression in Western Anatolia, which is relieved by N-S extension. Seyitoglu and Scott (1992) reported that extension in western Anatolia commenced most likely during the early Miocene.

The basement rocks in the vast areas of Western Anatolia are the Menderes metamorphics of Paleozoic or older period. This region has been experiencing N-S directed extension since, at least, latest Oligocene-Early Miocene and is currently under the influence of forces exerted by subduction of the African Plate beneath the southern margin of Anatolian Plate along the Aegean-Cyprean subduction zone and the dextral slip on the North Anatolian Fault system. E-W trending grabens (e.g. Gediz and Büyük Menderes grabens) are the most prominent features of western Turkey and they cut up the Menderes Massif into northern, central and southern submassifs, respectively (Bozkurt and Sözbilir, 2004; Fig.1).

2.2 Stratigraphy

The stratigraphy of the KB-5 well and surrounding region summarized here are based on the study made by Şimşek (1985). Accordingly, stratigraphic sequence of the studied area consists of Lower Pliocene Kızılburun, Sazak, and Kolonkaya and Plio-Quaternary Tosunlar units. The Kızılburun Formation, is the earliest continental-lacustrine deposits that cover the massif. It consists of alternating conglomerate, sandstone, mudstone and claystone. The Sazak Formation conformably overlies the Kızılburun Formation. Clayey and sandy limestones are the dominant lithology of this formation in addition to sandstones, marn and claystones. This formation is the primary reservoir for the geothermal water. Erupted oil like material in the KB-5 well is from the Sazak Formation. Kolonkaya Formation conformably overlies the Sazak Formation and is made up of sandstone, claystone and clayey limestones. Thickness of this unit reaches up to 500 m. Plio-Quaternary Tosunlar Formation overlies the Lower Pliocene units with an angular unconformity. Tosunlar Formation consists of alternating conglomerate, sandstone and mudstone with fossiliferous clayey limestones. Thickness is about 500 m. Terrace deposits, alluvium, slope debris, alluvial fans and travertine characterize the Quaternary sequence. In the study area, Tosunlar formation is not present due to recent

intense tectonic and subsequent erosion. Stratigraphic positions of Kolonkaya and Sazak formation are not regular and continuous due to again intense tectonic activity and resulting normal faults. Meteoric waters circulate through the deep-seated and interrelated these normal fault planes and fractures of the recent Büyük Menderes (Fig.3) and Gediz graben (Fig.4). systems. In the area of this study, the main reservoir unit is the calcareous Sazak Formation.

3. RESULTS AND DISCUSSION

Normalized percentage compositions of the fractions from Iatrosca analysis of the KB-5 petroleum and various samples from different part of the world are shown in Fig.5. Capillary gas chromatograms, m/z 191 GC-MS chromatograms, and m/z 217, m/z 218 GC-MS chromatograms of the KB-5 and Alaşehir-1 oils are given in Figs. 6, 7, and 8, respectively. For comparison purposes, ratios related to n-alkanes (Figs.6a and b), terpanes (Figs.7a and b) and steranes (Figs.8a, b and c, d) for the KB-5 oil with unknown origin and conventional Alaşehir-1 and hydrothermal Guaymas Basin oils are listed in Table 1.

3.1 Bulk Composition

Normalized percentage composition of the KB-5 petroleum plotted in a ternary diagram indicates that this petroleum is enriched in asphaltic (NSO) compounds (Fig.5). This feature of the KB-5 petroleum is very similar to hydrothermally generated Calcite Springs petroleum sample from Yellowstone Park, Wyoming USA (Clifton et al., 1990) and the other hydrothermal petroleum-bearing samples from Guaymas Basin, and Escanoba Through-USA (Kawka and Simoneit, 1987; Kvenvolden and Simoneit, 1990), Wakamiko Caldera-Japon (Yamanaka et. al., 2000), and Waitotapu- New Zealand (Czochanska et al., 1986). and geothermal resources (Şimşek, 1985; Şimşek and Güleç, 1994; Şimşek, 2003). Lower abundance of saturated hydrocarbons in the KB-5 petroleum is attributed to removal of these type of hydrocarbons by bacteria preferentially and also to water washing in hot water and evaporation in the surface.

The scattered plot of the all samples on the ternary diagram (Fig.5) refers that hydrothermal petroleum does not show fixed composition with respect to bulk fractions. In fact, Simoneit (1985) suggested that the wide variations in bulk composition of hydrothermal petroleum are mainly due to the great variability inherent in the hydrothermal processes and post-generation effects.

3.2 Normal Alkanes and Isoprenoids

The KB-5 petroleum sample from the Büyük Menderes Graben, whose generation mechanism is subject to this investigation, is compared to the geothermally (conventionally) generated petroleum sample from the Alaşehir-1 well in the Gediz Graben using distributions of n-alkanes and isoprenoid hydrocarbons.

The ratios related to these distributions are given in Table 1. The reason for selecting Alasehir-1 sample for the comparison purposes is that (1) Both samples are taken from the graben systems of the western Anatolia and (2) Alaşehir-1 petroleum is produced from 1991 m in depth and it is most likely generated via conventional way. Remarkable differences between the two petroleum samples exist in the nC_{13} - nC_{30} region of the gas chromatograms (Fig.6). Alaşehir-1 sample contain a homologous series of n-alkanes continuously from nC_{13} to about nC_{34} , whereas the KB-5 petroleum do not contain continuous series of n-alkanes by saying that it is lack of nC_{19} compounds. At a

first sign, the lack of nC_{19} compounds may refer bacterial alteration, which has been known to occur with the presence of aerobic bacterial activity and at a temperature of less than 80°C. As mentioned, the KB-5 petroleum came from a relatively shallow depth of 120-130 m, where the temperature was quite high and reached about 120°C. Such a high temperature does not let bacteria to survive and consume n -alkanes, on the other hand, it increases the solubility of hydrocarbons. We propose that lack of nC_{19} compounds in the KB-5 petroleum is caused by hydrothermal process and accompanying bacterial activity.

It has been well known that bacteria preferentially consume n -alkanes and then attack to acyclic isoprenoids to consume. However, n -alkanes and isoprenoids in the KB-5 petroleum (Fig. 6a) do not follow bacterial attack trend such that, the absence and/or very low concentrations of acyclic isoprenoids (i.e., Pr and Ph) and presence of nC_{19+} compounds.

Normal alkanes and acyclic isoprenoids carry information about the type and maturity level of organic matter in the source rocks, from which petroleum is generated. OEP (odd-even predominance) is a n -alkane related parameter that carries information about both maturity and type of organic matter. The distribution of n -alkanes in the nC_{18} and nC_{22} region in the Alaşehir-1 sample show slight odd-carbon predominance indicating terrigenous organic matter input because land-plant waxes, the likely precursors for such n -alkanes, are characterized by high OEP values (OEP20=1.07; Table 1; in the OEP20, the 20 indicates the center of the range of carbon numbers used in the calculations). OEP27 values for the KB-5 and Alaşehir samples, on the other hand, are 1.02 and 0.98, respectively. Both values are very close to unity referring that both samples are mature petroleums. OEP=1 is commonly observed in mature petroleums. Acyclic isoprenoid ratios, Pr/Ph, Pr/ nC_{18} , and Pr/ nC_{17} are frequently used for understanding of paleoenvironment of source rocks. In the case of our sample, the KB-5 petroleum, acyclic isoprenoid ratios would not be determined due to the result of process rather than source. For the Alaşehir-1 sample, these ratios are calculated and used paleoenvironmental indicator. For example, Pr/Ph>1 (1.20), Pr/ nC_{18} <1 (0.98), and Pr/ nC_{17} >1 (1.03) for the Alaşehir-1 sample indicate that source rock of this sample was deposited in an oxic environment and received some terrigenous organic input. High Pr/Ph ratio, which refers an oxic condition, is not consistent with the presence of β -carotene compound determined in the Alaşehir-1 sample. β -carotene is only present in petroleum, whose associated source rock could have been deposited in an anoxic condition.

The UCM (unresolved complex mixture) of saturated hydrocarbons for the KB-5 sample is much more larger than the that of Alaşehir-1 sample indicating that the KB-5 petroleum has been subjected to more of alteration (i.e., water washing, evaporation etc.) than the Alaşehir-1 sample. Larger UCM for the KB-5 sample is consistent with the absence of nC_{19} compounds.

3.3 Biomarkers

Biomarkers in petroleum have significant application areas in hydrocarbon exploration programs, such as predicting the nature and origin of source rocks (i.e., approximate age, lithology, type of organic matter, water conditions, and maturity). In order to better understand the nature and origin of the KB-5 and Alaşehir-1 petroleums analyzed in this study and the Guaymas Basin petroleum sample (the data taken from Kvenvolden and Simoneit, 1990) are

compared with each other based on the specific source and maturity biomarker ratios (Table 1). Comparisons are also made using terpane (m/z 191) and sterane (m/z 217 and 218) fingerprints of the KB-5 and Alaşehir-1 petroleum samples. As seen in Fig. 7, the dominant terpanes in both samples are hopanes ($17\alpha(H)$, $21\beta(H)$ -hopane series). The $\alpha\beta$ -hopanes ranged from C_{27} to C_{35} as a homologous series with a maximum at C_{30} .

Presence of $18\alpha(H)$ -oleanane in a petroleum refers a Tertiary source rock with terrigenous angiosperm input and low $C_{29}NH/C_{30}H$ (<1) ratios suggest clay-rich (i.e., clastic) source rock for all the three petroleums examined in Table 1. Higher concentration of C_{31} , C_{32} , C_{33} , C_{34} , and C_{35} extended hopanes (Fig. 7a) in the KB-5 petroleum indicates organic matter is also rich in bacteria. Presence of gammacerane and $C_{35}/C_{34}>1$ for the KB-5 petroleum (Fig. 7a) indicates that source rock deposited in an anoxic conditions (Peters and Moldowan, 1993).

Steranes (Figs. 8a and 8b) are also used as an organic matter type indicator. For example, $C_{27}/C_{29}>1$ and $C_{27}/C_{29}<1$ are commonly used by geochemists for an indicator of marine versus terrigenous organic matter, respectively (Huang and Meinschein, 1979). In this respect, the KB-5 (0.74) and Alaşehir-1 (0.33) petroleum samples exhibit terrigenous, whereas Guaymas Basin (2.20) petroleum exhibits marine-algal input. In the case of the Guaymas Basin, marine input was verified with presence of marine diatom algae in the sediments, from which petroleum was generated (Kvenvolden and Simoneit, 1990).

Careful comparison between the maturity related ratios in Table 1 shows a slight difference between the KB-5, Alaşehir-1, and Guaymas Basin petroleum samples. The C_{31} , C_{32} , and C_{33} 22S/22S+22R hopane ratios for the KB-5, Alaşehir-1, and Guaymas Basin petroleum samples are calculated as 0.55, 0.57, 0.55; 0.55, 0.53, 0.62; and 0.53, 0.57, 0.54, respectively (Table 1). The C_{31} , C_{32} , and C_{33} hopanes in petroleum occur as a mixture of two epimers, 22S and 22R, which provides relative maturity level of petroleum. With increasing maturity, the 22R, which is biological precursor, is converted to 22S epimer and the ratio increases from zero to equilibrium ratio of about 22S/22S+22R=0.60. Equilibrium value of 0.60 corresponds vitrinite reflectance (%Ro) value of 0.60 at onset of early petroleum generation. Epimer ratios of the three petroleum in Table 1 is lower than 0.60 (except C_{33} epimer ratio of Alaşehir-1 sample) suggesting that all the petroleums was generated at a maturity level of %Ro<0.60. This conclusion is firmly supported by the C_{29} 20S/20S+20R sterane ratios of the petroleums, which are 0.35, 0.32, and 0.28 for the KB-5, Alaşehir-1, and Guaymas Basin petroleum samples, respectively. The conversion of 20R to 20S occurs similar fashion to 22R to 22S conversion as described above and reaches an equilibrium value when 20S/20S+20R ratio is about 0.55, which corresponds %Ro= 0.80 at peak petroleum generation. Again, the considered three petroleum samples appear to be at an early mature stage with respect to oil generation (%Ro=0.50-0.60). The ratio of $17\beta(H)$ -22,29,30-trisnorhopane (Tm) to $18\alpha(H)$ -22,29,30-trisnorhopane (Ts), which has been used as a maturity indicator, are also high for the three petroleum samples (3.00, 7.75, and 2.90, respectively) indicating low level of maturity (Table 1).

Moretanes, $17\beta(H)$, $21\alpha(H)$ -hopane series are unusually absence or in low concentration in the KB-5 sample but present in high concentrations in the Alaşehir-1 sample

(Figs. 7a and b). Moretanes are sensitive and unstable compounds with respect to temperature, so their absence in the KB-5 sample implies that the sample has been subject to considerably high temperature.

4. CONCLUSIONS

The petroleum from the KB-5 well in the Büyük Menderes Graben, Sarayköy-Denizli, Turkey, was characterized by high asphaltic (NSO) (77%) and low saturated compounds (3.78%). This composition appears to be very similar to those of the hydrothermally derived petroleums from the other part of the world.

From a study of the biomarkers present in the KB-5 petroleum, the source rock was Tertiary in age, rich in clays (clastics), rich in terrigenous plus bacterial organic matter and deposited under anoxic conditions.

A high ratio of Tm/Ts (3.0), low ratio of C_{29} 20S/20S+20R (0.35) sterane, low C_{31} , C_{32} , C_{33} 22S/22S+22R extended hopane ratios (0.55, 0.57, and 0.55, respectively) suggest that the KB-5 petroleum was generated from an early mature source rock at %Ro value of 0.50-0.60. This level of maturation could not be reached by such a young (i.e., Early Pliocene sediments) and shallow (120-132 m) sediments unless an existence of "instantaneous" hydrothermal activity. This is supported by the existence of hot geothermal waters (i.e., 100°C) associated with the KB-5 petroleum.

High temperature petroleums-those associated with magmatically-driven hydrothermal systems are rare in the geologic environment. It appears that KB-5 petroleum from Denizli-Turkey with its erupted feature with the hydrothermal fluids is a unique case and deserves more attention in the near feature to understand mechanism of petroleum generation in the geothermal areas.

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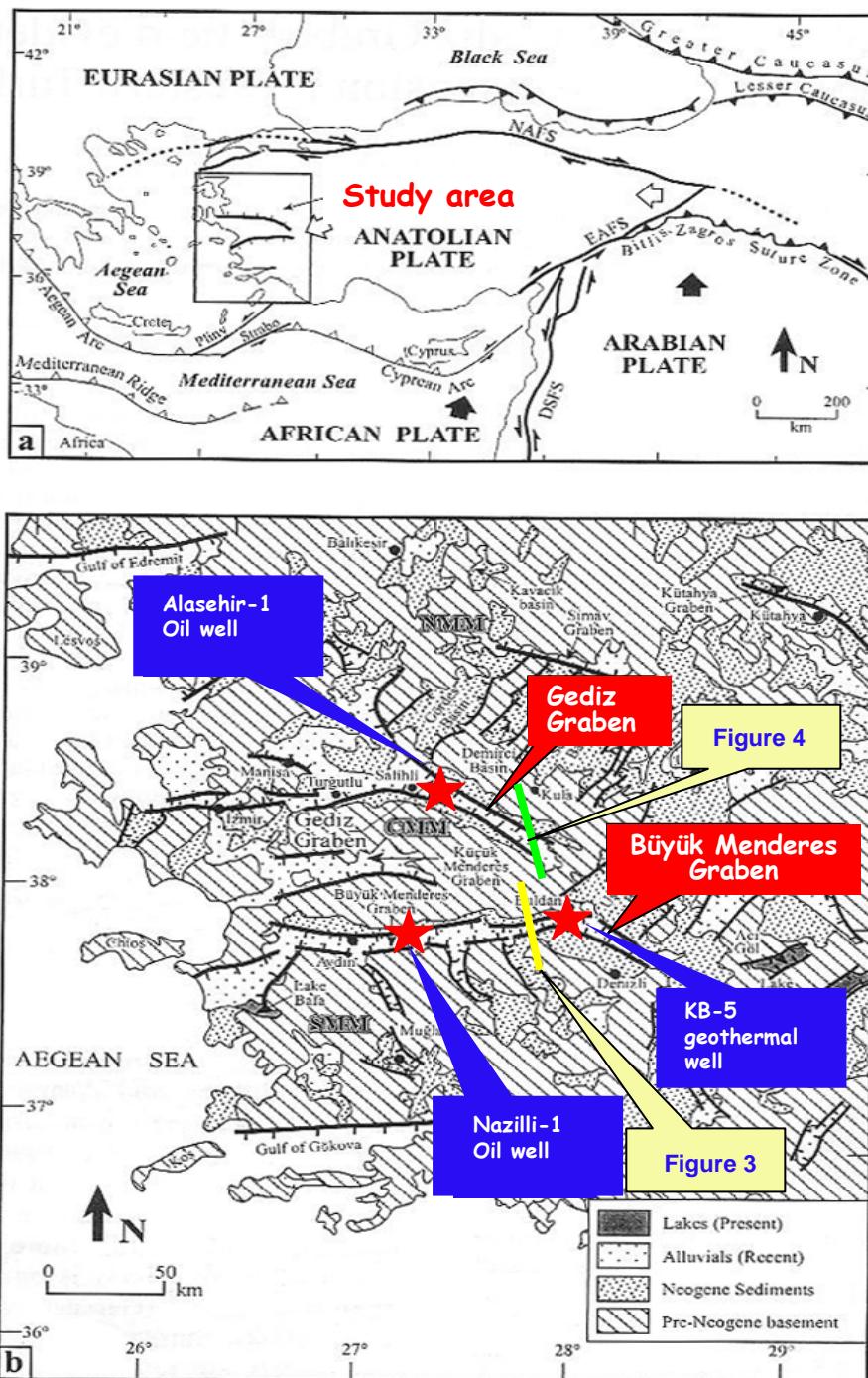


Figure 1. Map showing study area and simplified major tectonic elements of Turkey. DSFS=Dead Sea Fault System; EAFS=East Anatolian Fault System; NAFS=North Anatolian Fault System, from Barka (1992) (a) and (b) map showing Neogen and Quaternary basins and extension of Büyük Menderes and Gediz Grabens. Location of two oil wells and subdivisions of the Büyük Menderes Massif (Bozkurt and Sözbilir, 2004).

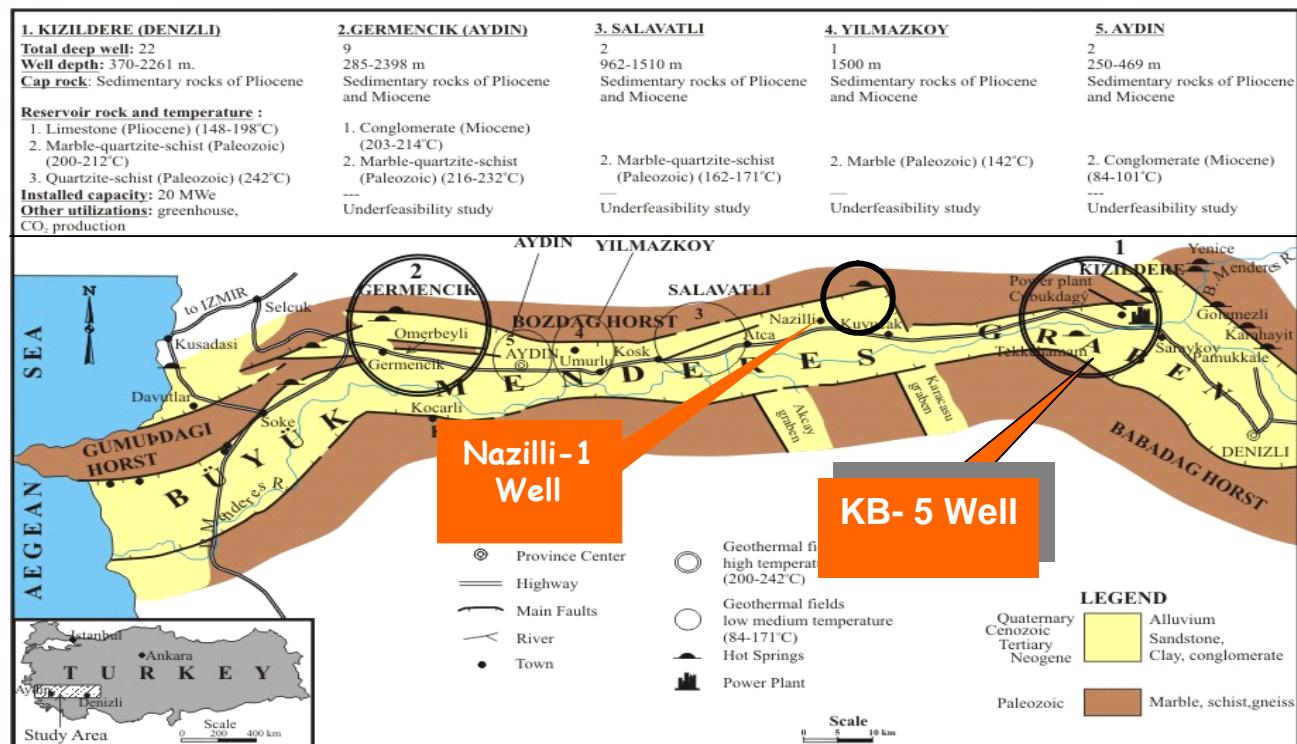


Figure 3. Close look at the Büyük Menderes Graben and locale of the KB-5 well, from which petroleum sample was taken for this study (Modified from Şimşek, 2002). Location of the Nazilli-1 dry-oil well is also shown.

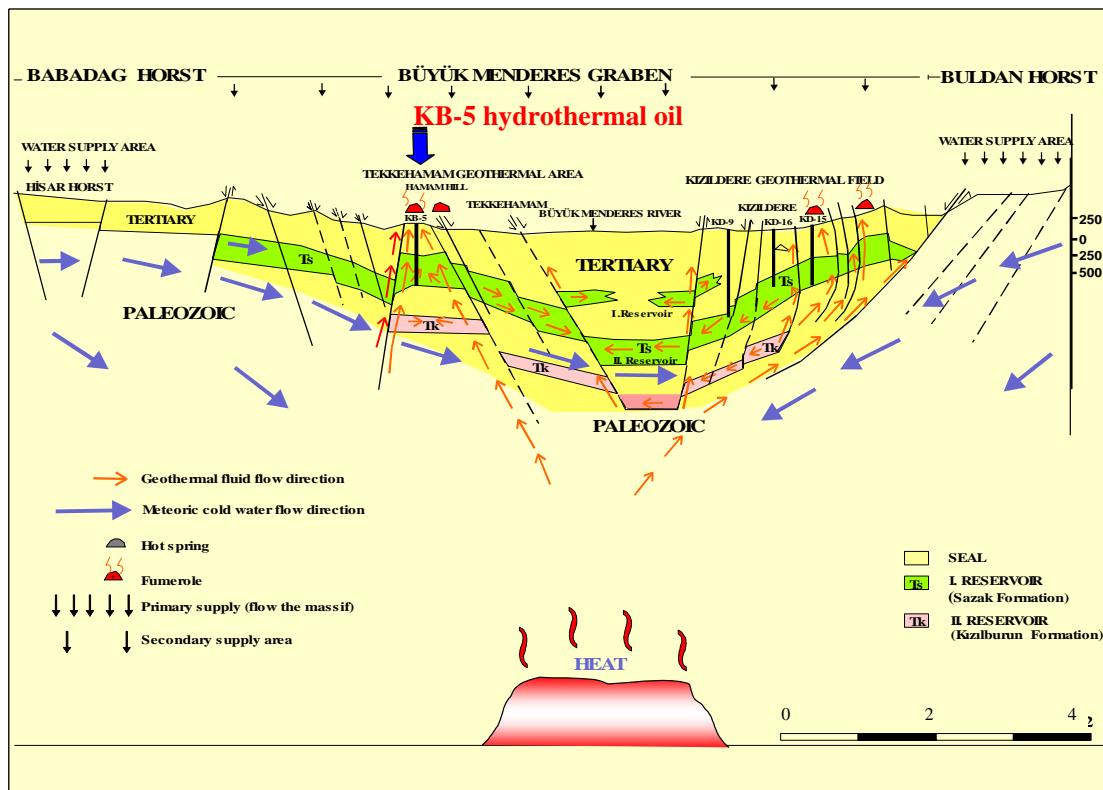


Figure 4. Schematic illustration of the hydrothermal system in Kızıldere- Tekkehıamam areas on a N-S geologic cross-section from the Büyük Menderes Graben (from Şimşek, 1985). Locale of the KB-5 well is also exhibited.

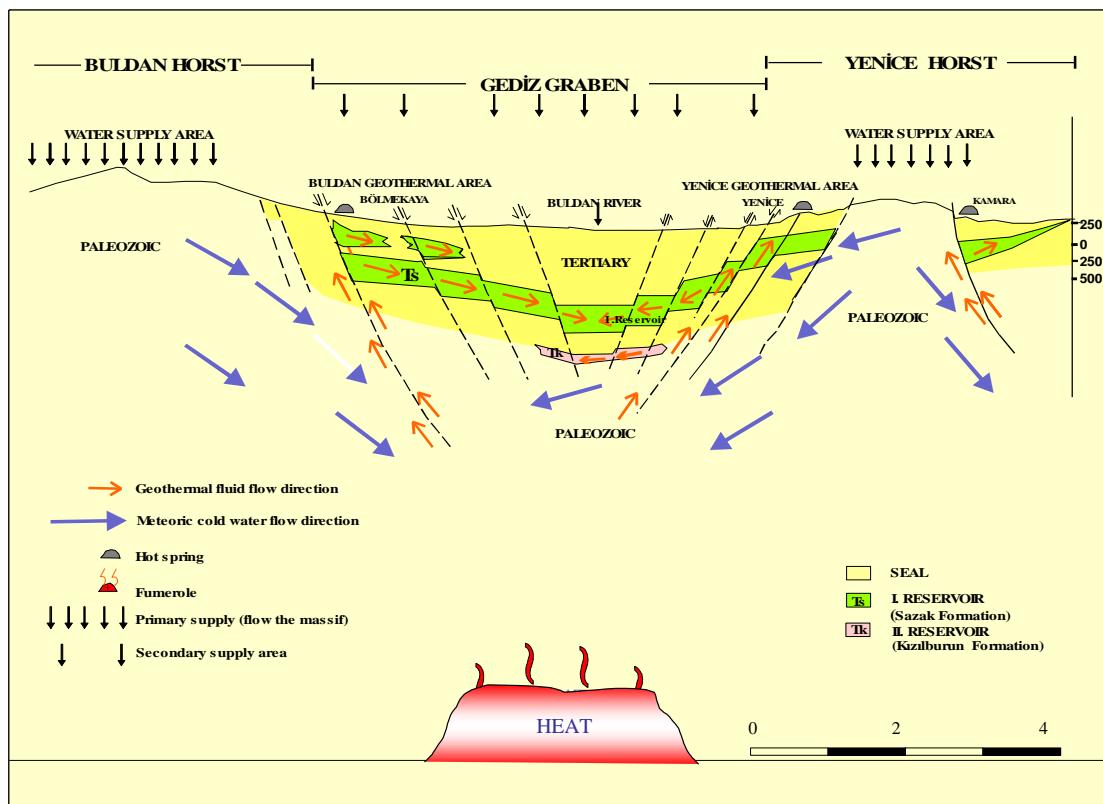


Figure 5. Schematic illustration of the hydrothermal system in Buldan-Yenice areas on a N-S geologic cross-section from the Gediz Graben (from Şimşek, 1984).

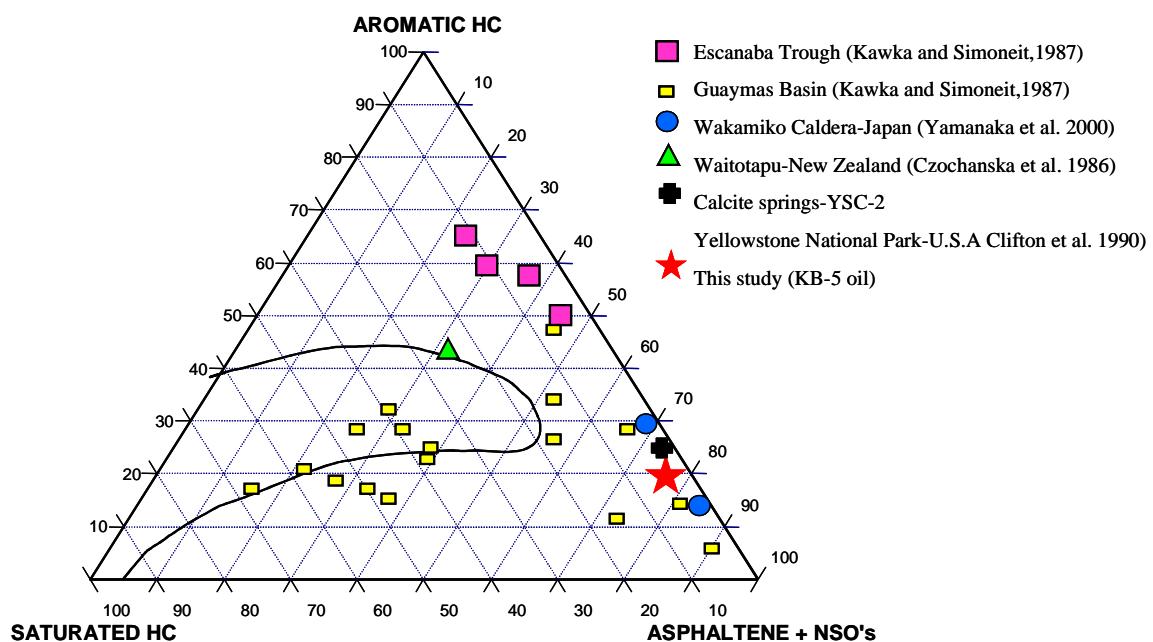


Figure 6. Ternary diagram of saturated hydrocarbons, aromatic hydrocarbons, and NSO plus asphaltic compounds. Various hydrothermal oils from different part of the world were selected for comparison purposes with the KB-5 oil from Gediz Graben. Typical crude oils fall within the curved area (Tissot and Welte).

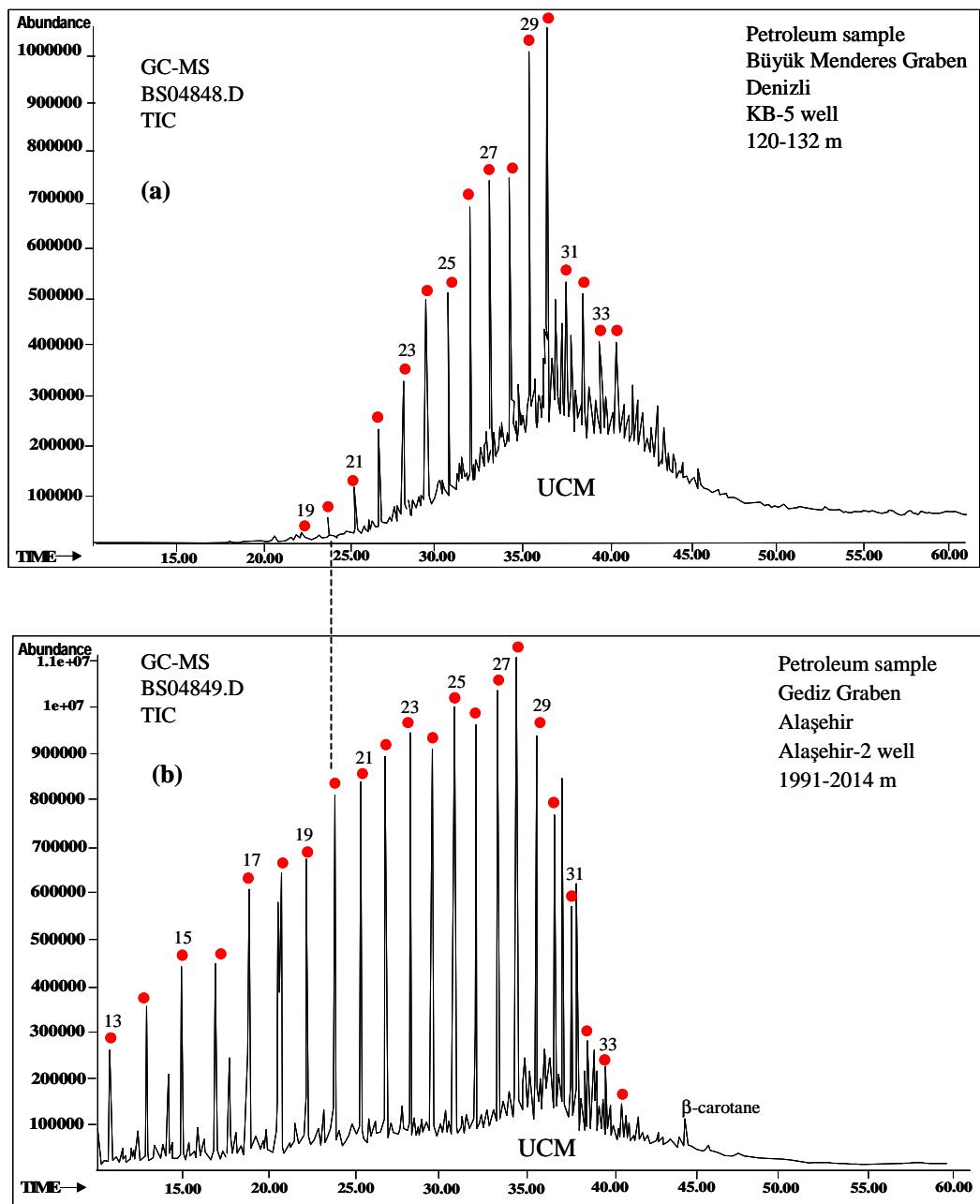


Figure 7. Capillary gas chromatogram of the saturated fraction of the KB-5 (a) and Alaşehir-1 oils (b) from the Büyüük Menderes and Gediz Grabens, respectively.

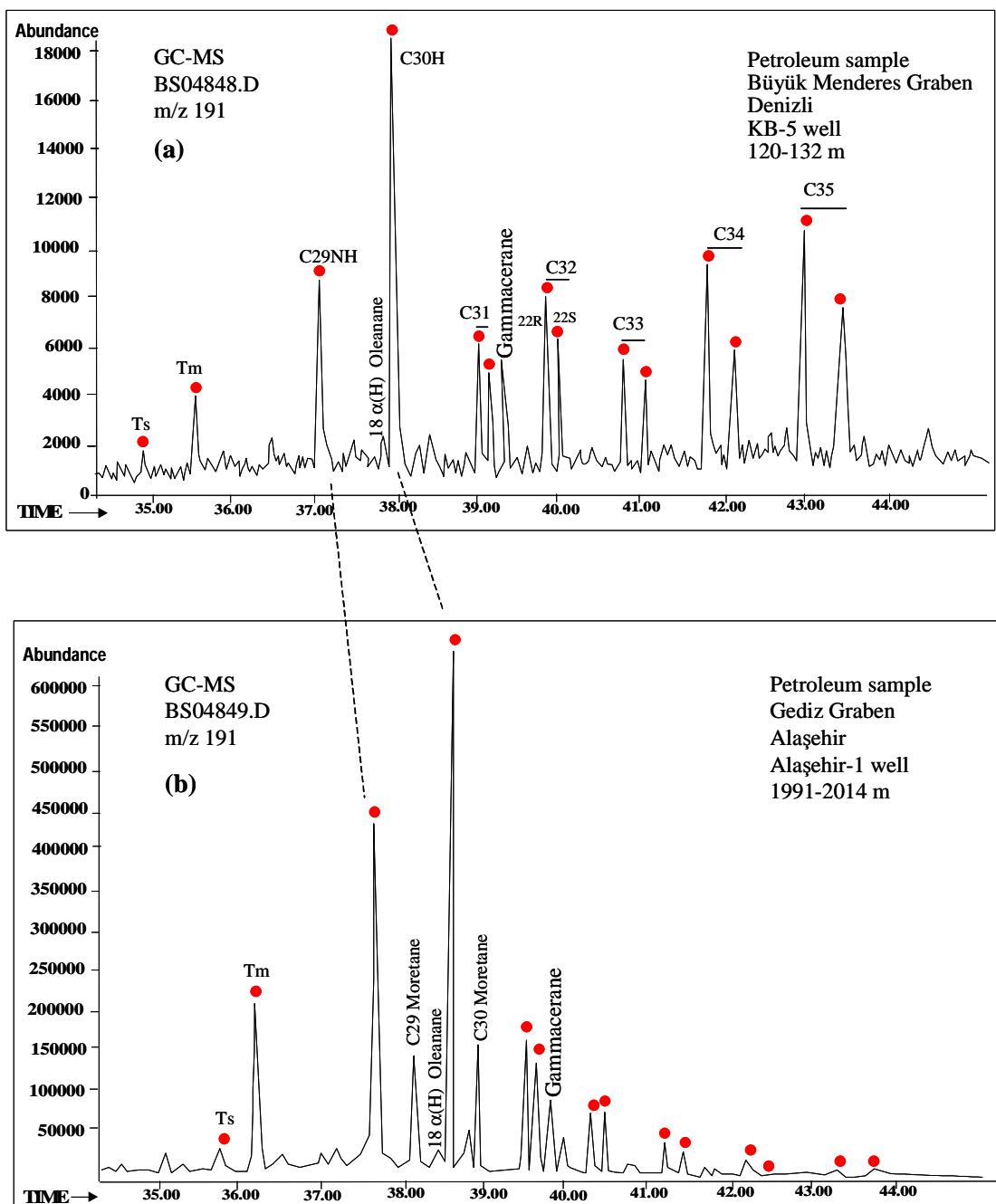


Figure 8. M/z 191 GC-MS chromatograms of the KB-5 (a) and Alaşehir-1 oils (b) from the Büyükk Menderes and Gediz Grabens, respectively.

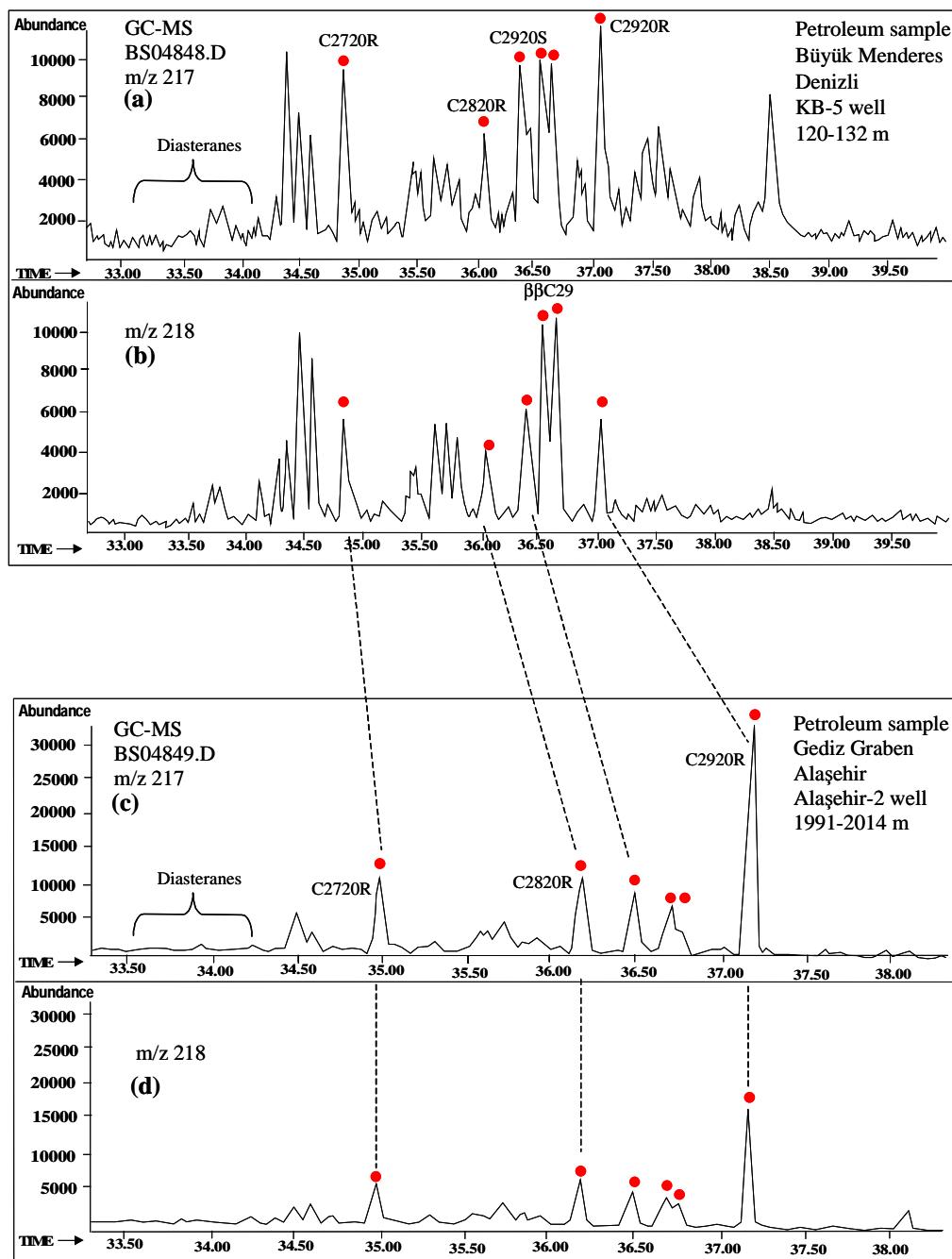


Figure 9. M/z 217 and 218 GC-MS chromatograms of the KB-5 (a,b) and Alaşehir-1 oils (c,d) from the Büyüük Menderes and Gediz Grabens, respectively.

Table 1. Comparison of geochemical parameters for oils from the KB-5 well (Büyük Menderes Graben-hydrothermal oil), Alaşehir-1 well (Gediz Graben-geothermal oil), and from the Guaymas Basin-Northern Atlantic (offshore surface sediment extract-hydrothermal oil).

	KB-5 Well	Alaşehir-1 well	Guaymas basin-1172-4
OEP 20 ¹	-	1.07	1.00
OEP 27 ²	1.02	0.98	1.01
Pr/Ph ³	-	1.20	1.40
Pr/nC18 ⁴	-	0.96	0.53
Pr/nC17 ⁵	-	1.03	0.69
(C29+C30)H/ (C29+C30)M ⁶	Moretane n.p.	4.11	3.00.
C30M/C29M ⁷	Moretane n.p.	1.15	2.20
Tm/Ts ⁸	3.00	7.75	2.90
C35/C34 ⁹	1.19	1.02	6.40
C31 22S/22S+22R ¹⁰	0.55	0.55	0.53
C32 22S/22S+22R ¹¹	0.57	0.53	0.57
C33 22S/22S+22R ¹²	0.55	0.62	0.54
C27R/C29R ¹³	0.74	0.33	2.20
C27R/C28R ¹⁴	1.93	1.00	2.10
C28R/C29R ¹⁵	0.38	0.33	1.00
C29 20S/20S+20R ¹⁶	0.35	0.32	0.28
C29NH/C30H ¹⁷	0.43	0.65	0.46
Oleanane ¹⁸	+	+	+
Gammacerane ¹⁹	+	+	+
β-Caratone ²⁰	n.p.	+	n.p.