



Mature petroleum hydrocarbons contamination in surface and subsurface waters of Kızılırmak Graben (Central Anatolia, Turkey): Geochemical evidence for a working petroleum system associated with a possible salt diapir

Adil Ozdemir^{*1}, Yildiray Palabiyik², Atilla Karataş³, Alperen Sahinoglu⁴

¹Adil Ozdemir Engineering & Consulting, Ankara, Turkey

²Istanbul Technical University, Department of Petroleum and Natural Gas Engineering, Istanbul, Turkey

³Marmara University, Faculty of Science and Literature, Geography Department, Istanbul, Turkey

⁴Istanbul Esenyurt University, Institute of Science and Technology, Istanbul, Turkey

Keywords

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ABSTRACT

Salt formations exist in Kızılırmak Graben (Central Anatolia, Turkey), which consists of volcano-sedimentary units, and it was stated in previous studies that these formations have a diapiric structure. The adjacent basin, Ayhan Basin, contains bituminous shale and operated coal deposits. For this reason, in this study, it is aimed to investigate the oil and gas potential of the Kızılırmak Graben by conducting TPH (Total Petroleum Hydrocarbons) analysis on the samples taken from natural cold-water resources by making use of the thought that hydrocarbon generation may come into existence from those units in the Ayhan basin. As a consequence of the analyses performed, hydrocarbons have been brought into the open in all the water samples. The organic geochemical methods have been used to find out the source of hydrocarbons determined in the water resources. The disclosed n-alkane hydrocarbons are the mature petroleum hydrocarbons derived from peat/coal type organic matter (Type III kerogen, gas-prone). These mature hydrocarbon-rich waters can be regarded as evidence for the availability of a working hydrocarbon system associated with possible salt diapir identified by using gravity and magnetic data obtained from the investigation area.

1. INTRODUCTION

The study area is geographically within the Central Anatolia and geologically within the Kırşehir Massif. It is located in the west of the Central Anatolia Fault Zone and in the region situated in the east of the Salt Lake Fault Zone. The regions uplifted between the Salanda and Dadağı faults in and around the study area have been named as the Hırkadağ and Ziyarettepe horst while the region in the south of the horst is called Kızılırmak Graben and Kuyulukışla Graben in the northwest of the horst (Fig. 1) (Demircioğlu, 2014). The investigation area was called Salanda Pull-Apart Basin by Koçyiğit and Doğan (2016). Paleozoic-Quaternary units are present in the area. The basement of the sequence contains Paleozoic-Mesozoic metamorphic rocks. Tertiary sedimentary units unconformably cover these units. Marine units are of Late Paleocene-Middle Eocene. Depending on the climatic conditions and tectonism in the region, lacustrine environments developed in

Miocene and then in micro-scaled basins. In the region, the units formed in the stream environment in different ages and lithologies are also observed. Besides, igneous rocks are quite common in the study area and its surroundings. For this reason, the region is also called the Cappadocia Volcanic Province (Demircioğlu, 2014). In the inspected area, the Alpine orogen phases have become effective and faults and folds have developed based on this orogenesis. NW-SE oriented Tuzköy Fault Set, Salanda, Tuzköy, Yüksekli, and Gülşehir faults are also remarkable faults in the area (Fig. 2).

In Turkey, old aged and thick Tertiary natural salt deposits currently exist. The main deposits of these formations are located in Central Anatolia (Çankırı and Nevşehir regions), Eastern Anatolia (Erzurum region), Southeastern Anatolia (Çukurova region), and Tuz Gölü (Salt Lake) region (Fig. 3) (Kayhan, 1976). The Tuzköy salt deposit in the study area (Nevşehir region) is set foot in the depth of 60 m from the surface and has the thickness of 150 m (Fig. 2). In the deposit, 75 million tons

* Corresponding Author

^{*}(adilozdemir2000@yahoo.com) ORCID ID 0000-0002-3975-2846
(palabiyiky@itu.edu.tr) ORCID ID 0000-0002-6452-2858
(atilla.karatas@marmara.edu.tr) ORCID ID 0000-0001-9159-6804
(alperensahinoglu@esenyurt.edu.tr) ORCID ID 0000-0002-1930-6574

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of proved, 96 million tons of probable and 959 million tons of possible NaCl reserves have been determined. The presence of rock salt deposits in Gülşehir is not only limited to the Tuzköy rock salt deposit but also continue apart from the deposit (Kayakıran, 1979; Ünüçok, 1985). The folded rock salt deposit formation compatible with an NW-SE extended anticline is covered by younger units (Barutoğlu, 1961; Burkay and Önder, 1986). Rock salt formations in the study area are characterized by a diapiric structure (Bilginer, 1982).

There are numerous oil and gas production areas related to salt structures in the world. In those types of structures in Turkey, any economically viable hydrocarbon field has not been discovered so far and so, any significant hydrocarbon potential could not be determined as well. Therefore, in this study, it has been

aimed to identify the salt structure and its hydrocarbon potential based on the findings that it has a diapiric and deep structure which was stated by Bilginer (1982) and that it continues on the outside of the deposit that is the fact that was expressed by Ünüçok (1985). For this purpose, firstly, the possible boundaries of the salt structure are determined by regional gravity and magnetic data. Then, TPH (Total Petroleum Hydrocarbons) analyses are carried out on the samples taken from natural water resources within and around the specified limits. As a result of the analyses, mature petroleum hydrocarbons have been determined in all water samples. The detected mature hydrocarbons can be considered as evidence for a working petroleum system associated with possible salt diapir in the study area.

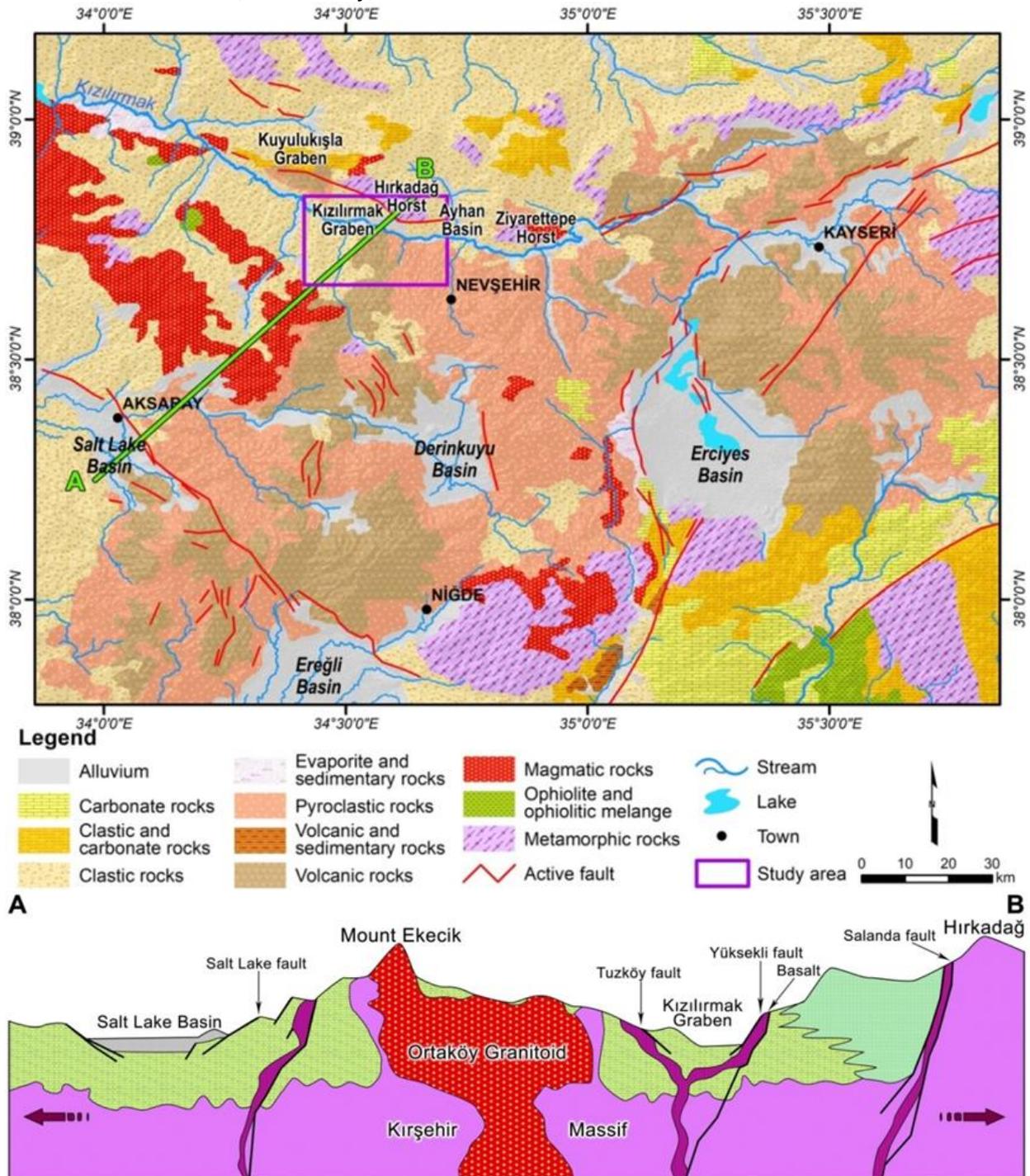


Figure 1. Location map of Kızılırmak Graben (modified from MTA, 2002 and Demircioğlu, 2014)

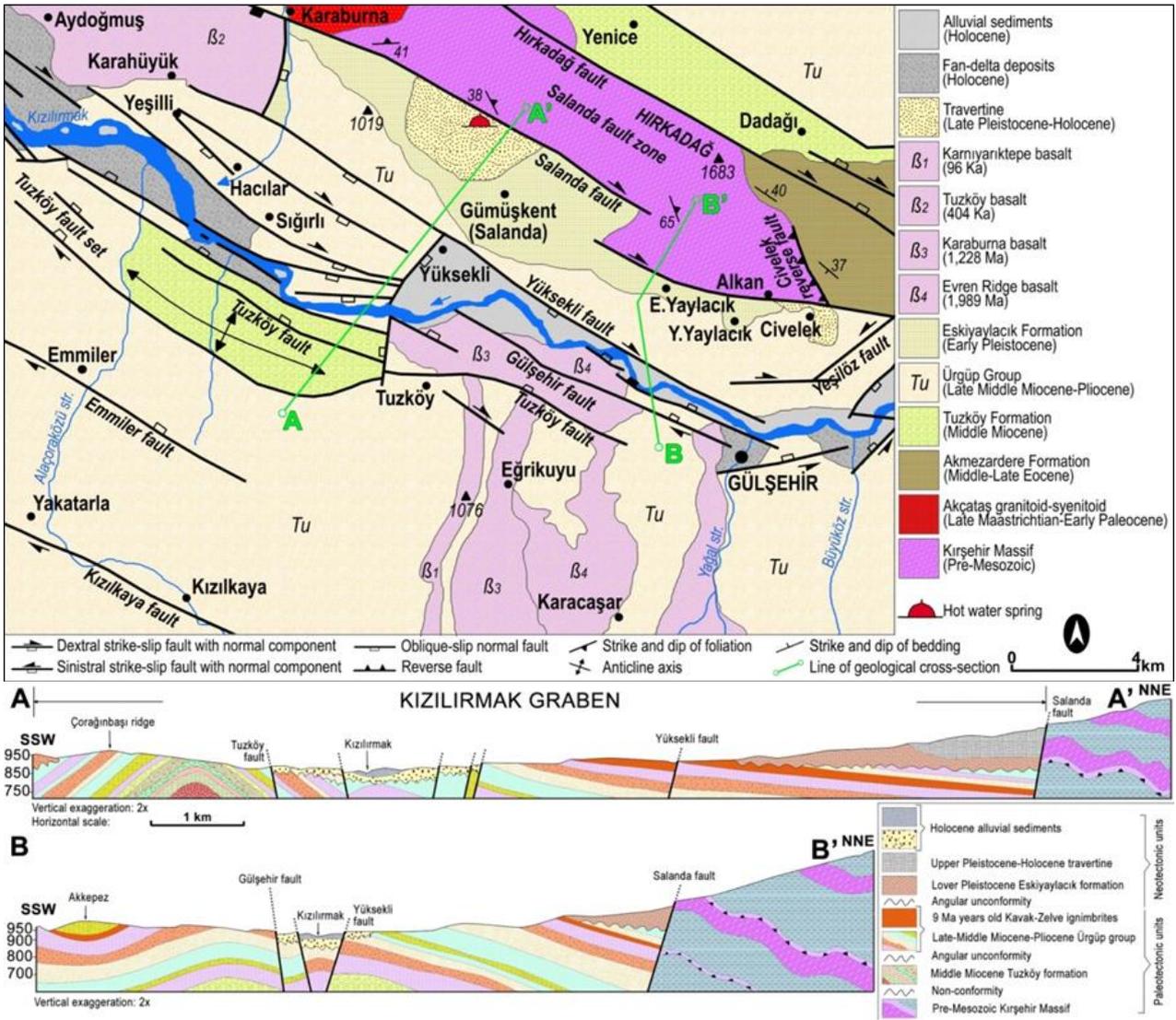


Figure 2. Geology map of the study area (modified from Koçyiğit and Doğan, 2016)

2. MATERIAL and METHOD

Eymold et al. (2018) have determined that shallow groundwaters above the basin formations containing shale gas are enriched in hydrocarbons. They have also expressed that the hydrocarbons in these hydrocarbon-rich waters migrated from deep source rocks to shallow aquifers. Moreover, Kreuzer et al. (2018) have mentioned that the faults in petroliferous basins make the transportation of hydrocarbon-rich brines to aquifer formations above source rocks easy by influencing the geochemistry of shallow groundwaters and cause a hydrocarbon enrichment in these waters. On the other hand, in recent times, TPH in water analysis has started to be utilized in petroleum exploration, which allows the determination of hydrocarbon-rich waters and organic geochemical properties in basins/regions where source rocks are not exposed at the surface as outcrops (covered basins) or has been exhausted (depleted or spent) (Ozdemir 2019a-c; Karataş et al., 2019; Palabiyik et al., 2019, 2020; Palabiyik and Ozdemir, 2020; Ozdemir et al., 2020). Furthermore, in recent studies, it has been come into existence that all organic geochemical analyses conducted on source rock and gas samples can also be applied to hydrocarbon-rich surface and subsurface waters determined by TPH in water analysis and that the

same analysis and interpretation results for the same basins/regions have been reached (Fig. 4) (Ozdemir 2018; Liu et al., 2018). According to these studies, if an oil and/or gas reservoir exists in a region, the surface and subsurface waters in the study area should be rich in mature petroleum hydrocarbons (Fig. 5). Therefore, it can be inferred that the technique of TPH analysis in water will significantly contribute to reservoir-targeted oil and gas exploration activities.



Figure 3. Location map of Tuzköy rock salt mine (Nevşehir) (edited from Kayhan, 1976)

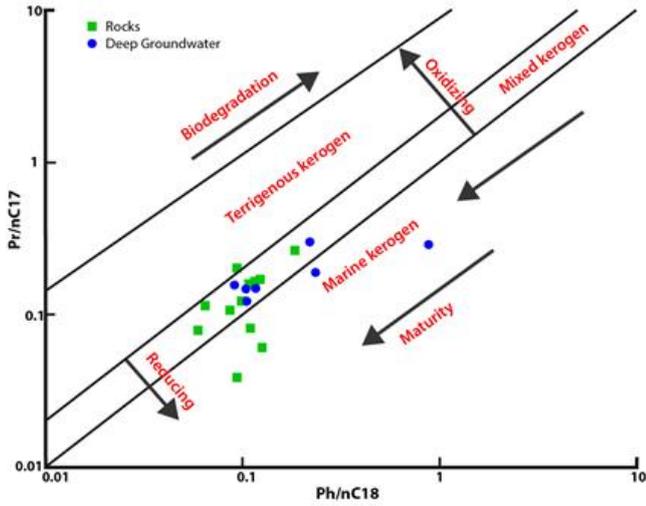


Figure 4. Ph/nC18 vs Pr/nC17 ratios of deep groundwater and rock samples taken from the same region (Liu et al., 2018) (Pr: Pristane and Ph: Phytane isoprenoid hydrocarbons, nC17 and nC18: n-alkane hydrocarbons)

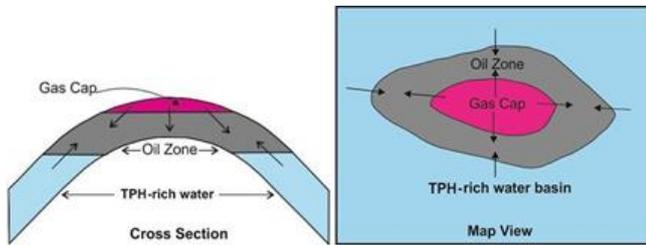


Figure 5. Components of an anticlinal type of petroleum reservoir that can be produced by primary methods (Ozdemir, 2018)

TPH value provides information on hydrocarbon contamination of water resources. Gas chromatography (GC) analyses are performed to detect the TPH concentrations of the water contaminated by

hydrocarbons. In the determination of TPH content, the standard test method “the Determination of Hydrocarbons: Solvent extraction and gas chromatography method (ISO 9377-2)” is used (other methods: EPA Method 1664 and ASTM D7678-11). In this technique, aromatic hydrocarbons are separated, and the total amount of petroleum hydrocarbons is determined in the samples taken from the surface, subsurface, and distribution waters. These samples are stored by an acidification process to prevent the issues, which may affect the number of hydrocarbons, such as evaporation or biodegradation in the samples. Samples are analyzed within 14 days if acidified, or they are performed within 7 days if not done, and stored at $5^{\circ}\text{C} \pm 3^{\circ}\text{C}$ before the analysis.

In the comprehension of this research, totally 25 samples have been taken by the scaled polyethylene bottles of 1 liter from the natural flowing waters (cold water fountains) in the region (Figs. 6 and 7). The water samples are taken from the untreated water resources that are not related to tap water (running water). Since the water samples collected from the study area have been analyzed a few days after the sampling, no acidification process has not applied to the samples. They were collected and preserved according to the standard procedures (ISO 5667-3) and analyzed in the laboratory for TPH in water employing the standard methods (ISO 9377-2). In the samples, the TPH analyses have been conducted by a gas chromatography device in the laboratory to generate data for organic geochemical evaluations. Thus, direct TPH concentrations of the water samples (in mg/l) have been determined depending on the analyses and the required geochemical parameters (CPI, NAR, etc.) to be discussed in detail in the next section of the paper have been calculated by making use of gas chromatograms. In the geochemical evaluations, the TPH concentrations and the calculated parameters are utilized.



Figure 6. Location map of the taken water samples. Yellow circles: the water samples, yellow polygon: Tuzköy rock salt mine



Figure 7. A view of water sampling procedure from cold water fountains (pure and clean natural flowing waters) in the study area by using scaled polyethylene bottles

3. FINDINGS and DISCUSSION

Based on the TPH analysis results regarding the water samples taken from the study area, concentrations, biodegradation conditions, source, maturity, and redox conditions of the depositional environment of the hydrocarbons in the waters are investigated in a geochemical point of view. Moreover, the aeromagnetic and gravity maps prepared for the study area are interpreted in terms of geological and tectonic aspects, and the construction of the conceptual occurrence, migration, and accumulation model of the hydrocarbons is targeted.

Table 2. TPH analysis results of the water samples and the calculated parameters

Sample No.	Water Resource	Coordinates		TPH (mg/l)	CPI	TAR	P _{aq}	P _{wax}	Waxiness Index	n-C17/n-C31	NAR	Pr/Ph	Pr/n-C17	Ph/n-C18	n-alkane maksimum
		X	Y												
1	Natural flowing water	4300669	635154	0.52	1.24	-	0.28	0.72	-	0.20	0.32	4.88	0.29	0.14	C31
3	Mineral water	4300753	632969	< 0.4	1.34	-	0.26	0.74	-	0.14	-	7.67	0.39	0.13	C31
4	Natural flowing water	4299892	633152	0.72	1.35	-	0.26	0.74	-	0.15	0.25	5.84	0.26	0.10	C31
5	Natural flowing water	4297621	635602	0.56	1.36	-	0.26	0.74	-	0.19	0.23	5.95	0.29	0.09	C29
6	Natural flowing water	4291566	640269	0.64	1.18	5.79	0.29	0.71	4.80	0.14	0.37	10.87	0.24	0.09	C31
8	Natural flowing water	4288798	633507	0.62	1.41	-	0.23	0.77	-	0.21	0.25	5.30	0.27	0.11	C31
10	Water well	4287546	635389	0.63	1.27	7.91	0.22	0.78	-	0.14	0.37	5.68	0.28	0.12	C31
11	Natural flowing water	4292077	630459	0.68	1.82	-	0.10	0.90	5.58	0.10	0.27	9.28	0.25	0.08	C31
13	Salt discharge water	4292277	629466	0.57	1.26	-	0.28	0.72	-	0.15	0.29	3.63	0.21	0.13	C31
14	Natural flowing water	4291278	620839	0.53	1.36	-	0.24	0.76	-	0.15	0.25	5.45	0.30	0.10	C31
16	Natural flowing water	4287752	618043	0.72	1.26	-	0.22	0.78	-	0.13	0.37	5.61	0.23	0.10	C31
17	Natural flowing water	4292465	619808	0.41	-	-	-	-	-	0.11	-	8.43	0.40	0.09	C31
19	Water well	4292981	617241	0.52	1.55	-	0.17	0.83	-	0.08	0.20	10.12	0.32	0.06	C31
20	Natural flowing water	4294291	614037	0.43	1.60	7.70	0.18	0.82	4.27	0.12	0.10	9.95	0.30	0.08	C31
21	Water well	4301664	614292	0.48	1.90	10.03	0.07	0.93	3.63	0.22	0.12	13.05	0.30	0.05	C29
23	Natural flowing water	4301125	619556	0.53	1.71	7.91	0.12	0.88	2.95	0.11	0.08	10.00	0.24	0.06	C31
26	Natural flowing water	4302065	622556	0.65	-	7.20	-	-	-	0.17	-	15.95	0.25	0.04	C31
27	Natural flowing water	4299109	624796	0.97	1.93	7.25	0.08	0.92	3.37	0.17	0.00	4.60	0.21	0.09	C31
28	Caisson well	4296453	626478	0.80	1.72	7.01	0.11	0.89	1.92	0.22	0.00	5.52	0.19	0.10	C29
29	Natural flowing water	4295935	630637	0.63	1.93	6.89	0.07	0.93	1.86	0.17	0.05	11.60	0.23	0.05	C31
31	Natural flowing water	4294888	639800	0.60	1.75	-	0.11	0.89	-	0.17	0.02	9.82	0.26	0.05	C20
32	Water well	4284729	637224	0.54	1.85	-	0.11	0.89	-	0.10	0.02	7.42	0.32	0.08	C20
33	Natural flowing water	4276181	638653	0.59	1.64	-	0.17	0.83	-	0.15	0.05	18.50	0.27	0.04	C29
35	Mineral water	4282436	655103	0.47	1.66	-	0.10	0.90	-	0.32	0.12	9.43	0.24	0.07	C20
36	Mineral water well	4283652	655804	0.66	1.27	-	0.10	0.90	-	0.10	-	7.43	0.25	0.07	C31

CPI = $\{[(C23+C25+C27) + (C25+C27+C29)] / [2 * (C24+C26+C28)]\}$ (Bray and Evans, 1961), TAR = $(C27+C29+C31)/(C15+C17+C19)$ (Bourbonniere and Meyers, 1996), NAR = $[\sum n\text{-alk} (C_{19-32}) - 2\sum \text{even } n\text{-alk} (C_{20-32})] / \sum n\text{-alk} (C_{19-32})$ (Mille *et al.*, 2007), Waxiness Index: $\sum (n-C21-n-C31) / \sum (n-C15-n-C20)$ (Peters *et al.*, 2005), P_{aq} = $(C23+C25)/(C23+C25+C27+C29+C31)$ (Ficken *et al.*, 2000), P_{wax} = $(C27+C29+C31)/(C23+C25+C27+C29+C31)$ (Zheng *et al.*, 2007), - : Could not be calculated.

3.1. Contents, Source, and Biodegradation of Hydrocarbons in Waters

Liu *et al.* (2018) have defined groundwater of which hydrocarbon concentration exceeds 0.05 mg/l as original hydrocarbon-rich groundwater. The TPH limit values recommended for surface and subsurface waters are given in Table 1. Surface and subsurface waters exceeding the TPH values in Table 1 are defined as hydrocarbon-rich waters. The n-alkane hydrocarbons have been found in all the water samples in the study area. The hydrocarbon content of the water samples is much higher than the limit values suggested for the waters (Tables 1 and 2). Hence, it can be mentioned that water-rock-hydrocarbon interactions have created this hydrocarbon enrichment in waters.

Table 1. The TPH limit values recommended for surface and subsurface waters

TPH (mg/l)	Reference
< 0.05	Liu <i>et al.</i> (2018)
< 0.1	Zemo and Foote (2003)
< 0.5	Ozdemir (2018)
< 0.2	Ministry of Agriculture and Forestry of Turkey (2004a), Surface Water Quality Regulation of Turkey (Appendix 5, Table 2: Oil and Grease)
< 0.02	Ministry of Agriculture and Forestry of Turkey (2004b), Water Pollution Control Regulation of Turkey (Appendices Table 1: Oil and Grease)

Source, maturity, migration, and biodegradation are the main elements responsible for the compositional changes in hydrocarbons. Ph/n-C18 value less than 1 indicates non-biodegraded hydrocarbons (Hunt, 1995). Ph/n-C18 values of all the water samples are less than 1 and according to these values, the hydrocarbons in the water samples are in a non-biodegraded character.

By making use of gas chromatography analysis outcomes, the Carbon Preference Index (CPI), Pr/Ph ratio, isoprenoid/n-alkane ratio (Pr/nC17 and Ph/nC18) have been computed, and the n-alkane distributions have been evaluated. In this study, Pr/Ph ratio (Didyk *et al.*, 1978; Tissot and Welte, 1984; Banga *et al.*, 2011), Carbon Preference Index (CPI) (Bray and Evans 1961, 1965; Tissot and Welte, 1984), Pr/Ph versus CPI (Onojake *et al.*, 2013; Hakimi *et al.*, 2018), and Pr/n-C17 versus Pr/Ph (Syaifudin *et al.*, 2015; Larasati *et al.*, 2016; Devi *et al.*, 2018) plots are utilized to assess the water samples.

CPI is an indicator for the source of n-alkanes. The CPI, a ratio between the amounts of n-alkanes with odd and even carbon number, is calculated by measuring the heights of the peaks in gas chromatograms. The dominant peaks in these chromatograms are represented by n-alkanes. In the computation of the CPI, various equations have been proposed by numerous researchers. This index can be applied to any range of the carbon sequence. It is utilized to assess the kind of organic matter, the depositional environment, and thermal maturity. It is remarkably greater than 1 (odd n-alkane preferential) or lower than 1 (even n-alkane preferential), indicating thermally immature oil or bitumen samples (Tissot and Welte, 1984; Peters and Moldowan, 1993). A high value of CPI in the immature or low-maturity sample means the input of organic matter derived from higher terrestrial plants (Tran and Philippe, 1993). According to the CPI values (Table 2), the source of n-alkanes in the water samples remarks the petrogenic hydrocarbons and old organic-rich sediments (Table 3).

Table 3. Source of n-alkanes in water according to CPI value (Ozdemir, 2018)

CPI	Source
> 2.3	Young terrestrial sediments (biogenic hydrocarbons)
1.2 - 2.3	Old organic matter-rich sediments (marine shales, limestones, etc.)
≤ 1.2	Petrogenic hydrocarbons (values < 1 biodegraded oils)

The term petrogenic sources describe unburned fossil resources like crude oil and coal. These types of sources were formed very slowly at moderate temperatures (between 100°C and 300°C) millions of years ago (Beyer *et al.*, 2010). The parameter NAR (Natural n-alkane Ratio) has been suggested to interpret the source of hydrocarbons in the environment (natural or petroleum n-alkane). This ratio is zero or approximately zero for natural petroleum hydrocarbons and crude oil. In other hydrocarbon sources, those ratios are greater. According to the parameter NAR (Table 2), all the n-alkanes in the water samples indicate natural petroleum (petrogenic) hydrocarbons.

The parameter TAR (Terrestrial/aquatic hydrocarbon ratio) shows the ratio of n-alkanes derived from terrestrial organic matter to n-alkanes derived from aquatic algae (Cranwell *et al.*, 1987; Goossens *et al.*, 1989; Meyers and Ishiwatari, 1993; Bourbonniere and Meyers, 1996). High values of TAR (greater than 1) sign terrestrial plant source and its low values (less than 1) mean marine algae source (Kroon, 2011). The TAR values have been calculated as quite high for the inspected water samples (greater than 1) (Table 2). These values show that the n-alkanes having high carbon numbers indicating terrestrial organic matter are dominant in the water samples in the examined area.

Waxiness index can be utilized to determine the amount of terrestrial organic matter. This index depends on the assumption that regional terrestrial organic matter contributes to extracts with the n-alkane components with high molecular weight (Peters *et al.*, 2005). It is observed that the water samples in the investigation area have high Waxiness values indicating high amounts of biomarkers derived from terrestrial plant (Table 2). This finding is also supported by the fact that the analyzed samples show high TAR values.

The n-C17/n-C31 ratio indicates the source of the hydrocarbons derived from the organic matter in the environment. High values (higher than 2) correspond to marine algae while low values (lower than 2) sign land plant sources (Forster *et al.*, 2004). The ratio of n-C17/n-C31 of the water samples ranges from 0.08 to 0.32 by indicating the terrestrial organic matter. This consequence is consistent with TAR and Waxiness index values.

By calculating P_{aq} and P_{wax} parameters, some interpretations can be made about the plant species that make up the organic matter and the paleoclimate conditions of the environment (Zheng *et al.*, 2007). These parameters are used only for coals. If P_{aq} value is less than 0.1, it will mean terrestrial plants; if it is between 0.1 and 0.4, it will refer to aquatic plants (floated in the swamp environment), and if it is between 0.4 and 1.0, it will mean that environments exist the presence of plants floated in the water (Ficken *et al.*, 2000). According to P_{aq} values (Table 2), the types of plants that constitute the organic matter-deriving hydrocarbons in the water samples are predominantly aquatic plants (floated in the swampy environment). If the P_{wax} value is lower than 0.7, it means dry climate conditions whereas if it is less than 0.7, it will mean the existence of humid climate conditions (Zheng *et al.*, 2007). According to the P_{wax} values (Table 2), the organic matter-deriving hydrocarbons in the water samples was formed in dry climate conditions.

3.2. Maturity of Hydrocarbons in Waters and Redox Conditions of Sedimentation Environment

CPI value of mature hydrocarbons is equal to 1 or close to 1 (Waples, 1985). The CPI values of oil and bitumen related to very salty carbonate or evaporitic environments are lower than 1 (Tissot and Welte, 1984; Peters and Moldowan, 1993). The maturity level of hydrocarbons is classified based on their CPI values (Table 4) (Onojake *et al.*, 2013), and depending on this

classification, all the hydrocarbons in the water samples (Table 2) can be classified as mature (more oxidizing) level.

The n-alkanes, which are the closest to isoprenoids in gas chromatograms, are utilized for isoprenoid/n-alkane ratios. The Pr/Ph ratio is an appropriate correlation parameter. Even though pristane (Pr) and phytane (Ph) define other sources, they are derived from phytyl, which is the side chain of chlorophyll, particularly in phototropic organisms. Under anoxic conditions, the side chain of phytyl breaks down to form the phytol, while phytol is also reduced to pristane under oxic conditions (Peters and Moldowan, 1993). Hence, the Pr/Ph ratio shows the redox potential of the depositional environment. Pr/Ph values lower than 1 reflect anoxic conditions while the values higher than 1 remark oxic conditions (Didyk *et al.*, 1978; Hunt, 1995). The water samples in the study area exhibit a high Pr/Ph ratio varying from 3.63 to 18.50 (Table 2). Therefore, the water samples contain the hydrocarbons derived from sediments deposited in an oxic environment (Pr/Ph > 1). The Pr/Ph ratio also provides information about paleoenvironment and maturity (Volkman and Maxwell, 1986) level. In the Pr/Ph versus CPI relationship, it can be observed that the hydrocarbons in the water samples are located in the more oxidizing zone and have similar maturity levels (Fig. 8).

Table 4. The maturity level of hydrocarbons according to CPI value (from Onojake *et al.*, 2013) (see Fig. 8)

CPI	Maturity
>1	Mature (oxidizing-reducing)
0.8 - 1	Mature
< 0.8	Immature

Pr/n-C17 and Ph/nC18 ratios are commonly made use of in petroleum correlation studies. Samples containing high Pr reflect an oxidizing source, and high Ph content indicates a reducing source. Thus, the plot of Pr/n-C17 versus Ph/n-C18 is utilized to distinguish petroleum or bitumen in different groups (Hunt, 1995). Although the Pr/Ph ratio above 1.5 shows settling conditions in an oxygenated environment based on a standard geochemical interpretation, it is well-known that it may be lower than 1 for an anoxic depositional environment. Lower values may indicate less suitable oxic conditions than the other parts of the same sequence (Hartkopf-Fröder *et al.*, 2007). The ratio of isoprenoid/n-alkane decreases with increasing maturity as more amounts of n-alkanes release from kerogen affected by a breaking down process (Tissot and Welte, 1984; Hunt, 1995) and is used as a degree of maturity for biodegradable oil and bitumen samples. It increases with the biodegradation (Hunt, 1995) and is also affected by organic matter input and secondary phenomena. Depending on their positions in the Pr/nC17 versus Ph/nC18 plot of the water samples, it is observed that the source rocks which generated the hydrocarbons in the water samples are deposited in oxic terrestrial (Type III kerogen, gas-prone) environment and exhibit a high maturity (Figs. 8-12 and Table 5).

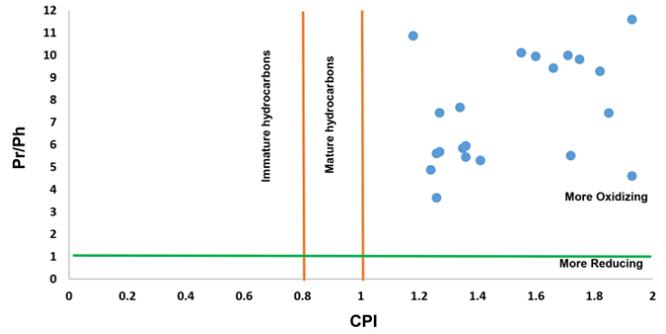


Figure 8. Pr/Ph vs CPI plot of the water samples (the plot: from Onojake *et al.* 2013)

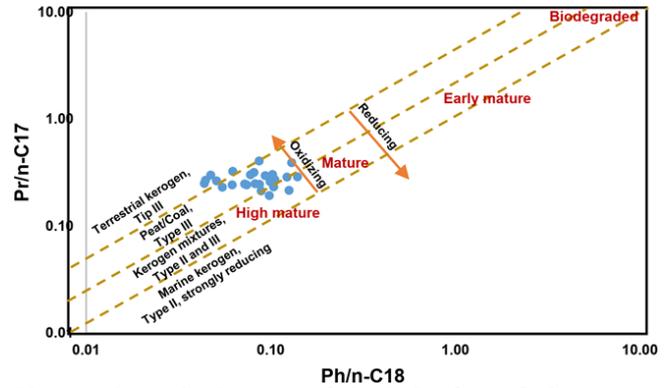


Figure 9. Pr/n-C17 vs Ph/n-C18 plot of the water samples (the plot: from Peters *et al.* 1999)

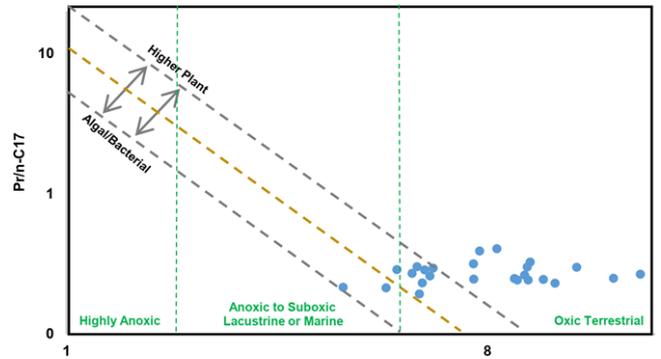


Figure 10. Pr/n-C17 vs Pr/Ph plot of the water samples (the plot: Syaifudin *et al.*, 2015; Larasati *et al.*, 2016; Devi *et al.*, 2018)

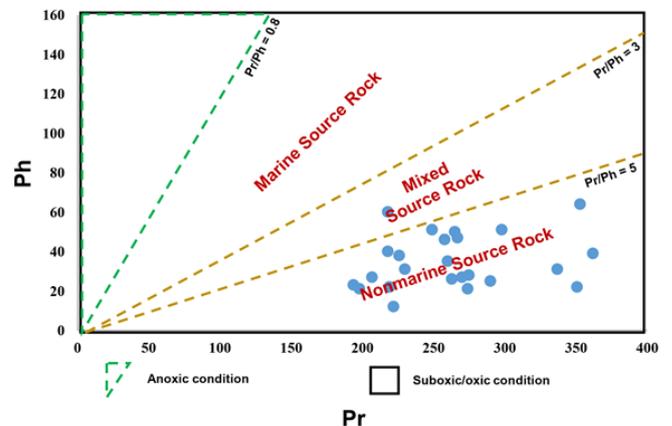


Figure 11. Ph vs Pr plot of the water samples (the plot: from Banga *et al.*, 2011)

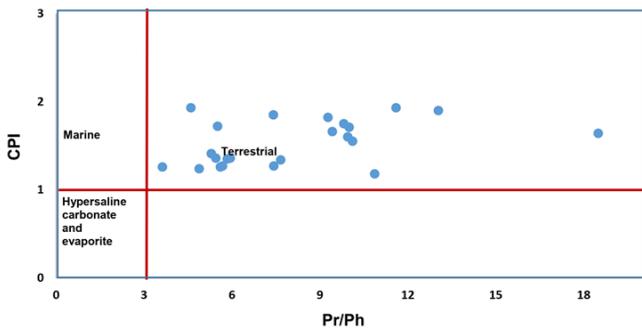


Figure 12. Pr/Ph vs CPI plot of the water samples (the plot: from Hakimi *et al.*, 2018)

Table 5. Source rock and depositional environment of hydrocarbons according to Pr/Ph value (from Banga *et al.* 2011) (see Fig. 11)

Pr/Ph	Source rock	Pr/Ph	Environment
< 3	Marine	< 0.8	Anoxic
3 - 5	Marine	> 0.8	Suboxic-Oxic
	Terrestrial		
> 5	Terrestrial		

3.3. Aeromagnetic and Gravity Maps of the Study Area and Geological Interpretations

There are a lot of studies regarding the methods and field applications of gravity and aeromagnetic data for use in oil and gas exploration (Griffin, 1949; Nettleton, 1976; Geist *et al.*, 1987; Lyatsky *et al.*, 1992; Gadirov, 1994; Piskarev and Tchernyshev, 1997; Pašteka, 2000; Aydın, 1997, 2005; Gadirov and Eppelbaum, 2012; Ivakhnenko *et al.*, 2015; Satyana, 2015; Eke and Okeke, 2016; Stephen and Iduma, 2018; Gadirov *et al.*, 2018; Ozdemir, 2019a-c; Ozdemir *et al.*, 2020). Analysis of gravity and magnetic anomalies has been a permanent component of hydrocarbon exploration and discovery in West Siberia for half a century (Piskarev and Tchernyshev, 1997). In the regions where the existence of mature petroleum hydrocarbons is proved by the determination of hydrocarbon-rich waters in this research, particularly seismic surveys are crucial to determine the locations of oil and gas reservoir(s)/trap(s). Unfortunately, no seismic lines/measurements are available in the investigated area. The contour maps specifically prepared for the

study area from regional gravity and aeromagnetic data measured by the General Directorate of Mineral Research and Exploration of Turkey (MTA) have been utilized to evaluate subsurface geology of the area.

Salt diapir-dome structures are characterized by negative gravity anomalies (reverse gravity anomaly) (Nettleton, 1976; Sharma, 1986; Blood, 2001; Jallouli *et al.*, 2005; Pinto *et al.*, 2005; Stadler *et al.*, 2014; Nava-Flores *et al.*, 2016; Constantino *et al.*, 2019; Pourreza and Hajizadeh, 2019). Jallouli *et al.* (2005) stated that as a result of the misinterpretation of the gravity anomaly observed in salt structures, a positive gravity anomaly of the calculated gravity anomaly can be produced, but the calculated gravity anomaly should produce negative gravity anomaly as compatible with the observed gravity anomaly (Fig. 12). The gravity anomaly of the study area containing Kayatuzu mine is also negative (Fig. 14).

The gravity map prepared for the study area (Fig. 12) contains young sediments composed of lower-density sedimentary origin rocks (siltstone, mudstone, claystone, conglomerate, shale, etc.) and metamorphic (slate, phyllite, etc.) rocks represented by dark blue, light blue, and green colors. In the areas featured by orange, red, and yellow colors, an anomaly is characterized by the rocks having relatively higher densities (crystallized limestone, marble, quartzite, schist, etc.). The study area is limited by strike-slip faults, and the part between the two NW-SE and SE-NW trending main faults has uplifted. This uplifting part was broken with many possible secondary faults.

The prepared aeromagnetic map for the study area (Fig. 12) demonstrates the anomalies that originated from fully non-magnetic sedimentary (sandstone, limestone, siltstone, mudstone, claystone, conglomerate, shale, etc.) and metamorphic (crystallized limestone, marble, quartzite, schist, etc.) rocks represented by blue, green, and light green colors. In the areas represented by yellow, red, and white tones, there are the rocks with magnetic properties (pebbly volcanic sandstones, ophiolites, dikes, etc.). In gravity and magnetic maps prepared for the study area, both the gravity and magnetic anomalies of the possible salt diapir, whose possible limits have been determined, are negative. This result is Koşaroglu *et al.* (2016) is compatible with the data.

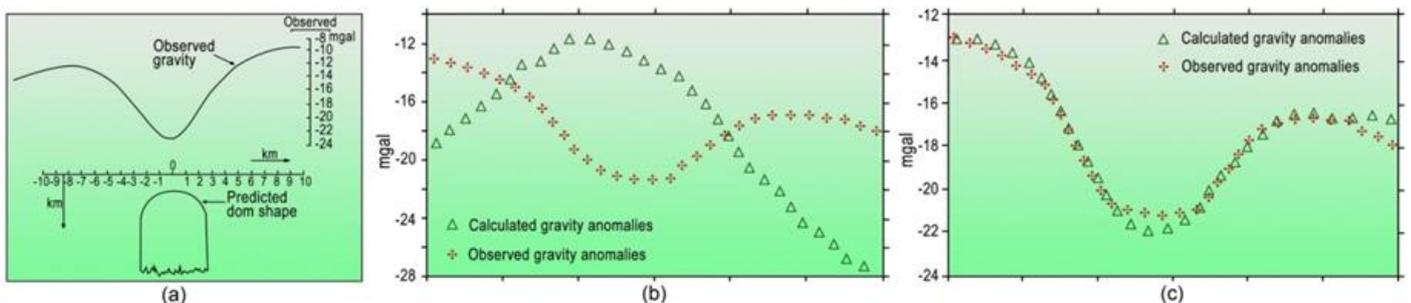


Figure 13. a. Gravity anomaly of Humble salt dome (Harris Country, Texas, USA) (modified from Nettleton, 1976). b) A mistakenly produced calculated gravity anomaly interpretation for a salt dome, c) a correctly produced calculated gravity anomaly interpretation (modified from Jallouli *et al.* 2005)

In the study, the interpretation method proposed by Svancara (1983) and Töpfer (1977) is used to convert the 2D (two-dimensional) residual gravity anomalies into the depth values for the estimation of the basin and structure depths. In this method, if the density contrast is known, the depth of the sedimentary basin or structure can be determined by simple relations established between gravity anomaly and parameters. The first stage of the interpretation gives the characteristic parameters of the anomaly (Figure 13). The relevant equations can be expressed as follows:

$$A = \frac{g_{mak}}{W_a \sigma} \quad (1)$$

Where

g_{mak} : Maximum amplitude of the gravity anomaly,
 W_a : Distance corresponding to the half amplitude ($g_{mak}/2$) value of the gravity anomaly,
 σ : Density contrast.

$$\frac{W_b}{W_a} = (-0.056A) + 1.827 \quad (2)$$

$$D_o = 23.866 \frac{g_{mak}}{\sigma} \quad (3)$$

For the condition $0 < A < 9$ (Töpfer, 1977):

$$\frac{D}{D_o} = 0.072A + 1.00 \quad (4)$$

For the condition $9 < A < 13$ (Töpfer, 1977):

$$\frac{D}{D_o} = 0.12A + 0.57 \quad (5)$$

where

W_b : Full width of the gravity anomaly,
 D : Depth corresponding to the gravity anomaly value,
 D_o : Depth obtained from the flat-plate formula,
 D : Maximum depth.

3.4. Conceptual Occurrence, Migration, and Trapping Model of Hydrocarbons in the Investigation Area

Ozdemir (2019a, b), Ozdemir and Palabiyik (2019a, b) and Ozdemir *et al.* (2020) have mentioned that petroleum source rocks are formed in the mid-ocean ridges and the continental rifts (spreading centers). Hence, Hasandağı, Kızılırmak, and Keçikalesi volcanisms accompanying the extensional regime in the study area, possibly in the Lower (?) - Middle Miocene period, should have caused the formation of the source rocks which generated the hydrocarbons in the water samples (Figs. 1 and 17). Intense volcanic activities (Gürbüz *et al.* 2019) that have taken place during all Neogene and Quaternary periods are also thought to be effective in maturing of hydrocarbons detected in the water samples.

In the Ayhan basin which is the adjacent basin of the study area, bituminous shale formations and coal deposits characterizing the swampy environment exist in the villages of Avcıköy, Alemlı, Dadağı, and Ayhan along

with their surroundings (Figs. 1 and 18). In the Ayhan Basin, a partial lacustrine-swampy environment prevailed in the Middle Miocene. Coal units in the basin were deposited in this environment. Lignite veins have the thicknesses ranging from 80 to 100 cm and their calorific values are quite high (around 5000 kcal) (Erguvanlı, 1954; Reul, 1954; Becker, 1956; Lebküchner, 1957; Wedding, 1967; Inoue, 1967; Akgün *et al.*, 1995; Taka *et al.* 2004; Demircioğlu, 2014). The n-alkane hydrocarbons determined in the water samples are the mature petroleum hydrocarbons derived from predominantly peat/coal type organic matter (Type-III kerogen, gas-prone) (Fig. 9). Therefore, the hydrocarbons in the water samples may have migrated from the coal beds in the neighboring Ayhan basin and Kuyulukışla graben to the study area. Another possibility is that they may have been derived from the possible coal deposits in the Kızılırmak graben or in the Derinkuyu basin. Gürbüz *et al.* (2019) have claimed in their paleontological and palynological study that there was a swampy environment in and around the investigation area in the Lower (?) - Middle Miocene and the units of this period contain lignite seams (Fig. 18).

The mature hydrocarbon-rich waters are evidence for a working petroleum system in the study area. The possible gas reservoirs in the study area are the structures around this salt diapir, which is determined by the gravity and magnetic maps (Figs. 16 and 19). The fact that the main structure is located between two strike-slip fault zones empowers the possibility of having accumulated of the hydrocarbons in this structure. The hydrocarbons in the water samples are enriched as a consequence of the water-rock-hydrocarbon interaction with the hydrocarbon-rich geological units in the subsurface and/or mixed with shallow groundwaters migrated from the potential reservoir(s) determined by gravity and magnetic maps.

4. CONCLUSION

In this study, which is aimed to investigate the oil and gas potential of Kızılırmak Graben using TPH analysis performed on the samples taken from water resources, the hydrocarbons have been determined in all the water samples. The TPH values are remarkably greater than the hydrocarbon limit values suggested for surface and subsurface waters. The source of the n-alkanes in the water samples are petrogenic hydrocarbons and organic-rich sediments. Water-rock-hydrocarbon interactions have caused a hydrocarbon enrichment in the waters in the inspected area, and the hydrocarbons in the water samples are featured by a non-biodegraded character. It is brought out that the source rocks which generated the hydrocarbons in the water samples were deposited in the swamp environment (Type III kerogen, gas-prone) as well as corresponding to the mature-overmature level. The possible gas reservoirs in the study area are the structures around this salt diapir, which is determined by the gravity and magnetic maps.

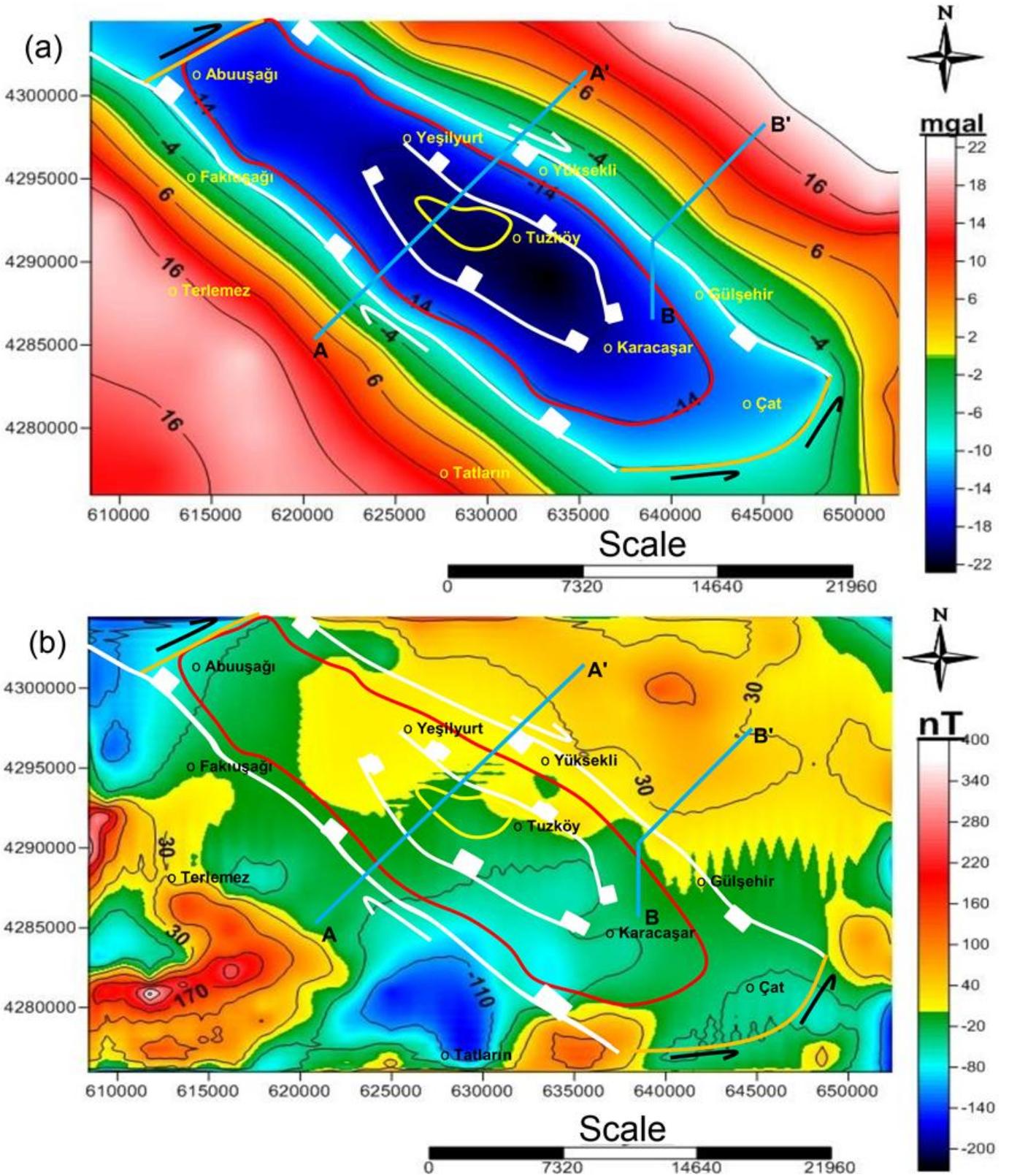


Figure 14. The colored contour maps and geological interpretations of the regional gravity (a) and aeromagnetic (b) anomalies of the study area. Red polygon: possible gas trap in the Kızılırmak Graben, white lines: oblique strike-slip faults (teeth show the hanging wall), A-A 'and B-B': geological cross-section lines (Fig. 3), A-A' line: profile taken to determine the depth of the structure (Fig. 12).

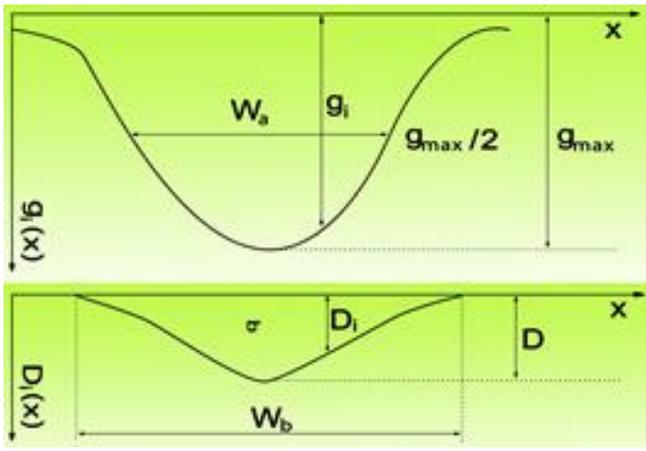


Figure 15. Ideal gravity anomaly of a basin and characteristic parameters (Svancara, 1983)

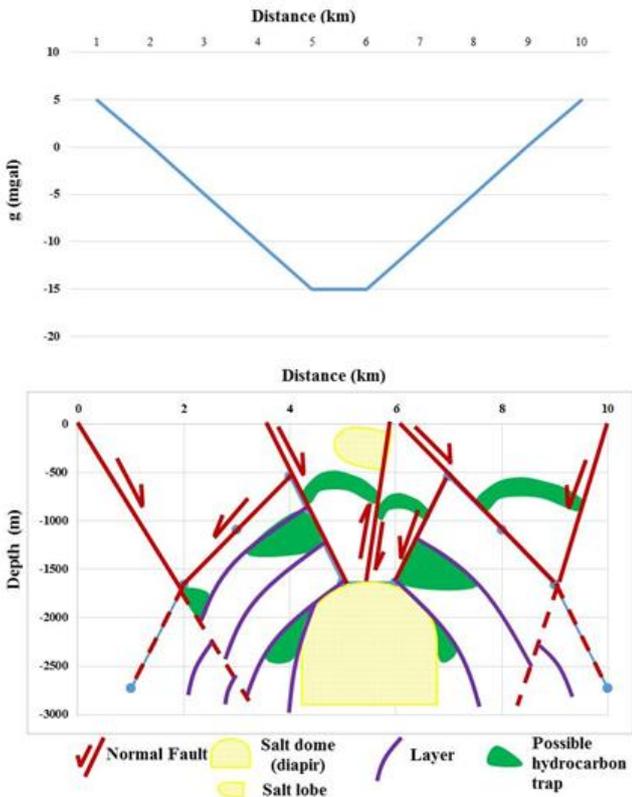


Figure 16. Geological interpretation of A-A ' profile and depth of the possible salt diapir in the study area (see Fig. 12).

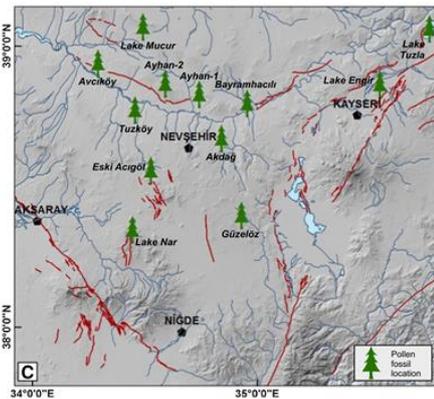


Figure 18. The Miocene paleoenvironment reconstruction based on the geological unit, volcanic age, and palynological data of the study area and its surroundings (Gürbüz *et al.* 2019)

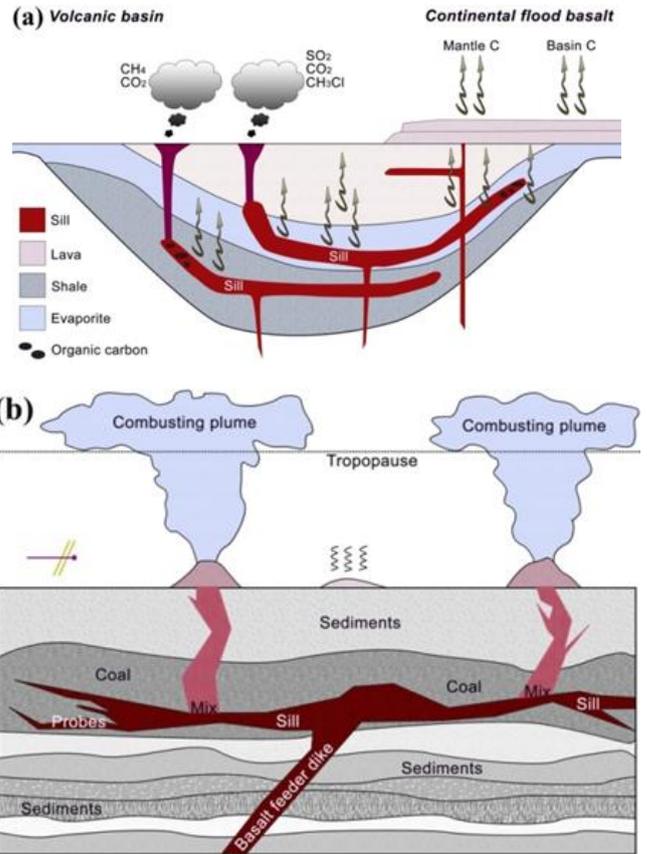


Figure 17. Schematic cross-section showing sill and dykes across a volcanic basin. The chemical composition of sedimentary rocks heated by igneous intrusions has a significant effect on the composition of the metamorphic fluid. For example, organic-rich shale produces CH_4 during contact metamorphism, while coal produces CO_2 -derived fluids and also water. Many sedimentary basins with sill settlement may contain hydrogen-rich kerogen and oil and gas deposits, and fluids such as methane (CH_4) and ethane (C_2H_6) can be enriched in the basin (Svensen *et al.*, 2015). (a: Svensen *et al.*, 2015; b: Ogden and Sleep, 2011)

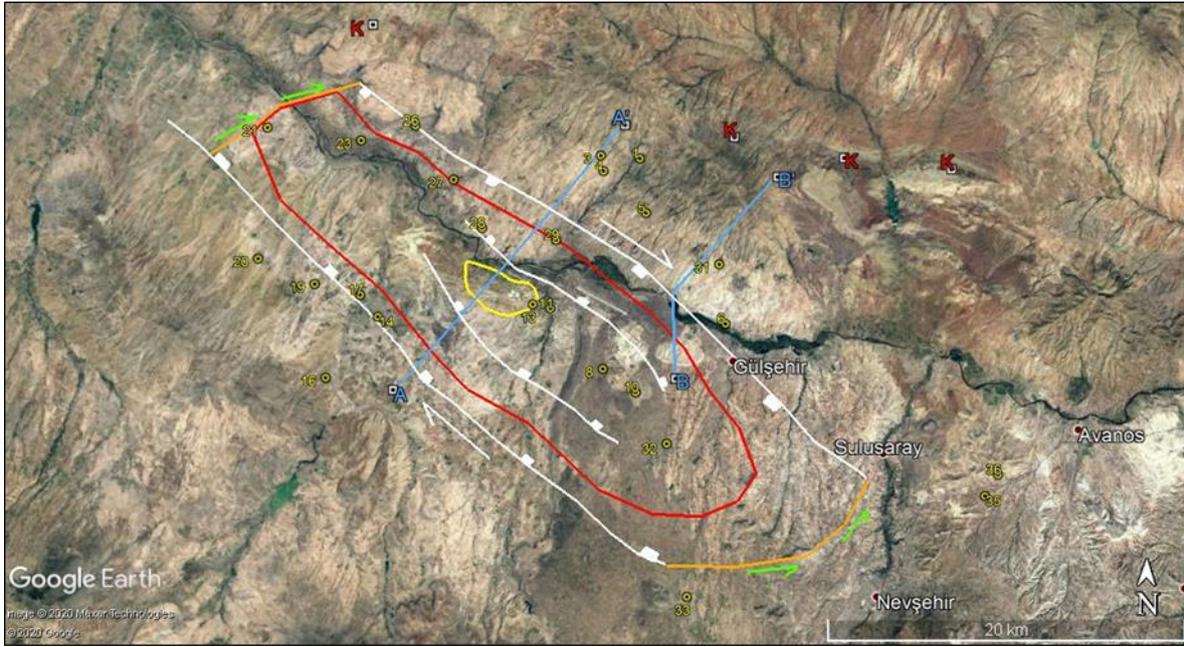


Figure 19. Possible gas trap in the study area (red polygon). White lines: oblique strike-slip faults (teeth show the hanging wall), yellow polygon: Tuzköy rock salt mine, K: coal (lignite) field, A-A' and B-B': geological cross-section lines (Fig. 3), A-A' line: profile taken to estimate the depth of the structure (Fig. 14)

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Author Contributions

Adil Ozdemir conceived of the presented idea. He developed the theory and wrote the manuscript. **Yildiray Palabiyik** verified the analytical methods. He encouraged Adil Ozdemir to investigate [a specific aspect] and supervised the findings of this work. **Atilla Karataş** carried out the experiments and plotted figures. **Alperen Sahinoglu** contributed to the design and implementation of the research. All authors discussed the results and contributed to the final manuscript.

Conflicts of interest

The authors declare no conflicts of interest.

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