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ORGANIC GEOCHEMICAL CHARACTERISTICS AND SOURCE ROCK POTENTIAL OF UPPER PLIOCENE SHALES IN THE AKÇALAR LIGNITE BASIN, TURKEY

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Abstract. *In this study, organic geochemical characteristics of Upper Pliocene bituminous shale beds in the Akçalar lignite basin, southwest of Konya, Turkey, were investigated using total organic carbon (TOC) and pyrolysis analyses. Additionally, the palaeontological study was carried out to assess the depositional environment of the area. The TOC contents of the studied samples are mostly high, varying between 0.4 and 50 wt%, with an average of 21.35 wt%. The low Hydrogen Index (HI = 17–395 mg HC/g TOC) and* S_2/S_3 *ratio (0.15–11.4) values indicate that kerogen of the vast majority of the samples is of Type III and only a few samples contain Type II and IV kerogens. Consequently, most of the organic matter (OM) tends to generate gas, even though a small portion of it exhibits a tendency to generate gas-oil mixture. The studied bituminous rock samples characterized by low* T_{max} *(408–511 °C) and Production Index (PI = 0.08–0.36) values show that the degree of thermal maturity is in the range of immature to early mature.* S_1, S_2 *and Potential Yield (PY) values range from 0.04 to 16.16 mg HC/g rock, 0.07 to 115.56 mg HC/g rock and 110 to 131720 ppm, respectively. These values imply, in terms of hydrocarbon generation potential, that the studied samples have a source rock potential from poor to excellent. Additionally, the* S_1 *hydrocarbon type values indicate no external contribution of migrated hydrocarbons to the bituminous rocks of the studied area.*

The lacustrine sediments including coal and bituminous shale beds in the Akçalar lignite basin were deposited in a fresh water lacustrine environment with periodically changing depositional conditions from deep to shallow and to stagnant swamp.

Keywords: *Akçalar lignite basin, total organic carbon, oil shale pyrolysis, maturity degree.*

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1. Introduction

As petroleum is a natural resource that is being rapidly consumed, there are made great efforts worldwide to seek possible substitutes for it. The reserves of oil shale stand out as an important source potentially substituting petroleum [1]. Oil shale is commonly defined as a fine-grained sedimentary rock containing organic matter (OM) that yields substantial amounts of oil and combustible gas upon destructive distillation [2]. The widely distributed deposits around the world and their vast amounts make them a promising option as a source of oil. In Australia, oil shales have been the source of energy products for a long time [1]. The United States has the largest oil shale resources in the world, totaling 3340 billion tons, which constitutes 62% of the world's known recoverable oil shale potential. The most important deposits are the Green River oil shale and the Devonian black shales [2, 3]. Brazil has the world's second largest oil shale reserve in the Irati shale in Sao Mateus do Sul, Parana. China has an important amount of oil shale potential in Manchuria and particularly near Fushun. Estonia has the largest oil shale industry active today; for example, having accumulated in shallow sea basins, its Ordovician Kukersite deposit is of high oil shale potential [4].

Oil shale is an important potential energy resource for Turkey, and is considered the second largest fossil energy source after lignite. Oil shale deposits in Turkey are mainly distributed in central and western Anatolia. These are predominantly found in the organic rich marl, carbonate and clay successions of Paleocene-Eocene and Middle-Late Miocene ages [5]. The most important oil shale deposits, in terms of quality, amount and exploitability, are Miocene Beypazarı (Ankara), Seyitömer (Kütahya), Himmetoğlu (Bolu) [6–9] and Paleocene-Eocene Hatildağ (Bolu) [10] deposits. The reserves of these deposits are 330, 120, 660 and 360 million tons, respectively.

The oil shale of the Seyitömer field located north of Kütahya City consists of a variety of predominantly grey, green-grey and brown marl with intercalated brightly coloured marly limestone and silicified limestone and chert [11]. The main maceral components are laminated algae, pollen and planktonic algae, with traces of liptodetrinite and humic organic material [12].

The oil shale of the Beypazarı field consists mostly of well consolidated marl, clay, bituminous marl, dolomitic limestone, dolomite and magnesite, with intercalated tuffaceous horizons and occasionally chert. The microscopically recognizable organic material consists mainly of liptinite, huminite and prebitumen [12, 13].

The Himmetoğlu and Hatıldağ oil shale fields are located in the Bolu Basin, south-east of Bolu City, northwestern Anatolia. The Miocene-aged Himmetoğlu oil shale is comprised of bituminous banded marl and liptinite and huminite as the main maceral components. On the other hand, the Paleocene-Eocene Hatıldağ oil shale is made up of brown-grey, brown and beige banded bituminous calcareous or dolomitic marl and bituminous marl;

intercalated bands or strips of sterile marl are frequent. At the same time, the microscopically recognizable organic matter consists of liptinite, prebitumen and huminite [12, 13].

Moreover, there are other potentially important oil shale resources in Mengen (Bolu), Ulukışla (Niğde), Bahçecik (İzmit), Burhaniye (Balıkesir), Beydili (Ankara), Dodurga (Çorum) and Demirci (Manisa), but these deposits have not been investigated in detail [4, 5].

The area studied in this paper, the lacustrine Akçalar lignite basin of Late Pliocene age, is located in the south of Bayafşar, a village between Seydişehir and Beyşehir, 90 km southwest of Konya, central Turkey (Fig. 1). The lacustrine sediments in the basin include coal and bituminous shale beds of varying thickness. The potential reserves of lignite are estimated at about 45,326,000 tons [14]. The studied area and its surroundings have been studied earlier [14–16] for geological and economical (coal reserve determination) purposes. The goal of this study is to investigate the organic geochemical characteristics (total organic carbon (TOC) content, type and thermal maturity), depositional environment and hydrocarbon potential of bituminous shales in the Akçalar lignite basin.

2. Geological setting

The studied area lies in the central part of the Tauride Orogenic Belt [17]. The main tectonic units in this area are the autochthonous Geyikdağı Unit and allochthonous Bozkır [18] and Beyşehir-Hoyran Napı Units [19]. The latter two units, Bozkır and Beyşehir-Hoyran Napı, overlap the autochthonous Geyikdağı Unit by thrust fault.

The Geyikdağı Unit dominates in the studied area and is represented by the Upper Cambrian-Lower Ordovician Seydişehir Formation [20], composed essentially of a succession of shale-sandstone alternating with meta-sandstone. The Seydişehir Formation is followed by Mesozoic neritic carbonates and detritic units. Then, Mesozoic sedimentary units were unconformably succeeded by Miocene-Pliocene fossiliferous limestone, marl and poorly fossiliferous massive mudstone and sandstone of the Taşlıca Formation [21]. Over the whole region, all the aforesaid formations are followed with angular unconformably by Quaternary travertine and alluvium deposits (Fig. 1).

The Taşlıca Formation in the studied area is comprised of interbedded argillaceous limestone as the dominant lithology, bituminous claystone, fossiliferous limestone and marl, clayey coal and bituminous and fossiliferous coal (Figs. 2, 3 and 4). Based on the borehole data obtained from Egemad Enerji Mandencilik Company in the region, the thickness of the organic carbon rich-sediment in the Taşlıca Formation varies between 1 and 99 meters, whereas its total thickness in an exposed coal quarry in the area is 21.17 meters.

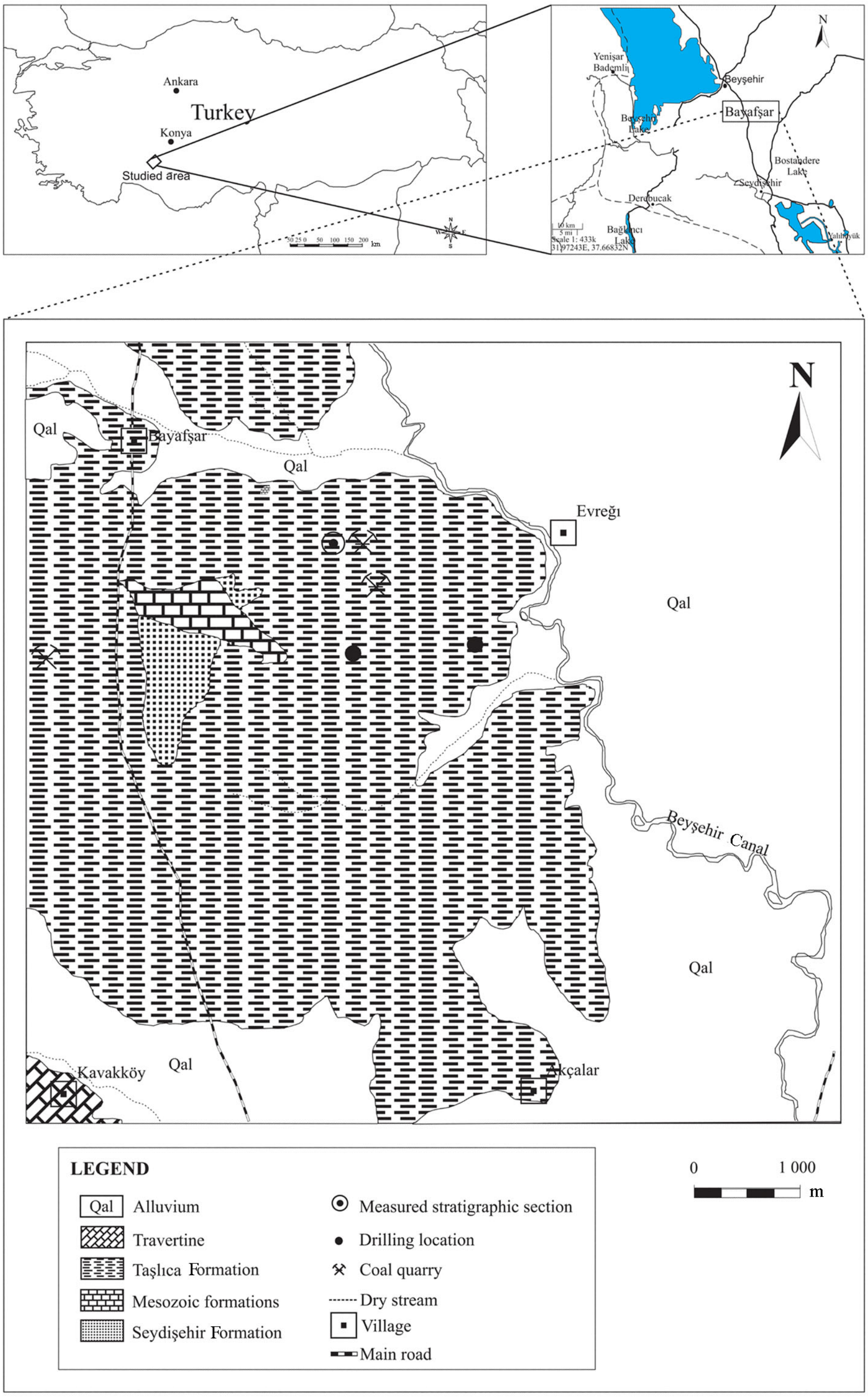

Fig. 1. Location and geological map of investigated area and its surroundings (geological map modified from [22]).

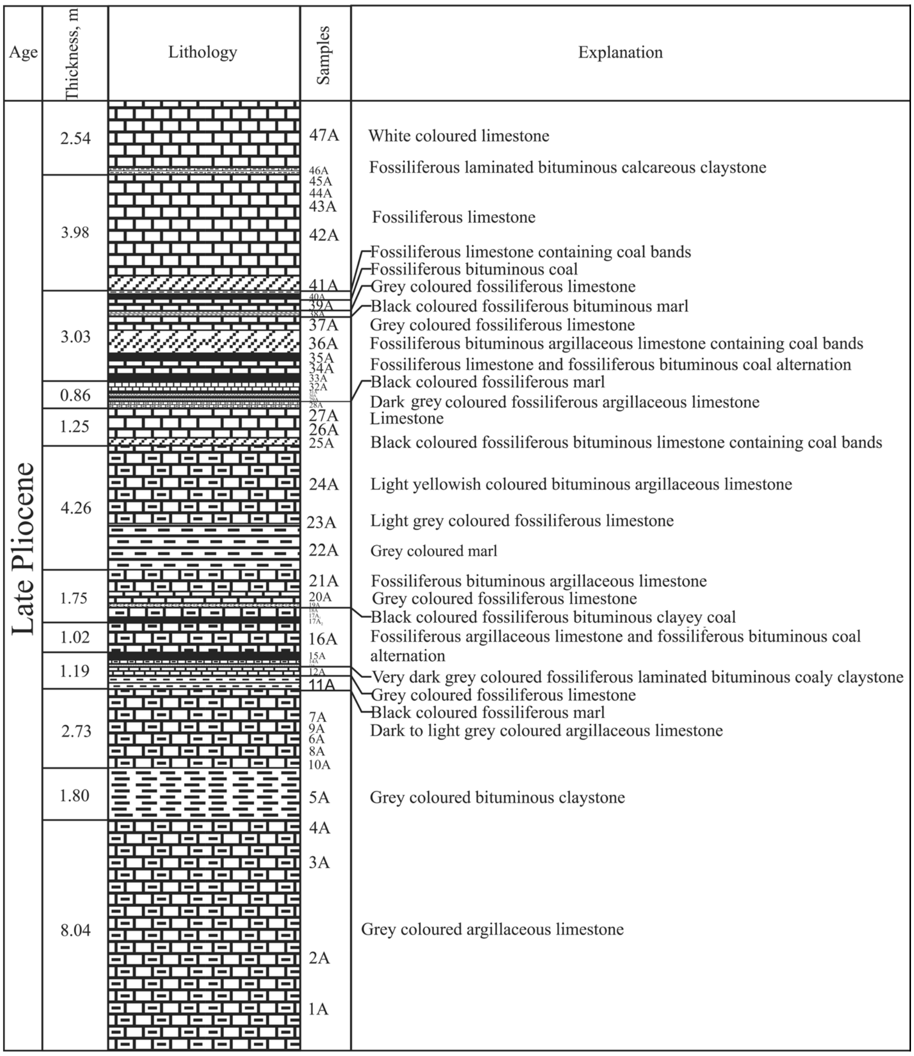

Fig. 2. The measured stratigraphic section through the Taşlıca formation showing the sample positions.

Within the fossiliferous lithologies of the Taşlıca Formation in the area under study, fossils of *Valvata piscinalis (O.F. Müller), Valvata pulchella Studer, Lithoglyphus acutus decipiens Brusina, Bithyinia tentaculata (Linnaeus), Acella acuaria Neumayr, Planorbarius corneus (linnaeus), Planorbarius thiollierei (Michaud), Anisus vortex (linnaeus), Gyraulus albus (O.F. Müller) and Segmentina filocincta (Sandberger)* were identified. The Taşlıca Formation in the area was dated as Late Pliocene based on its fossil assemblage.

Fig. 3. Levels of bituminous and fossilliferous coal (arrows direction) in the studied area.

Fig. 4. Sample of bituminous and fossilliferous coal.

3. Materials and methods

Altogether 48 samples from a measured section exposed in a quarry (Fig. 2) were collected from the Taşlıca Formation in the Akçalar lignite basin (Bayafşar village, Konya, central Turkey). Of them, 20 bituminous samples were subjected to TOC and Rock Eval pyrolysis analyses (Table and Fig. 5) in the Laboratory of Applied Petroleum Technology AS in Norway by following the instructions given in NIGOGA – The Norwegian Industry Guide to Organic Geochemical Analyses [23].

The fossil determination of 28 fossiliferous rock samples from the studied stratigraphic section has been realized by Dr. Sevinç Karan Yeşilyurt at the Paleontology Laboratory at Çanakkale 18 Mart University, Turkey.

4. Results

Abundance of organic matter, its type, thermal maturation and hydrocarbon potential of rock samples are typically assessed by Rock-Eval pyrolysis [24–27]. Results of Rock-Eval pyrolysis are summarized in Table and Figure 5.

The total organic carbon content of the studied samples varies between 0.4 and 50.0 wt% with an average of 21.35 wt%. Samples with TOC values greater than 10 wt% are considered as organic-rich [24], and as excellent [25] and qualified [28] source rocks. These high values of TOC are also viewed as a favorable environment for production and preservation of organic matter.

The Rock-Eval pyrolysis parameter S_1 values vary between 0.04 and 16.16 mg HC/g rock (average 5.26 mg HC/g rock) for bituminous rock samples, whereas the Rock-Eval pyrolysis parameter S_2 values for the

Table. Results of total organic carbon (TOC) and Rock-Eval pyrolysis analyses for the studied samples

Sample No.	TOC, wt%	S_1, mg HC/g rock	S_2, mg HC/g rock	S_3, mg CO_2/g rock	S_2/S_3	PY, mg HC/g rock	PY, ppm	PI	T_{max}, °C	HI, mg HC/g TOC	OI, mg CO_2/g TOC	RC, wt%	PC, wt%
1A	0.8	0.35	1.18	0.45	2.62	1.53	1530	0.23	431	149	57	0.67	0.13
2A	1.3	0.75	5.13	0.45	11.4	5.88	5880	0.13	431	395	35	0.81	0.49
5A	0.8	0.29	1.47	0.42	3.5	1.76	1760	0.16	433	181	52	0.65	0.15
7A	0.6	0.08	0.46	0.44	1.05	0.54	540	0.15	438	82	79	0.56	0.04
13A	39.3	6.7	53.08	18.92	2.81	59.78	59780	0.11	434	135	48	34.34	4.96
14A	8.2	1.15	13.59	3.32	4.09	14.74	14740	0.08	437	166	41	6.98	1.22
15A	33.9	6.33	43.1	31.42	1.37	49.43	49430	0.13	420	127	93	29.8	4.1
$17A_2$	35.1	5.1	43.21	12.51	3.45	48.31	48310	0.11	430	123	36	31.09	4.01
19A	34.4	7.98	80.2	12.9	6.22	88.18	88180	0.09	434	233	37	27.08	7.32
21A	24.5	4.79	45.78	20.17	2.27	50.57	50570	0.09	438	187	82	20.3	4.2
24A	0.5	0.07	0.25	0.45	0.56	0.32	320	0.22	478	49	88	0.47	0.03
25A	38	5.55	53.76	17.54	3.06	59.31	59310	0.09	440	141	46	33.08	4.92
31A	3.4	0.14	1.63	2.32	0.7	1.77	1770	0.08	441	48	68	3.25	0.15
33A	44.7	10.83	78.12	17.03	4.59	88.95	88950	0.12	430	175	38	37.32	7.38
35A	50	15.54	88.08	18.69	4.71	103.62	103620	0.15	408	176	37	41.4	8.6
36A	25	9.48	63.05	15.86	3.98	72.53	72530	0.13	422	252	63	18.98	6.02
38A	40.6	16.16	115.56	26.64	4.34	131.72	131720	0.12	415	285	66	29.67	10.93
40A	45	13.73	80.59	28.11	2.87	94.32	94320	0.15	412	179	62	37.17	7.83
44A	0.4	0.04	0.07	0.47	0.15	0.11	110	0.36	511	19	129	0.39	0.01
46A	0.5	0.04	0.08	0.47	0.17	0.12	120	0.33	505	17	102	0.49	0.01
Average	21.35	5.26	38.42	11.43	3.2	43.68	43674.5	0.15	439.4	155.95	62.95	17.73	3.63

Note: PC – pyrolyzable carbon.

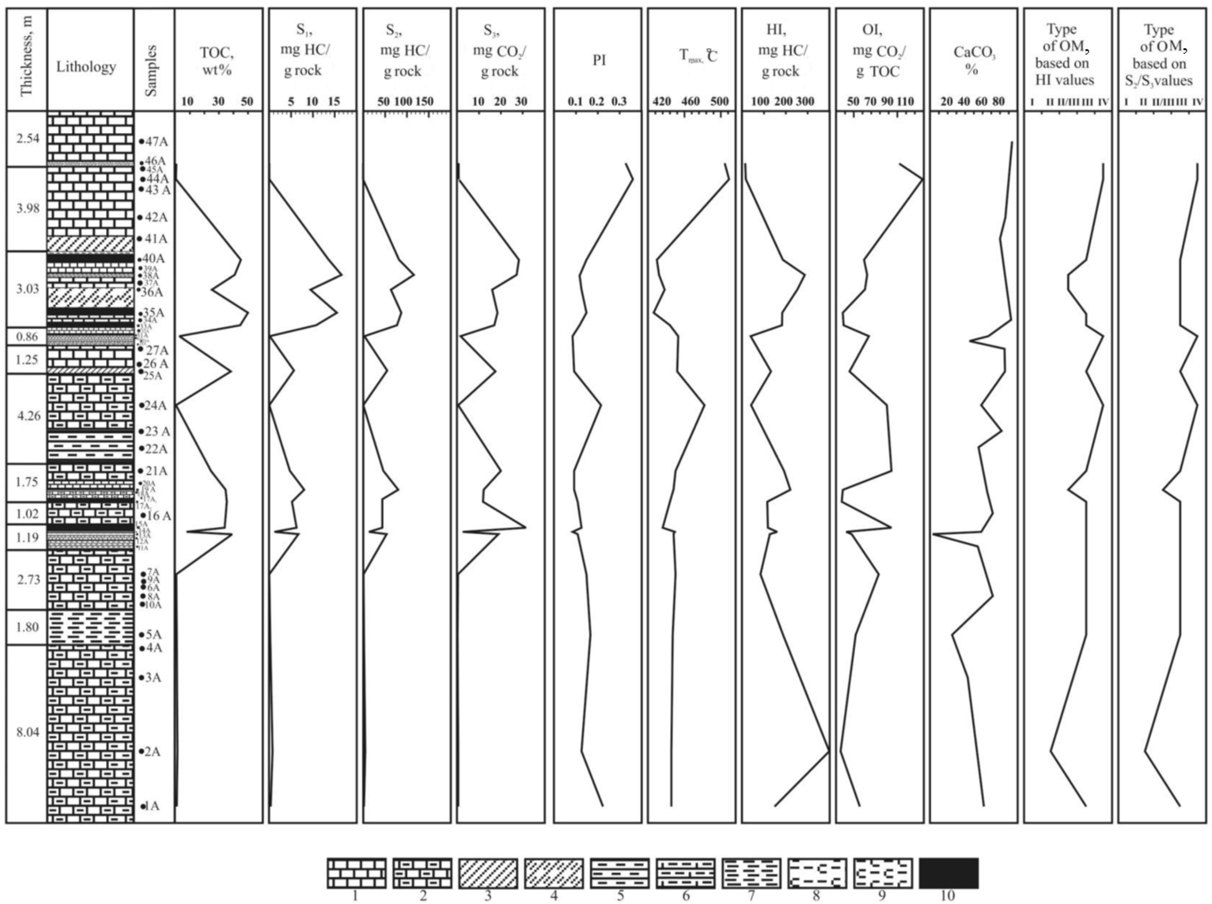

Fig. 5. Rock-Eval pyrolysis and carbonate percentage data for the studied samples. 1 – limestone; 2 – argillaceous limestone; 3 – limestone containing coal bands; 4 – argillaceous limestone containing coal bands; 5 – marl; 6 – calcareous claystone; 7 – claystone; 8 – coaly claystone; 9 – clayey coal; 10 – coal.

samples range from 0.07 to 115.56 mg HC/g rock (average 38.42 mg HC/g rock). The Hydrogen Index (HI) values of the studied samples are generally low, varying between 17 and 395 mg HC/g TOC (average 155.95 mg HC/g TOC), while the Oxygen Index (OI) values for the analyzed samples are relatively high, ranging from 35 to 129 mg CO_2/g TOC (average 62.95 mg CO_2/g TOC). The moderate to relatively high S_2, low HI and relatively high OI values indicate that the studied bituminous rock samples have a good source rock potential with a tendency to gas generation [25].

The hydrocarbon type index (S_2/S_3 ratio) is used in determining the kerogen type [25], generated hydrocarbon type (oil or gas) and thermal maturity level [29, 30]. Potential Yield or genetic potential (PY = $S_1 + S_2$) values are the reflection of hydrocarbon potential of a rock sample [24]. Finally, production index (PI = $S_1/(S_1 + S_2)$) ratio is used for maturity evaluation [25].

Generally, the studied samples have low S_2/S_3 ratio values, varying between 0.15 and 11.4 with an average of 3.2. PY values for the investigated bituminous rock samples are usually high, ranging from 110 to 131 720 ppm (average 43 674.5 ppm). Moreover, the samples, in general, are represented by relatively low PI values, varying between 0.08 and 0.36 (average 0.15).

The T_{max} values show the maturity level of organic matter [24, 31, 32]. The studied samples exhibit relatively low T_{max} values, ranging from 408 to 511 °C with an average of 439.4 °C.

5. Discussion

5.1. Depositional environment

The paleontological data show that the sequences in the studied area were deposited in a fresh water lacustrine environment with periodically changing water depth. The presence of *Valvata piscinalis* and *Valvata pulchella* in high quantities, and the relative abundance of *Lithoglyphus acutus decipiens* and *Bithyinia tentaculata* imply the dominance of a lacustrine environment fed by fresh water and rivers. *Acella acuaria, Anisus vortex, Segmentina filocincta* and *Gyraulus albus* indicate the existence of shallow, fresh and stagnant water conditions. At the same time, *Planorbarius corneus* and *Planorbarius thiollierei* which were found within coal levels give evidence of a shallow and stagnant swamp depositional environment.

5.2. Type of organic matter and generated hydrocarbon (oil or gas)

Kerogen types of the bituminous rock samples were determined using HI [25], S_2/S_3 ratio [25], HI-OI [26] and HI-T_{max} [33, 34] kerogen classification diagrams.

The low HI and S_2/S_3 ratio values (Table) for the studied samples indicate that kerogen is mostly of Type III with minor Type IV and Type II/III. On the HI-OI diagram, the investigated samples mainly fall in the Type II kerogen field and the rest of the samples plot in the Type III kerogen field (Fig. 6). Contrary to the HI-OI diagram, the HI-T_{max} diagram classifies the organic matter of the vast majority of the studied samples as Type III kerogen and that of a few samples as Type II kerogen (Fig. 7).

The organic matter classified mainly as Type III kerogen is interpreted as having a high gas generating potential with a minor existence of gas-oil mixture. Additionally, on the HI-TOC [35] diagram, the studied samples mostly plot in the gas and/or oil and gas field (Fig. 8).

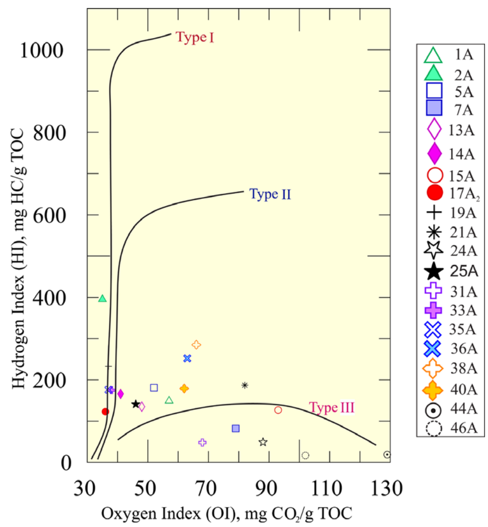

Fig. 6. HI vs OI plot for the studied bituminous rock samples.

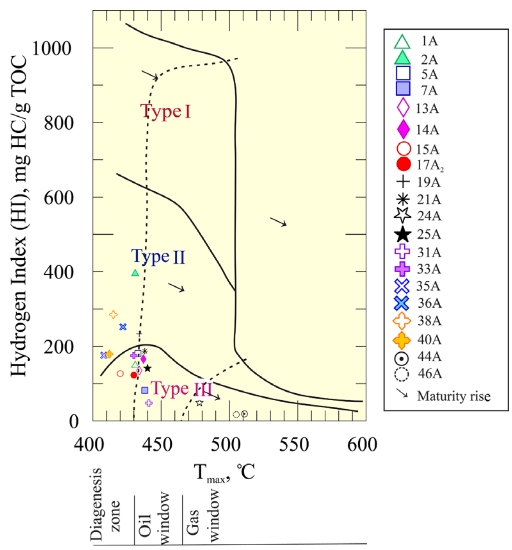

Fig. 7. HI vs T_{max} plot for the studied bituminous rock samples.

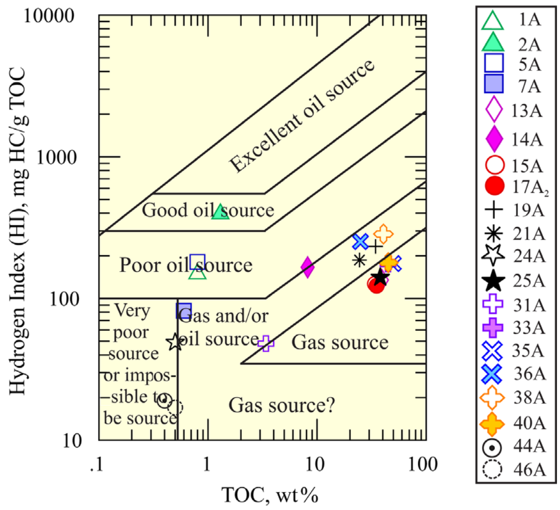

Fig. 8. Configuration of bituminous rock samples selected from the studied area on the HI-TOC cross-plot.

5.3. Thermal maturity of organic matter

The T_{max} value measured during the pyrolysis and the calculated PI values are closely related to thermal history of organic matter [24, 36]. Therefore, in this work the maturity of kerogen is interpreted using the T_{max}, PI and S_2/S_3 ratio values obtained from pyrolysis analysis.

The calculated T_{max} values (Table), according to [31], indicate that the studied samples mostly range from immature to oil prone and a few samples are gas prone. According to [25], T_{max} values are indicative of immature to early mature organic matter and a small part of samples is at the post mature thermal level. On the other hand, based on [32], these values show that the vast majority of the investigated samples are characterized by immature to early mature thermal level and the rest of the samples show oil and gas prone characteristics. The PI values (Table) of the studied bituminous rock samples are relatively low, and, based on [25], for most of the samples these values correspond to the early mature character and for the rest, to the immature and mature character. Moreover, the low S_2/S_3 ratios (Table) of the studied samples, according to [29, 30], reflect the catagenetic-post mature character for most of them, whereas a few samples show catagenetic-mature characteristics.

Additionally, HI-T_{max} [33, 34] and PI-T_{max} [37] diagrams have also been used to assess the maturity of organic matter. Both HI-T_{max} (Fig. 7) and PI-T_{max} (Fig. 9) diagrams exhibit that the investigated samples, in general, have clustered in the immature (diagenesis) zone and marginally fall in the beginning of the oil zone.

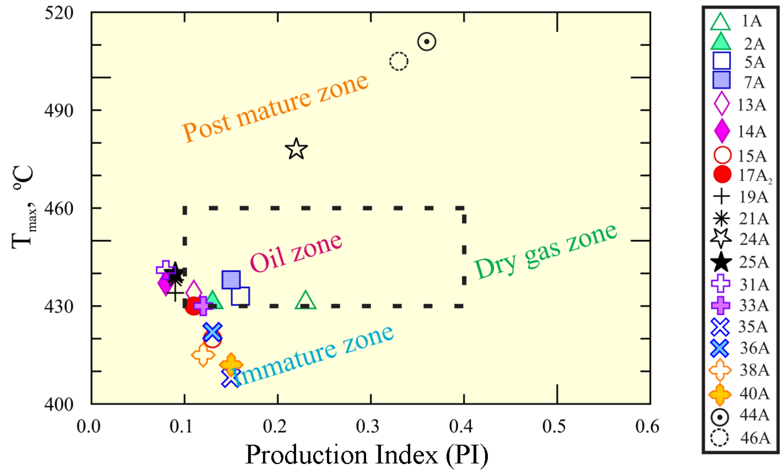

Fig. 9. Configuration of the studied samples on the PI-T_{max} cross-plot.

5.4. Potential of hydrocarbon generation

The values of Rock-Eval pyrolysis parameters S_1 and S_2 (Table) [25] for the studied bituminous rock samples suggest variation from poor to excellent hydrocarbon potential. In terms of Potential Yield values [24] (Table), the samples are characterized by variation from no oil source rock to good source rock potential. On the diagram of TOC vs S_2 [38], they plot in the fields indicating respectively poor, fair, good, very good and excellent source rock potential (Fig. 10). However, on the TOC-RC (Residual carbon) [39] diagram, the studied samples mainly clustered very near to the residual carbon line (TOC = RC), indicating poor source rock potential (Fig. 11).

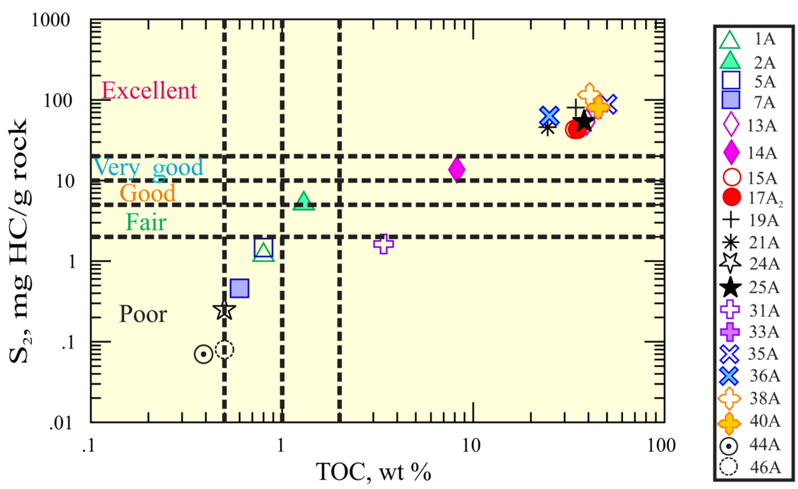

Fig. 10. Source rock potential for the studied samples based on the S_2-TOC diagram.

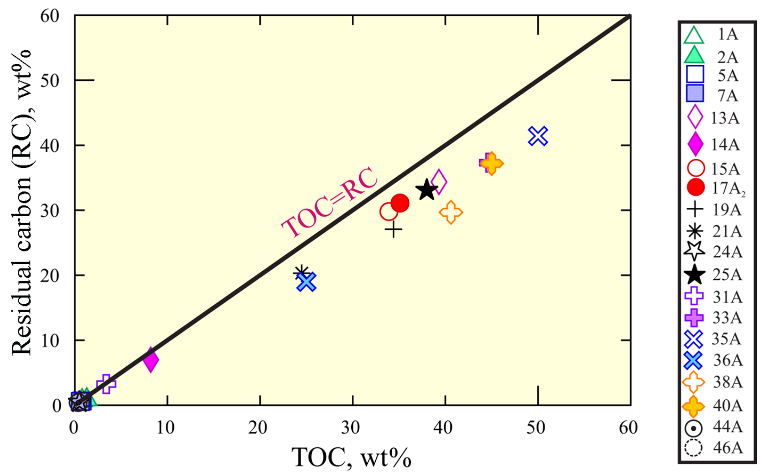

Fig. 11. Distribution of the studied samples on the RC-TOC plot.

5.5. Migrated hydrocarbons

Nonindigenous hydrocarbons can be detected if S_1 is high and TOC low [33]. Therefore, TOC vs S_1 diagram was used to separate migrated hydrocarbons from non-migrated ones in the bituminous rock samples from the studied area (Fig. 12). Thus, all the analyzed samples plot in the indigenous hydrocarbons field, reflecting no external contribution of migrated hydrocarbons to the bituminous rocks of the area.

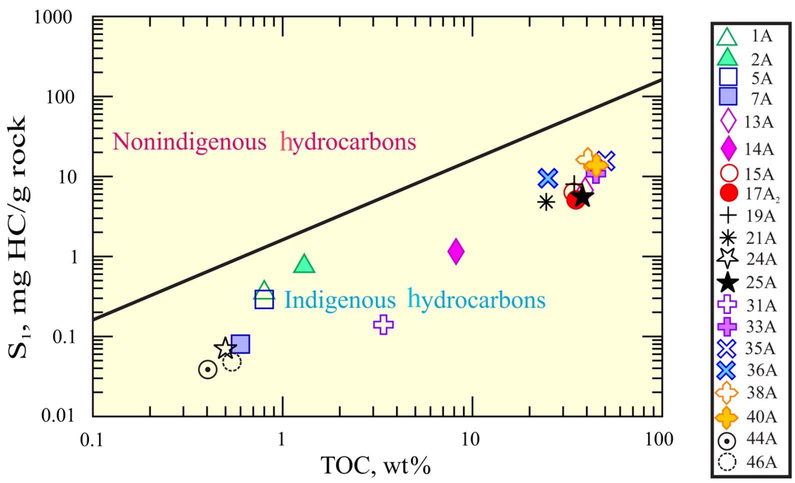

Fig.12. Distribution of the analyzed bituminous rock samples on the S_1-TOC plot.

6. Conclusions

Twenty bituminous rock samples from the measured stratigraphic section in the Taşlıca Formation in the Akçalar lignite basin were investigated with regard to their organic geochemical characteristics. Additionally, the palaeontological study was conducted on twenty-eight fossiliferous samples taken from the same section to interpret the depositional environment.

The TOC values, with an average of 21.35 wt%, show the organic matter content of the studied samples to be very high.

HI (17–395 mg HC/g TOC) and S_2/S_3 (0.15–11.4) ratio values imply that the samples contain mostly Type III kerogen with minor Type IV and Type II/III kerogens. The HI-OI diagram shows that kerogen type is mainly II, while the HI-T_{max} diagram classifies the kerogen type for the majority of the studied samples as III.

In terms of generated hydrocarbon type, HI and S_2/S_3 ratio values mostly indicate a high gas generating potential with a minor existence of gas-oil mixture. On the HI-TOC diagram, the studied samples mainly fall in the gas and/or oil and gas field. With regard to the thermal maturity of organic matter of the samples, T_{max} values in the range from 408 to 511 °C generally indicate immature to oil prone stage, and in some cases, immature to early mature stage. PI values of 0.08–0.36 reflect the early mature character, whereas S_2/S_3 ratio values show the catagenetic-post mature character for most studied samples and the rest exhibit catagenetic-mature characteristics.

Moreover, plotted on HI-T_{max} and PI-T_{max} diagrams, the studied samples fall in the immature (diagenesis) zone and marginally enter the beginning of the oil zone.

S_1 (0.04–16.16 mg HC/g rock), S_2 (0.07–115.56 mg HC/g rock), PY (110 to 131720 ppm) values and TOC vs S_2 diagram used to assess the hydrocarbon potential suggest its variation from poor to excellent. However, the TOC-RC (Residual carbon) diagram exhibits poor source rock potential. On the TOC vs S_1 diagram, all the analyzed samples concentrated in the indigenous hydrocarbons field signifying that the source rock had not been subjected to any external contamination.

The paleontological data show that the sequences in the studied area were deposited in a fresh water lacustrine environment with periodically changing depositional conditions from deep to shallow and to stagnant swamp.

Acknowledgements

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