Biomarker Characteristics of Crude Oil Blends from Some Flow-Stations in Bayelsa State, Nigeria

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Authors' contributions

This work was carried out in collaboration between all authors. All authors read and approved the final manuscript.

Original Research Article

ABSTRACT

Three different crude oil samples from three crude oil flow stations in Bayelsa state, Nigeria, were chosen for this study and were used to analyze the geochemical characteristics such as thermal maturity, depositional environments, sources of organic matter and extent of biodegradation. The crude oil samples were separated into saturated hydrocarbons, aromatic hydrocarbons and polar compounds. The saturated hydrocarbons were determined by Agilent (HP) 5890 Series II gas chromatography equipped with a Flame Ionization Detector (California, USA). Ratios of the biomarkers pristane/phytane, Isoprenoids/n-alkanes and CPI were determined. The pristane/phytane ratio in the oils reflects that the oils originated mainly from a source rock with a significant terrestrial contribution deposited under oxic environment with high maturation level due to the high pr/ph ratio of the oils. The ph/n-C_{18} ratios of the oils were less than one (< 1.0) suggesting that the oils were non-biodegraded. The CPI values of the three crude oil samples were below 1.0 indicating that the crude oil samples can be described as matured.

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1. INTRODUCTION

The origin of petroleum lies in plants and animals which lived on earth and in sea many millions of years ago. These organisms died and their remains became buried under the earth. Due to the bacterial decomposition and under the action of earth's heat and pressure, these remains were converted to liquid hydrocarbon, petroleum and other compounds. Today petroleum and natural gas rank as the main sources of our fuel and the bulk of organic chemicals used in industries.

The Niger Delta is a region that is rich in crude oil, is also significant in crude oil correlation studies because it is one of the major hydrocarbon provinces of the world with an estimated reserve of about 25 billion barrels of oil and 256 trillion cubic feet of natural gas [1].

Crude oil, a non-renewable source of energy is a complex naturally occurring mixture containing mostly hydrocarbons with varying proportions of non-hydrocarbon constituents and traces of organometallic compounds. Most of the hydrocarbon found in crude oil ranges from C_1 to C_{60+} and are arranged in chains or rings. Crude oil may also contain some compounds of oxygen, nitrogen and sulfur. In general, crude oil has an average composition of 85% carbon, 13% hydrogen and 2% of sulphur, nitrogen and oxygen. The petroleum occurs in sedimentary rock deposits throughout the world. Crude oil also contain numerous "chemical fossils" or "biomarker molecules" which are resistant to biodegradation and whose origin in the crude oil is related through transformation to organic molecules produced by living organism [2].

The name geochemistry was first used by the Swiss chemist Christian Schobein in 1838. He mapped out the programmme for this new branch of science and emphasized the importance of studying in detail the properties of all geological formation, their physical and chemical properties and their age relationship, thus geochemistry can be defined as the science concerned with the chemistry of the earth as a whole and of its component parts [3].

Geochemistry is helpful for determining the relative and absolute abundance of chemical components in the earth of which Characterization of crude oils is a routine part of petroleum geochemistry. Various crude oil classifications have been proposed by geochemist and petroleum refiners, but the most important area of interest to the geochemist and geologists is in identifying and characterizing the crude oils, in order to relate them to specific source rocks and also to know the degree of their thermal maturation [1]. Due to the variety of geological conditions and age under which crude oil was formed, every crude oil exhibits a unique biomarker fingerprint depending on the oil source, the thermal regime during oil generation, the geological migration and the reservoir conditions [4].

Geochemical methods of crude oil characterization include biomarker finger prints, use of bulk parameters, stable isotope ratios, hydrocarbon contents, etc. [5-8]. Biomarker analysis mainly relies on the use of gas chromatography- mass spectrometry (GC-MS).

Biomarkers are compounds found in oil that holds information of the oil. They are mainly from living organisms that have been preserved through time. Petroleum biomarkers are complex organic compounds derived from formally living organism found in oil [9]. They show little or no changes in their structures from the parent organic molecules and this distinguishes biomarker from other compounds [10]. These compounds are also called geochemical fossils because of their origin from living organisms [11]. They are also the 'DNA' of oils providing information not only on the biological source of the oil, but also on the environment in which the parent organic matter was deposited and on the thermal history of the buried organic matter [12].

The biomarker patterns of crude oils provide information on the organic matter type, environmental conditions during deposition, thermal maturity experienced by a source rock and the degree of biodegradation [8,13]. In analytical chemistry, biomarkers are also used to trace spilled, weathered and biodegraded petroleum pollution in the environment. The information from biomarkers analysis can be used also to determine the migration pathways from a source rock to the reservoir through oil-to-oil and oil-to-source rock correlation [4].

Several hundreds of biomarkers have been identified in crude oils and sediments. Amongst these biomarkers, the most frequently studied are the n –alkanes, isoprenoid, porphyrins and steranes. The two best known and abundant biomarkers in petroleum are the isoprenoid molecules; pristane and phytane.

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A variety of studies have been carried out in the Niger Delta in order to determine the location and effectiveness of the region source rocks [14,15]. But despite these in-depth studies, no consensus has yet emerged concerning the true identity of the petroleum systems that contributed to the greater Niger Delta oil field [16].

Onojake et al. [17], recognized that the crude oils in Southwest Niger Delta are formed from mixed sources (marine and terrestrial Kerogen) deposited in an oxic paleoenvironment with no particular maturity trend. In another study Onojake et al. [18], concluded that the crude oil from Umutu/Bomu fields, Southwest Niger Delta originated mainly from terrestrial organic sources deposited in an oxic paleoenvironment and are matured and non-biodegraded.

This work attempts to evaluate the organic geochemical characteristics of crude oils blends from three different flow stations within Bayelsa state, Nigeria, to: Characterize the oils; differentiate the source depositional environments; degree of thermal maturity and the degree of biodegradation.

2. MATERIALS AND METHODS

The materials and methodologies applied in the Sample collection, Fractionation of the Crude oils and the GC-FID analysis are as described in the subsections below.

2.1 Sample Collection

Three samples of blends crude oil were collected from three different Flow-stations in Bayelsa state, Nigeria. The crude oils were collected with glass vials with Teflon Caps, labeled appropriately and then transported to the laboratory for analysis. To avoid evaporative losses, of the lower hydrocarbons, the vials were filled with no air space and kept in a ice chest. The crude oil samples were collected from LNG Obunagha (AT-01), Osiama oil field (KD-02) and Etelebou Flow Station (ET-03).

2.2 Fractionation of the Crude Oils

The crude oil samples were fractionated into saturates, aromatic hydrocarbons and polar compounds by column chromatography on a silica gel. The standard glass column, which is 50 cm in length and 0.5 cm in internal diameter, was rinsed first with dichloromethane (DCM) and later with light petroleum spirit (petroleum ether).The column was then plugged with cotton wool, to serve as a resting pad for the stationary phase (silica gel (SiO_2)) and filled with petroleum ether. Two (2 g) of Alumina was added to keep the surface stable. A crude oil sample was introduced, followed by the eluents, gently.70 mL of petroleum ether was added to elute the aliphatic fraction and 70 mL of DCM was used to elute the aromatic fractions, while 70 mL of methanol was used to elute the polar fractions. The aliphatic fractions were reduced with nitrogen stream to near dryness and then diluted with DCM for GC-FID analysis. The aromatic and polar fractions were not used in the study. The procedure was done separately for the three crude oil samples.

2.3 Gas Chromatography-Flame Ionization Detector Analysis

The saturated hydrocarbon fractions (n-alkanes) obtained were analyzed using Agilent (HP) 5890 Series II gas chromatography equipped with a Flame ionization detector (California, USA). The column used was a silica capillary column (30 mx0.25 m i.d.0.25 film thickness) coated with methyl silicone (OV-1). One micro litre of the aliphatic fraction of each crude oil sample was injected in a splitless mode by means of a syringe through a rubber septum into the column. The Detector and Injector temperatures were kept at 320°C and 273°C respectively while oven temperature was programmed from 50°C to 320°C at 10°C /min, with an initial hold time of 8mins. Helium at a linear velocity of 2 mL/min was used as a carrier gas. This procedure was done in triplicates for each crude oil sample.

3. RESULTS AND DISCUSSION

The results obtained from the GC Analysis are stated in Table 1. Figs. 1-3 are representative chromatograms of the GC Analysis of the Crude oil samples. The n-alkanes from n-C_9 to n-C_{40} can be seen in the chromatograms. The lack of n-alkanes <n-C_8 in early retention windows of the chromatograms may be due to evaporative losses during sample processing.

The mean values of pr/ph, pr/n-C_{17}, ph/n-C_{18} and CPI obtained are stated in Table 2.

Table 1. Amounts of total hydrocarbon (C8-C40) compounds in the crude oil samples

Carbon range	AT-01						KD-02						ET-03					
	Retention time(min)	Amount (ppm) Replicate 1	Amount (ppm) Replicate 2	Amount (ppm) Replicate 3	Average (ppm)	Std. Dev.(ppm)	Retention Time(min)	Amount (ppm) Replicate 1	Amount (ppm) Replicate 2	Amount (ppm) Replicate 3	Average (ppm)	Std. Dev.(ppm)	Retention Time(min)	Amount (ppm) Replicate 1	Amount (ppm) Replicate 2	Amount (ppm) Replicate 3	Average (ppm)	Std. Dev.(ppm)
C-8	2.39	-	-	-	-	-	2.3	-	-	-	-	-	2.3	-	-	-	-	-
C-9	3.31	129.7	129.0	130.0	129.57	0.51	3.3	-	-	-	-	-	3.4	28.2	29.3	28.5	28.67	0.57
C-10	4.77	3.9	3.5	4.0	3.80	0.26	4.7	-	-	-	-	-	4.7	73.3	73..3	73.5	73.40	0.14
C-11	6.26	1.7	1.6	1.5	1.60	0.10	6.3	-	-	-	-	-	6.3	1.3	1.0	1.2	1.17	0.15
C-12	7.97	10.2	10.0	10.6	10.27	0.31	7.9	-	-	-	-	-	7.9	-	-	-		
C-13	9.42	165.4	164.2	165.2	164.93	0.64	9.4	81.4	80.0	81.7	81.03	0.91	9.3	642.5	642.1	641.3	641.97	0.61
C-14	10.8	2490.0	2489.1	2490.3	2489.80	0.62	10.7	361.7	361.5	360.5	361.23	0.64	10.6	2193.3	2193.0	2192.5	2192.93	0.40
C-15	12.0	1128.4	1129.0	1130.0	1129.13	0.81	11.9	5.2	5.1	5.0	5.10	0.10	12.0	159.0	158.5	160.0	159.17	0.76
C-16	13.3	505.0	504.2	505.5	504.90	0.66	13.1	362.3	362.3	361.7	362.10	0.35	13.3	4352.1	4352.3	4352.0	4352.13	0.15
C-17	14.4	501.0	501.3	502.0	501.43	0.51	14.3	51.0	49.8	50.2	50.33	0.61	14.3	26.1	26.5	26.3	26.30	0.20
Pristane	14.7	4822.9	4822.0	4823.5	4822.80	0.75	14.5	2458.2	2458.1	2457.1	2457.80	0.61	14.5	23729	24126	23391	23748.67	367.89
C-18	15.6	1559.9	1560.0	1559.0	1559.63	0.55	15.6	329.2	329.1	330.0	329.43	0.49	15.4	995.6	994.5	995.0	995.03	0.55
Phytane	15.7	841.3	839.1	840.2	840.20	1.10	15.6	328.2	328.1	329.1	328.47	0.55	15.6	140.5	140.0	140.8	140.43	0.40
C-19	16.6	1041.2	1041.1	1040.0	1040.77	0.67	16.5	530.3	532.0	531.2	531.17	0.85	16.5	926.6	926.3	926.0	926.30	0.30
C-20	17.5	839.6	840.2	839.2	839.67	0.50	17.5	503.2	502.2	503.0	502.80	0.53	17.5	893.2	893.3	892.8	893.10	0.26
C-21	18.5	501.8	500.2	501.5	501.17	0.85	18.4	253.0	253.5	252.2	252.90	0.66	18.4	71.6	70.7	70.9	71.07	0.47
C-22	19.4	282.6	283.1	282.1	282.60	0.50	19.4	476.8	477.2	477.4	477.13	0.31	19.2	881.0	881.0	882.3	881.43	0.75
C-23	20.3	196.7	196.6	195.4	196.23	0.72	20.3	149.0	150.0	149.3	149.43	0.51	20.3	386.0	385.5	386.5	386.00	0.50
C-24	21.1	101.8	101.8	100.6	101.40	0.69	21.0	802.0	801.3	802.5	801.93	0.60	21.1	3022.3	3021.0	3022.7	3022.00	0.89
C-25	21.5	39.7	38.4	40.0	39.37	0.85	21.9	825.6	825.9	826.0	825.83	0.21	21.9	2494.6	2494.0	2493.6	2494.07	0.50
C-26	22.7	-	-	-	-	-	22.6	80.7	80.4	80.0	80.37	0.35	22.7	1.3	1.5	1.4	1.40	0.10
C-27	23.4	-	-	-	-	-	23.3	7.7	8.0	7.5	7.73	0.25	23.3	70.1	70.5	70.3	70.30	0.20
C-28	24.1	-	-	-	-	-	23.6	152.7	151.6	152.2	152.17	0.55	24.1	274.6	273.5	274.2	274.10	0.56
C-29	24.8	-	-	-	-	-	24.8	-	-	-	-	-	24.9	166.5	164.6	165.1	165.40	0.98
C-30	25.5	-	-	-	-	-	25.4	-	-	-	-	-	25.5	149.2	150.4	149.6	149.73	0.61
C-31	26.1	-	-	-	-	-	26.1	-	-	-	-	-	26.1	156.1	156.2	156.0	156.10	0.10
C-32	26.8	-	-	-	-	-	26.7	-	-	-	-	-	26.7	226.2	227.0	226.2	226.47	0.46

| Carbon range	AT-01						KD-02						ET-03					
	Retention time(min)	Amount (ppm) Replicate 1	Amount (ppm) Replicate 2	Amount (ppm) Replicate 3	Average (ppm)	Std. Dev.(ppm)	Retention Time(min)	Amount (ppm) Replicate 1	Amount (ppm) Replicate 2	Amount (ppm) Replicate 3	Average (ppm)	Std. Dev.(ppm)	Retention Time(min)	Amount (ppm) Replicate 1	Amount (ppm) Replicate 2	Amount (ppm) Replicate 3	Average (ppm)	Std. Dev.(ppm)
C-33	27.4	-	-	-	-	-	27.3	-	-	-	-	-	27.3	494.1	493.2	494.2	493.83	0.55
C-34	27.9	-	-	-	-	-	27.9	-	-	-	-	-	27.9	356.0	356.7	356.7	356.47	0.40
C-35	28.7	3.8	3.6	3.3	3.57	0.25	28.5	86.1	87.0	86.3	86.47	0.47	28.6	764.0	764.5	764.0	764.17	0.29
C-36	29.2	477.0	476.3	477.2	476.83	0.47	29.2	18.8	17.7	18.7	18.40	0.61	29.2	439.1	437.2	439.2	438.50	1.13
C-37	29.9	876.2	876.0	875.7	875.97	0.25	29.6	767.7	767.5	767.2	767.47	0.25	30.0	3293.9	3292.0	3293.1	3293.00	0.95
C-38	30.4	1227.1	1227.0	1226.0	1226.70	0.61	30.6	1162.0	1162.3	1161.0	1161.77	0.68	30.5	2817.5	2817.2	2817.0	2817.23	0.25
C-39	31.5	1330.0	1331.1	1330.3	1330.47	0.57	31.7	1207.4	1207.0	1206.5	1206.97	0.45	31.5	2615.9	2613.2	2614.5	2614.53	1.35
C-40	32.6	1173.2	1173.4	1173.0	1173.20	0.20	32.2	887.5	888.7	887.2	887.80	0.79	32.5	2251.2	2251.7	2251.0	2251.30	0.36
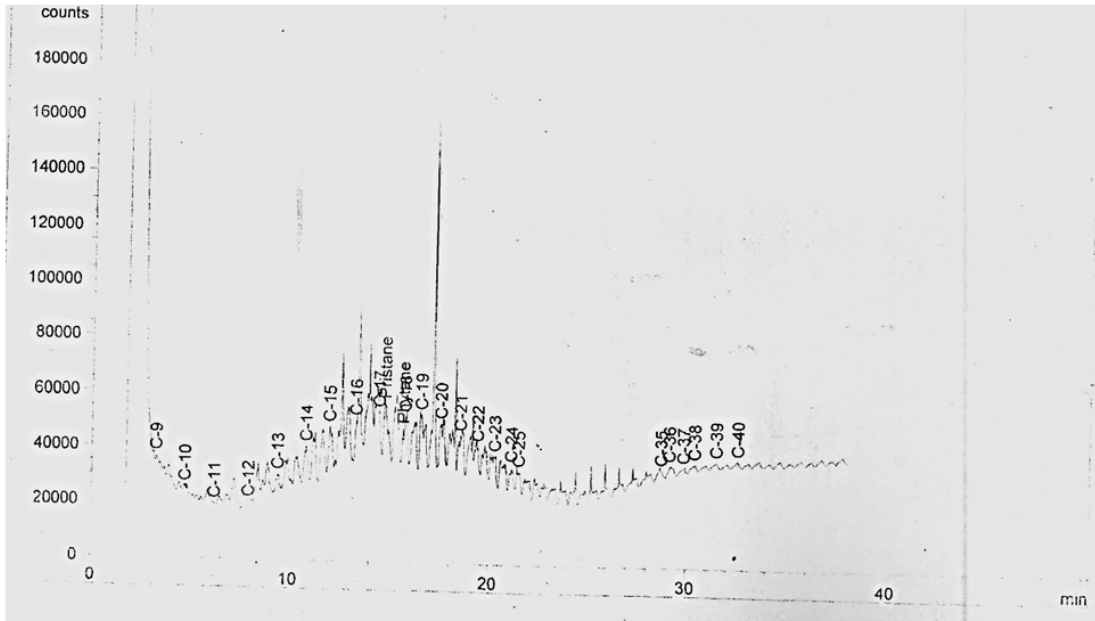
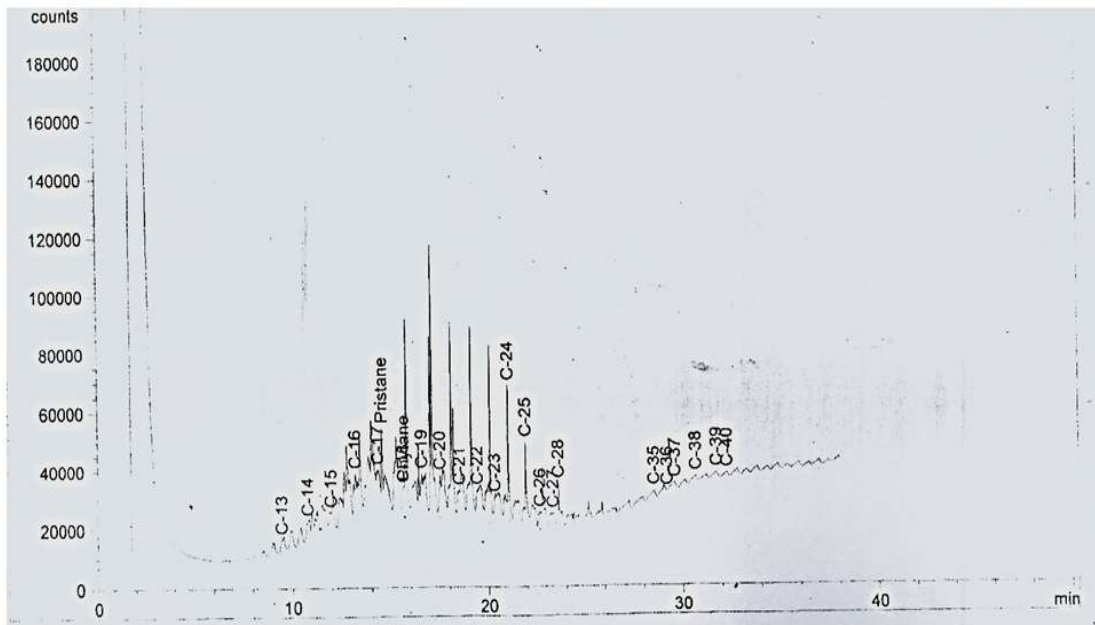**Fig. 1. Representative chromatogram of sample AT-01**

Fig. 2. Representative chromatogram of sample KD-02

3.1 Pristane and Phytane

Both pristane and phytane are derived from the phytol side chain of chlorophyll, either under reducing conditions to give phytane or oxidizing conditions to give pristane. The ratio of the pristane to phytane is used to assess redox conditions of the sediments during diagenesis [19]. The pristane/phytane ratio is one of the most commonly used correlation parameters which have been used as an indicator of depositional environment [20,21].
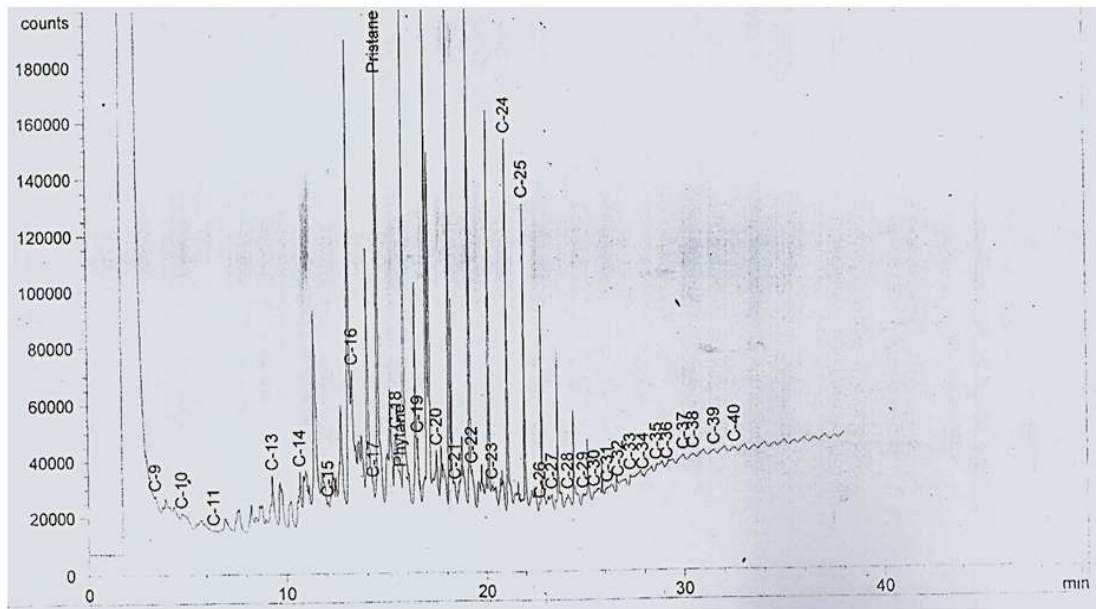
Fig. 3. Representative chromatogram of sample ET-03

Table 2. Geochemical parameter derived from GC-FID analysis

Sample	pr/ph	pr/n-C_{17}	ph/n-C_{18}	CPI
AT-01	5.74	9.62	0.54	0.68
KD-02	7.48	48.83	0.10	0.77
ET-03	64.55	902.99	0.14	0.06

Generally pristane/phytane ratio greater than 3 is thought to be associated with terrigenous bed with oxidizing conditions [22]. ten Haven et al. [23], stressed that high pr/ph (>3.0) indicates terrrigenous input under oxic conditions and low pr/ph (<0.8) indicates anoxic /hypersaline or carbonate environments. According to Volkman and Maxwell [24], pristane/phytane ratio >3 indicates terrestrial organic matter input under oxic conditions. Values less than 0.60 show anoxic, commonly hypersaline environments. Very high ratios (more than 3) are associated with terrestrial sediments. Pr/ph ratios ranging between 1 and 3 reflect oxidizing depositional environment [25]. According to Lijmbach [26], low values (pr/ph<2) indicate aquatic depositional environments including marine, fresh and brackish water (reducing conditions), intermediate values (2-4) indicate fluviomarine and coastal swamp environments, whereas high values (up to 10) are related to peat swamp depositional environments (oxidizing conditions). The ratios of the pristane/ phytane for the studied oil samples were generally high and ranged from 5.74 to 64.55 (Table 2). The pr/ph ratio for samples AT-01 (5.74), KD-02 (7.48) and ET-03 (64.55) suggest that the oils are derived from

terrestrial organic input under oxic conditions [23,24]. The high pr/ph ratios also indicate that the source rock for these oils may have been deposited in oxic environment and where are rich in terrestrial organic matter [3]. The relatively high pr/ph ratios of the oils indicate their high maturation [17].

3.2 Normal Alkanes and Isoprenoid Distribution

In crude oil correlations, the ratios of isopreniods to n-alkanes are widely used since they provide information on maturation and biodegradation as well as source [27,28]. Source, maturation, migration and biodegradation are the major factors responsible for the differences in crude oil composition [17]. Accordingly, isoprenoid/n-alkanes (pr/n-C_{17} and ph/n-C_{18}) ratios provide valuable information on biodegradation, maturation and diagenetic conditions. Generally pr/n-C_{17} and ph/n-C_{18} ratios decrease with increasing thermal maturity [18]. Waples [29], stated that by increasing maturity, n-alkanes are generated faster than isoprenoid in contrast to biodegradation. The pr/n-C_{17} and ph/n-C_{18} ratios of isoprenoid to n-alkane are as computed Table

2. According to Joshi and Pandey [3], high pr/n-C_{17} ratios indicate that the source rock is deposited in an oxic environment. Ph/n-C_{18} values less than 1.0 are indicative of non-biodegraded oils. From the results (Table 2), pr/n-C_{17} ratios range from 9.62 to 902.99, while ph/n-C_{18} ratios range from 0.10 to 0.54. All samples recorded ph/n-C_{18} less than one (<1.0), suggesting that these samples were non-biodegraded [17,25]. The high pr/n-C_{17} for the samples suggests that the source rock for these oils were deposited in an oxic environment and were rich in terrestrial organic matter [3].

3.3 Carbon Preference Index (CPI)

CPI is used for the measure of strength of the odd-carbon predominance in n-alkanes [22]. The CPI was the first maturity indicator applied to crude oils [20]. CPI is affected by both source and maturity of crude oils [30]. If the number of odd and even carbon members is equal the CPI is 1.0. If odd carbon homologs, predominate the CPI is greater than 1.0 and vice versa. Some researchers observed that immature rocks often had high CPI values (> 1.5), whereas those of mature oils were almost always below 1.0 [17]. According to Peters et al. [20], CPI around and below 1.0 indicates mature crude oils. The studied crude oil samples are characterized by CPI ratios of 0.68, 0.77 and 0.06 for AT-01, KD-02 and ET-03 respectively (Table 2). The CPI values of the studied oil samples were below 1.0, indicating that the oils are mature.

4. CONCLUSION

Biomarkers play an important role in the classification of petroleum oils with respect to depositional environment, organic matter input, thermal maturity and extent of biodegradation. It can be concluded that from the results obtained in the study, that the crude oils were non-biodegraded, the analysis also reveals that the studied oil samples, originated mainly from terrestrial organic source deposited in an oxic environment. The calculated carbon preference indices for the crude oils, which were used in determining the maturity level of the crude oils, show that the studied crude oils were matured.

COMPETING INTERESTS

Authors have declared that no competing interests exist.

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