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Evaluation of Yemen Crude Oil in Relation to its Maturity and heavy Metals Magnitude

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ABSTRACT

Trace metals in Yemen crude oils have been investigated; The samples of Yemen crude oils were collected and digested by a dry digestion method and analyzed by ICP-Optical emission Spectrometer technique and Universal of petroleum (UOP-915-92) used to determine the distribution of normal alkane and analyzed by gas chromatography technique. The levels of most of the trace elements obtained in this study were generally low. The relatively higher levels of Ni in Malik, 32, 43 (3.942, 4.058, 4.005) and V (4.919, 3.582, 3.633) respectively, Fe in Jannah, Alnaser and Malik, 94.3, 92.7 and 33.45 respectively. Ni, V and Fe observed in some results should be expected because these metals are commonly associated with crude oil of Marine organic materials with no signs of biodegradation. Carbon preference index (CPI) of Yemen oils ranges between (1.04-1.1) which indicate Yemen crude oil maturity. In this study, the analysis of crude oils shows a slightly lower abundance of C_{28} and C_{29} and relatively higher concentrations of C_{27} steranes indicate more input of marine organic source. In this study is <2 (0.4037 - 1.6585) which indicates that the Yemen crude oil was derived from marine sources with increasing maturity. Ph/n-C18 (0.4484-0.4921) values less than 1.0 are indicative of non-biodegraded; the decrease of both Pr/n-C_{17} and Ph/n-C_{18} indicates maturation of the Yemen crude oils. The V/Ni index decreases with the age of oil so Yemen crude oils are young age because V/Ni less than

INTRODUCTION

Heavy metals are often found to be part of a crude oil sample (Fausnaugh, 2002). Possible sources of trace metals in crude oil are: through incorporation and diagnosis of metal complexes of the original biological materials; through incorporation into the organic matrix during diagnosis of the biological materials in the source rocks either from clay minerals or interstitial aqueous solution through an aqueous phase during primary and secondary migration and from formation waters or reservoirs' rock minerals (Fausnaugh, 2002). According to (Odebunmi and Adeniyi, 2007), the ever-increasing chemical utilization of crude oils and petroleum products call for a better knowledge of the composition, structure and properties of their fractions. Parameters often determined in crude oil include: Metal/mineral contents. These important parameters are used in the specification and classification of crude oil blends (Oyekunle and Famakin, 2004).

OBJECTIVES:

To check the heavy metals in some Yemeni crude oils and to investigate the age and maturity of that oils.

MATERIALS AND METHODS

Sample collection: Collection and analysis of oil samples were carried out in line with recommended procedures of the American Society of Testing and Materials (ASTM). The studied area is Yemen from three governorates Marib, Shabowh and Hadramout. It consist of three principal sectors, Marib-Aljowf, Jannah, and Almaasilah many oil sectors discovered in Yemen about one hundred and five in 2014, but fourteen sectors only enter production at the end 2014. Twelve crude oil samples were collected from various Yemen crude oil sectors. Sample collection was carried out in collaboration with field technicians from the wellheads of the various producing wells; the oil wells sampled include: S-2, 34, Jannah, Alnaser, Malik, 32, 43, Al-Masilah, 51, 10, Dabah and Safer. During sampling all bottles were rinsed with water and air-dried. The bottles were later rinsed with the crude oil to be sampled before the sample for analysis was collected. The oil samples were coded samples according to the sector name in (March, 2014).

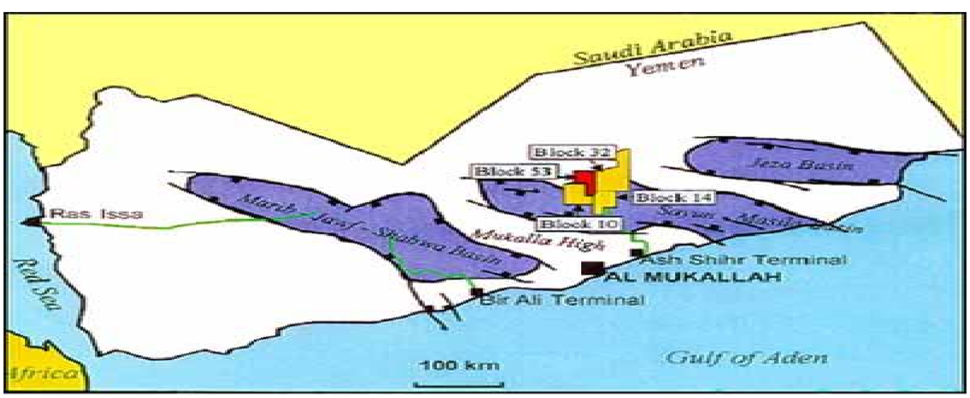

Figure 1: illustrate major sector and transportation pipeline crude oils in Yemen

Determination of the Metal content

Apparatus

* Inductively Coupled Plasma Atomic Emission Varian.
* Specimen Solution Containers, glass and plastic vials.
* Glassware, borosilicate 250-mL beakers, volumetric flasks of various capacities, and pipettes of various capacities. All glassware was cleaned and rinsed with deionized water.
* Electric Muffle Furnace, capable of maintaining 550°C and sufficiently large to accommodate 400-mL beakers.
* Hot plate capable of maintaining 300°C.

Reagents

Aqueous Standard Solutions, individual aqueous standards with 1000 mg/l concentrations of vanadium, nickel, iron and other elements.

H_2SO_4 concentrated and nitric acid HNO_3.

Summary of Test Method

10 g of sample was weighed into a beaker and decomposed with concentrated sulfuric acid. The residual carbon was burned off by heating at 525°C in a muffle furnace. The inorganic residue was digested with nitric acid, and made up to 50 ml volume. The solution was nebulized into the plasma of an atomic emission spectrometer. The intensities of light emitted at characteristic wavelengths of the metals were measured simultaneously. These intensities are related to concentrations by the appropriate use of calibration data.

Whole – Oil, Gas Chromatography Analysis

The Crude oil samples were subjected to whole oil- Gas chromatographic analysis. This was achieved by using Varian CP 3800 Gas Chromatograph, equipped with Flame Ionization Detector; 50m × 0.2 mm film thickness 0.5µm fused silica capillary PONA column. The sample (1µL) was injected. Injector temperature was kept at 280°C and (FID) detector at 320°C. The oven temperature was programmed from 60°C (Held 5 min) to 150°C at the rate 10°C /min, then to 300°C at rate 20°C/min (Held 60 min) finally to 310 at rate 20°C/min (Held 15 min). Helium flow 1.5 ml/min was used as carrier gas. The data were collected from retention time: 0-97 minutes.

Results

Table (1) shows the levels of heavy metals present in the studied Yemen crude oil. The results show range of values for physicochemical concentrations obtained from the oil samples as follows: For heavy metals, the range of values were Ag (0.0035-0.0846), Ca (3.24-1876), Cd (<0.0009-0.0162), Co (<0.0018-0.1632), Cr (<0.0013-1.776), Fe (<0.0062-94.3), K (<0.0360-17.52), Na (<0.0360-319.4), Ni (0.1649-4.005), Pb (0.015-402.2), V (0.0531-4.919) respectively.

Table 1: Parameter of heavy metals of Yemen crude oils

	S-2	34	Jannah	Alnaser	Malik	32	43	Almasilah	51	10	Dabah	Safer
Ag	<0.0035	<0.0035	0.0614	0.0437	0.0846	<0.0035	<0.0035	<0.0035	<0.0035	<0.0035	<0.0035	<0.0035
Ca	3.849	5.671	1876	1868	231	3.727	14.66	83.41	13.81	3.240	9.863	6.900
Cd	0.0041	<0.0009	0.0195	0.0162	<0.0009	0.0047	<0.0009	<0.0009	0.0092	<0.0009	<0.0009	<0.0009
Co	<0.0018	<0.0008	0.1615	0.1632	0.0147	<0.0018	<0.0018	<0.0018	<0.0018	<0.0018	<0.0018	0.0686
Cr	<0.0013	<0.0013	1.776	1.759	0.1619	<0.001	<0.0013	0.0018	<.0013	<0.0013	<0.0013	0.0084
Fe	0.6345	0.3801	94.3	92.7	33.35	1.1020	4.850	18.50	3.514	<0.0062	3.2	1.684
K	1.262	0.4209	4.169	5.164	17.52	<0.0360	0.1873	12.59	1.963	0.0360	1.132	0.3198
Na	6.173	<0.0360	19.67	59.19	31.98	3.877	6.648	319.4	42.66	4.462	29.77	<0.0360
Ni	0.1611	0.1200	0.3915	0.3823	3.942	4.058	4.005	0.1894	0.1643	2.460	0.1649	1.601
Pb	<0.015	0.7000	0.3915	402.2	0.1372	<0.015	<0.015	0.0534	0.0086	<0.015	0.1162	1.312
V	0.0679	0.0531	0.2073	0.2109	4.919	3.582	3.633	0.0667	0.0674	1.082	0.0722	0.459

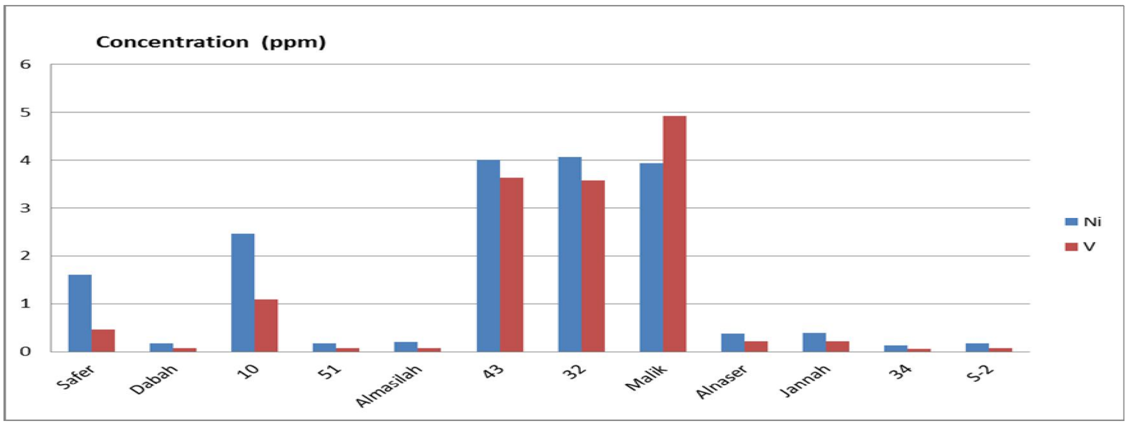

Figure 2: Cconcentration (ppm) of Ni and V ratio of Yemen crude Oil

Table 2: Parameters in Yemen crude Oils

Sampling sectors	Ratios of V to Ni, V to Cr, V to Fe and Ni to V			
S2	0.3521	37.05	0.0036	2.37
34	0.4425	40.86	0.1397	2.3
Jannah	0.5516	0.1198	0.0022	1.92
Al naser	0.5295	11.67	0.0021	1.81
Malik	1.274	30.38	0.1474	0.801
32	0.9293	2275	3.256	1.133
43	0.9071	2794	0.794	1.10
Masilah	0.3521	37.05	0.0036	2.77
51	0.4102	51.84	0.0198``	2.44
10	0.4398	832.3	174.5	2.27
Dabah	0.44	55.54	0.023	2.28
Safer	0.29	54.65	0.27	3.49

Table (2) shows the range of Ni/ V ratio was found to be (0.80 –3.49), V/Ni (0.29-1.274), V/Cr (0.1198-2794), V/Fe (0.0036-174.5).

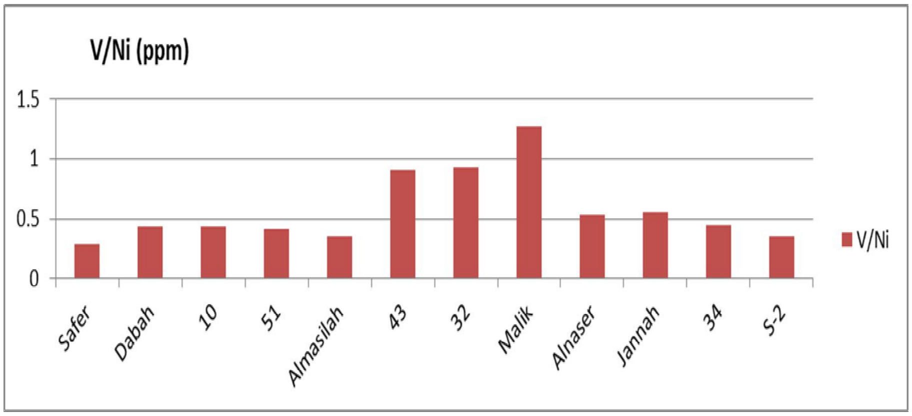

Figure 3: Concentration (ppm) of V/Ni ratio for Yemen crude oil

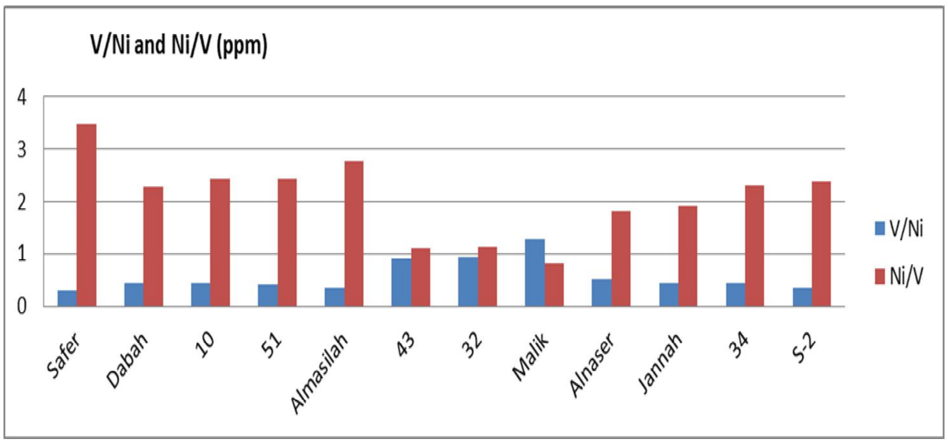

Figure 4: Concentration (ppm) of V/Ni to Ni/V ratio for Yemen crude oil

Table 3: Normal alkanes/isoprenoid ratios

Sample	Pr/Ph	Pr/n-C17	Ph/n-C18	n-C25/n-C18	CPI	Pr+C17/Ph+C18
Gannah	0.4037	0.4152	0.4737	0.5635	1.0476	0.4423
Almailah	1.6585	0.4947	0.4484	0.1622	1.1645	1.7153
Marib	1.6030	0.5834	0.4921	0.2695	1.09	1.5643

Table (3) the range of Normal alkanes/isoprenoid ratios Pr/ph (0.4037- 1.6585), pr/n-C17 (0.4158-0.5834), ph /n-C18 (0.4484-0.4921), n-C25/n-C18 (0.1622-0.5635), CPI (1.09-1.1645), Pr+n-C17/Ph+n-C18 (0.4423-1.7153).

Table 4. Mean a standard deviation of element of Yemen crude oil.

Properties	Mean	Standard deviation	CV
Ag	0.8484	1.2238	0.0614

Ca	27.3442	0.8560	1876
Cd	34980	1.2272	0.0195
Co	. 50083	2.3875	0.1615
Cr	6.4986	1.6214	1.7760
Fe	3.0091	1.7985	94.3000
K	19.2976	1.2951	4.1690
Na	33.9723	2.6766	19.6700
Ni	11.7285	1.2240	0.3915
Pb	0.01354	2.7469	0.3915
V	2.0366	0.8103	0.2073

3.1 Statistical analysis of the results

Table (4), the Statistical analysis of the results shows that, the light crude oil samples obtained Positive correlations observed in most of the metallic parameters. With the exception of Ca, Fe and Na, no homogenous, but generally the crudes have similar physicochemical characteristics and may also have a common geological characteristic. At divided by in tow basic area exploration and product from the above table, it is shown the CV-test is greater than significant level (0.05) that means there is no difference between the properties of Yemen crude oil.

Discussion

The levels of most of the trace elements concentration obtained in this study were generally low. This agrees with previous reports that light crude oil samples usually contain relatively low trace metal contents compared to the heavy crudes (Osuji, L. C; et al; 2005) The relatively higher levels of Ni in Malik, 32, 43 (3.942, 4.058, 4.005) and V (4.919, 3.582, 3.633) respectively, Fe (Jannah, Alnaser, Malik), (94.3,92.7, 33.45) respectively. Ni, V and Fe detected concentration on some results should be expected because these metals are commonly associated with crude oil samples
(Madu, A. N; et al 2011). V/Ni ratio generally used in the reconstruction of the depositional environment Mohialdeen, I.M.J., et al; 2012). The high ratio of V/Ni (4.64) is the best indicator for marine source rock. The high V/Ni ratio a reducing, carbonate-rich environment that receives more marine organic matter (Peters, K.E., et al., 2005). Possibly the source rock of this heavy oil is the Kolosh Formation (Paleocene-Early Eocene) which is composed of a sequence of clastic sediments and some carbonate shelf deposits. Older rock units, such as Shiranish Formation (Late Cretaceous) also can be considered as a source of this heavy oil. The studied crude through oil samples from bazian area, Suleiman, Kurdistan Region show high V/Ni ratio, which indicates oil originated from Suleiman carbonate-rich. The absence of main groups of biomarkers, except of tricyclic turpanes and some diasteranes, indicates heavy biodegradation process (Ibrahim M; 2013). V and Fe were derived from the original source material (Ball; et al; 1960 and Hyden; et al; 1961). However conclude that the V/Ni ratio increased with reservoir age. Much of this disagreement probably arises between the porphyrin and non-porphyrin forms of the metals in these

oils and the probable variation in the trace metal assembles age of the source rocks (Bonham. L.C; et al; 1956). (AL-shahristani and AL-tyiab 1972) measured the Ni and V contents of crudes from their field in Iraq or study designed to resolve controversy on the origin of the Oils of northern Iraq. One school of thought (AL-shahristani and AL-tyiab 1972) maintains that the source rocks from which the oils are derived are contemporaneous with the latter (cretaceous and Paleocene reservoirs in which they are found. Another school of thought (Dnnington, H.V; 1967). The nature of metals in crude and residual oils is of interest to the refiner as they are the source of environmental pollutants and the cause of corrosion of equipment and poisoning of process catalysts V and Ni have been studied more thoroughly than any other metallic element found in petroleum. One of the reasons is that these elements occur, in part as nitrogen complexes (porphyries) closely related to chlorophyll and hemoglobin, thus suggesting a biogenic origin for petroleum. Many correlations based on vana-V and Ni content have been made in attempts to obtain information on the geological origin of petroleum. (Hodgson; 1954) in a study of the oils of Western Canada measured V, Ni, and Fe and concluded that the V/Ni ratio decreased with increasing maturation. (Hyden; 1961 and Ball et al; 1960), however, presented data to show that V/Ni ratio increased with the age of the host rock. Similar inclusions were used by (Al-Shahristani; 1972) to suggest that a V/Ni ratio of less than 1 indicates that the oil is of a younger age, e.g. Paleozoic, while, a ratio of more than 1 indicates that oil is of an older age, e.g., cretaceous. More recently, (A bu- Elgheit W; et al 1979) studied the total V and Ni contents of petroleum residues from different oil fields in Egypt and concluded that the V/Ni index decreases with the age of oil. A V/Ni index of 1-1.44 was reported for the Early Cretaceous oils and an index of 3.25-3.53 for the Miocene oils. Heavy metals are often found to be part of crude oil samples. Possible sources of trace metals in crude oil are: through incorporation and diagnosis of metal complexes of the origin a biological materials through incorporation into the organic matrix during diagnosis of the biological materials in the source rocks either from clay minerals or interstitial aqueous solution through an aqueous phase during primary and secondary migration and from formation water or reservoirs' rock minerals (Fausnaugh, 2002). The levels of most of the trace elements obtained in this study were generally low. This agrees with reports that light crude oil samples usually contain relatively low trace metal contents compared to the heavy crudes. (Dusseault, M.B; 2001 and Osuji, L. C; et al; 2005). V and Ni are commonly associated with iron ores. Studies such as (Akpan, I.O; 2005 and Oderinde, R. A; 1984). Most of the crude oil samples used for this study were obtained from desert and oil wells which are associated with Paleocene or Cretaceous environment. The result obtained in the present study is also within the range of the results from previous studies a further comparison of the levels of these heavy metals was made with recommended limits of these parameters in soil and water environment by the World Health Organization (WHO, 2011). According to (Udeme John Dickson; et al 2012), the levels of some trace metals such as Pb, Cd, Cr, Mn, Zn, Cu, and Co were observed to be low when compared to recommended levels of these parameters by (WHO, 2011), in the environment, high values of Ni, V and Fe were observed in the crude oil blends. The results therefore, show that crude oil samples from Nigeria could be a source of heavy metals particular Ni and V in the environment. From the study result, it is concluded that crude oil obtained from Nigeria have a common geological and formative history (Oderinde, R. A; 1984). From Table (3) normal alkanes/isoprenoid ratios the distribution of n-alkanes in crude oils can be used to indicate the organic matter source (Udeme John Dickson; et al; 2012). For example, the increase in the n-C_{15} to n-C_{20} suggests marine organic matters with a contribution to the biomass from algae and plankton (Peters, K.; Moldowan, J; 1993 and Ficken, K.; et al; 2000). In this study n-C_{15} to C_{20} increases, which implies that Yemen crude oil was generated marine organic matters. Oil samples characterized by uniformity in n-alkenes distribution patterns suggest that they are related and have undergone similar histories with no signs of biodegradation (Peters, K. J; 1993). CPI is affected by both source and maturity of crude oils (Tissot, P ; Welte, D; 1984). CPI of petroleum oils ranges to about 1.00, generally shows no even or odd carbon preference which indicates mature samples. Also, it can be used in source identification; petroleum origin contaminants characteristically have CPI values close to one Maioli, O. L.; et al; 2011). From table (3) Carbon preference index CPI of Yemen oils ranges to about 1.00 (1.04-1.1) which indicates maturity of the Yemen crude oil.

$$CPI = 0.5\left[\frac{C_{25}+C_{27}+C_{29}+C_{31}+C_{33}}{C_{24}+C_{26}+C_{28}+C_{30}+C_{32}}+\frac{C_{25}+C_{27}+C_{29}+C_{31}+C_{33}}{C_{26}+C_{28}+C_{30}+C_{32}+C_{34}}\right]$$

(Bray, E. E.; Evans, E. D; 1961). The relative amounts of C_{27}-C_{29} steranes can be used to give an indication of source differences (Hunt, J; 1996). For example, the predominance of C_{28}, C_{29} and C_{30} steranes indicate an origin of the oils derived mainly from mixed terrestrial and marine organic sources, while oils showing a slightly low abundance of C_{28} and C_{29} and relatively higher concentrations of C_{27} steranes indicate more input of marine organic source. In this study, the results show a slightly low abundance of C_{28} and C_{29} and relatively higher concentrations of C_{27} steranes which indicates more input of marine organic source.

Pr/Ph believed to be sensitive to diagenetic conditions; Pr/Ph ratios substantially below unity could be taken as an indicator of petroleum origin and/or highly reducing depositional environments. Very high Pr/Ph ratios more than 3 are associated with terrestrial sediments. Pr/Ph ratios ranging between 1 and 3 reflect oxidizing depositional environments (Lijmbach, G; 1975). Low Pr/Ph values <2 indicate aquatic depositional environments including marine, fresh and brackish water (reducing conditions), intermediate values 2-4 indicate mixed organic source matter, terrestrial environments, whereas high values up to 10 are related to peat swamp depositional environments (oxidizing conditions) (Hunt, J; 1996). pr/ph ratio in this study is <2 (0.4037 - 1.6585) these indicate that the Yemen crude oil was derived from marine sources, with increasing maturity. Accordingly, isopreniods/n-alkanes (Pr/n-C_{17} and Ph/n-C_{18}) ratios provide valuable information on biodegradation, maturation and diagenetic conditions. The early effect of microbial degradation can be monitored by the ratios of biodegradable to the less degradable compounds. Isoprenoid hydrocarbons are generally more resistant to biodegradation than normal alkanes. Thus, the ratio of the pristane to its neighboring n-alkane C_{17} is provided as a rough indication to the relative state of biodegradation. This ratio decreases as weathering proceeds (Waples, D; 1985). The Ph/n-C_{18} values less than 1.0 are indicative of non-biodegraded oils. Both Pr/n-C_{17} and Ph/n-C_{18} decrease with maturation due to increasing prevalence of the n-paraffin. From Table (3) ph/n-C18 (0.4484-0.4921) values less than 1.0 are indicative of non-biodegraded Yemen crude oils but, the decease of both Pr/n-C_{17} and Ph/n-C_{18} indicate maturation of the Yemen crude oil.

CONCLUSION

The levels of most of the trace elements obtained in this study were generally low. This agrees the fact that light crude oil usually contains relatively low trace metal contents compared to the heavy crudes. Ni, V and Fe observed in some result should be expected because these are metals commonly associated with crude oil. Yemen crude oil was derived from marine organic matters. CPI of Yemen oils average value of 1.00 (1.04-1.1) indicates the maturity of Yemen crude oil. The ratio of pr/ph in this study is <2 (0.4037 - 1.6585) which indicate that the Yemen crude oil was derived from marine sources, with increasing maturity. Ph/n-C18 (0.4484-0.4921) values less than 1.0 are indicative of non-biodegraded Yemen crude oils, but both Pr/n-C_{17} and Ph/n-C_{18} decrease which indicate that the Yemen crude oil is mature. Most of the crude oil samples used for this study were obtained from desert and oil wells which are associated with Paleosen/Neosen or Cretaceous sediments.

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