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Abstract

The present investigation was designed to determin of residual of petroleum hydrocarbons in seawater and sediments from Aden coasts, Yemeni, during April 2004. Additional hydrographic data such as air and water temperatures, salinity, dissolved oxygen, pH and total alkalinity were also determinated in order to give a better appraisal of survey data. The mean concentrations of residual petroleum hydrocarbons in seawater were varied from 19ng/ l at Al-Omal Island to 239ng/l at free area and in sediment from 9ng/g at Al-Omal Island to 45 infront of refinery. The study revealed comparable level of petroleum hydrocarbons in this region to that reported for other parts of the world, which showed no pollution with petroleum hydrocarbons

Introduction

The Gulf of Aden and the Red Sea are among the busiest tanker routs. Most of the oil produced in the region is exported via sea and pipeline, while local refineries and consumption are located in the coastal area. The widespread of oil pollution in the Red Sea and Gulf of Aden are not surprising (Wennink and Nelson- Smith, 1979; Dicks, 1987; DouAbul and Heba, 1995; DouAbul and Al- Shiwafi, 1996). The growing in pretence of Yemen as an oil producing nation, and it's close proximity to one of the world's busiest shipping lanes means a high risk of oil pollution in various forms. Chronic and incidental oil pollution takes place in the following forms:

1) Shipping discharges in the Gulf of Aden of dirty ballast/ tank washing overboard at sea. These discharges from tar balls and oil droplets, which have been found throughout the coastline. Such practices are wide spread, largely due to lack of surveillance.
2) Shore facilities in Aden port. It was reported that a major in – land fuel oil tank failure in 1986 resulted in a continual seepage of oil into the harbor area.
3) Poor maintenance has resulted in low level, but locally significant leaks oil from bunkering facilities in the harbor.
4) Aden refinery effluent- oily water resulting from refining process is separated, and the effluent water discharged into Khor Ghadir.

5) Minor- medium spills- alarge number of minor spills (18 liters to 2 tones, mostly of fuel oil) have been reported in Aden.

Oil pollution has its impact on man, fish and shellfish, seabirds, animals and plants. The effect on fish and shellfish may be toxic effect or tainting. Sea birds may be killed, because the oiled bird, in attempting to clean itself from oil, may ingest some of it, also oil has a physical effect on the bird when penetrating the feather and the feather losers its waterproof. It is estimated that 150.000 birds are killed each year by oil pollution in the North Sea and North Atlantic (Hont, 1969).

Oily and tarry substances may discolor the water and seriously foul boats, fishing nets and gear, buoys, mooring lines and other marine structures. Oil floating on the nearshore waters deters bathing and swimming the tarry deposits on the beach may adhere to clothing or bodies of beach visitors and users.

During the recent years, the subject of hydrocarbons concentration in seawater and sediments and its impact on marine environment has attracted many scientists concerning with the water quality around industrial countries.(Badawy *et al*., 1993; Burns, 1993; Al-Lihaibi, 2003;Pandit *et al*., 2006). Meanwhile, others concentrated on the impact of these hydrocarbons on certain marine animals (Klionsky and Emr, 2000; Livingstone, 2001; Hwang *et al*., 2002; Lowe *et al*,2006)

Objective of the study

The main objective of the study was to perform a baseline study on the state of pollution on beaches of Aden City. The objective were:

*To determine residue levels of potentially hazardous petroleum Hydrocarbons

Which there in the sea water. Consequently to identify the most important sources of contamination in our country environment.

* To collect and review the relevant existing data and to arrange the results that obtained during this study.

* To serve as a baseline data for further follow-up study in beaches of Aden City.

* To compare the collected data locally and internationally.

Description of area (Aden city)

The Gulf of Aden coast is dominated by the Indian Ocean monsoon system. From January to March the northeast monsoon blows in a southwesterly direction. During this period the highest annual rainfall occurs and after flash floods the wadis

discharge into the sea forming the alluvial fans that are a characteristic feature of the region. Between May and September the southwest monsoon generates winds in a northeasterly direction. The relatively warm surface water of the Gulf of Aden is blown offshore and replaced by cooler, nutrient rich water from deeper layers. These upweelings limit the growth of coral reefs, but they trigger high primary production which supports the regions rich pelagic fish stocks (UNDP/GEF, 1996).

Aden is the largest coastal city and farms for transit trade, boat building, shiprepairs and bunkering. Before the closure of the Suez Canal in 1967, Aden was the third largest bunkering port in the world. Besides port related activities, major economic resources in the coastal zone are fisheries, maritime traffic and oil and gas exploitation. Coastal and marine tourism has so far played a mineral.Crude oil is supplied by pipeline from the shore, then transferred from the vessel to tankers. A barge is available for transport of ship based garbage and oily waste, which is carried by lorry to a municipal landfill where it is burned. The port and entrance channel are dredged regularly and dredge spoil is dumped offshore (Aden Port Development, 1996).

Material and Methods

Sample collection and Analysis

Samples of sub- surface water (~1m depth) were collected from 9 stations around Aden City- Gulf of Aden during the period April 2001 by means of hands (Figure, 1). Water samples were full into 2.5-Lamber glass bottle containing 50-ml re-distilled hexane Ehrhardt (1987). Solvent extraction was carried out during sampling and later on for about 30 min by a mechanical stirrer equipped with a stainless steel rod. The organic layer was taken and concentrated to 5 ml by a rotary evaporator and kept in refrigerator. In the laboratory the samples were applied to a column 200* 6mm o.d., slurry- filled with fully activated silica gel. Elution was with 20 ml (≈ 5 bed volumes) of re- distilled n- hexane plus 20% re- distilled dichloromethane. This procedure removes lipids and most other materials interfering with the measurements. Only some carotenoids are eluted with this solvent which apparently did not affect the measurement. In some cases considerable amounts of lipids remained on the silica gel column. The eluted was concentrated by rotary evaporation and brought to volume in 10-ml volumetric flasks. Aliphatic hydrocarbons (n-C_{13} to n-C_{34}), pristane, and phytane is analyzed by infrared Spectrometer (IR) at 2930^{-1} absorbance (APHA, 1985). Blank determinations were

carried out by repeating the procedure with a pre- extracted samples. Using a calibration with Maribe light crude.

Air, water temperature were measured at the time of sampling, using standard reversing protected thermometers, with accuracy of $0.01C^{o}$. Salinity was determined by water sampling was in the order of oxygen, pH and total alkalinity. The samples were collected in dark bottles and kept in dark box for salinity, pH and alkalinity determination in the laboratory.

In the water samples, dissolved oxygen was analyzed according to the modified winkler's method. Salinity of each sample was analyzed against Copenhagen Standard Seawater at the Department of Earth and Environmental Science, Sana'a University. The titration method described by Grassoff, (1982) was used, total alkalinity of each sample was determined by the method of Anderson and Robinson, (1964) as developed by Culberson, (1970).

The combination electrode was standardized with pH. 4.00 and pH 7.00 buffers at $25C^{o}$. The pH of each sample was calculated from:

$$pH_{sw} = (E_{sw} - E_4) / S$$

Where pH_{sw} is the pH of the sample, E_{sw} and E_4 are the measured potential of the sample and 4.0 buffer, respectively, and S is the measured slope of the electrodes obtained from:

$$S = (E_4 - E_7) / (4.00 - 7.00)$$

Where E_7 is the measured potential of the buffer solution 7.00. The slope of the electrode response was constant over long periods, and measurements of 7.00 buffer were made of the beginning and the end of the day. All measurements of pH and total alkalinity were made at $25 \pm 0.1C^{o}$ where the samples were placed in water bath for 10-20 minutes before measurements. The pH-meter used was digital pH/mV meter. The precision of the measurements were ± 0.01 pH and ± 0.009 $meq1^{-1}$ for alkalinity.

It is well established that aquatic sediments are the final accumulation site of water- borne constitutes derived from natural sources (living organisms and their detritus) in situ and surrounding, and artificial (domestic, urban- industrial and agricultural wastes) sources (DouAbul *et al*., 1984). The extraction method is that of Wade *et al*., (1988). A total of 10 g of dried sediment was soxhlet- extracted with methylene chloride and concentrated in Kudema- Danish tube. The extracts were

fractionated by alumina: silica gel (80-100 mesh) chromatography. The extracts were sequentially aluted from the column with 50 ml of *n*-hexane (aliphatic fraction).

The determination of petroleum residues was carried out following the Infra- red spectrophotometric method that approved by standard Methods Committee, 1985 (APHA, 1985). Using a Calibration with Marib light crude. Blank determinations were carried out by repeating the procedure with a pre-extracted samples.

Total organic carbon (TOC) and grain size analysis were performed on representative slips of the bulk sediment samples in order to give a better appraisal of petroleum hydrocarbons in the sediment. Percentage organic carbon (%TOC) was carried out according to the procedure of El-Wakeel and Riley (1957). Granulometery were conducted by the combined dry sieve and pipette method (Folk, 1974).

Results and Discussion

Data present in tables 1 and 2 and figures 2 and 3 were shown:

The residual of petroleum hydrocarbons in sea –water ranged from 19ng l^{-1}at Al-Omal station to 239ng l^{-1} at free area with mean 152.11ng l^{-1} and marine sediment from 9 ng/g at Al- Omal island to 45 ng/g infront of refinery with mean 30.89 ng/g (API light marib crude oil equivalents).

Table (1): Results of Aden city stations (sea water)

No	Nearest location	Concentration ng l^{-1}
1	Al-Omal Island	19
2	Abyan Coast (1)	135
3	Abyan Coast (2)	145
4	Syra'a	209
5	Free area	239
6	Algader area	150
7	Infant of refinery	217
8	Steam Station	139
9	Gold Moor	116
Range		19-239
Mean		152.11

Table (2): Results of Aden city stations (marine sediment)

No	Nearest location	Concentration ng /g
1	Al-Omal	9
2	Abyan Coast (1)	27
3	Abyan Coast (2)	29
4	Syra'a	33
5	Free area	40
6	Algader area	35
7	Infant of refinery	45
8	Steam Station	42
9	Gold Moor	18
Range		9-45
Mean		30.89

From the result presented here is evident that all the site are contaminated to some extent with petroleum hydrocarbons. The operational discharges from shipping are restricted. Nevertheless evidence suggest that oil pollution from these sources has a far greater effect on the marine environmental than accidental spills. An example of chronic oil pollution sources on the Gulf of Aden with vessels deballasting at the Aden refinery. However, the problem of passing vessels deballasting in the Gulf of Aden or the Red Sea appears to be the greater cause of oil pollution in republic of Yemen waters. Oil spillage is common and there is no organization to deal with spillage.

Table (3) Comparison of Hydrocarbon in seawater collected from different region of the world

Area	Concentration (ngl^{-1})	Source
Coastal waters of UAE	5600	Shriadh,(2000)
Coastal of Qater	2500	El-Samra *et al.*, (1986)
Coastal waters of Saudi Arabia	4300	Fowler,(1985)
Coastal of Oman	7500	Fowler,(1985)
Gulf of Mexico	3900	Marchand *et al*.,(1982)
West Coast of India	120.0-244.0	Sen Gupta *et al* ., (1978)
Coastal waters of Aden city	152.11	Present Study

Table (4) Comparison of hydrocarbon content in sediment collected from different region of the world

Area	Concentration (ng/g	Source
Coastal waters of UAE	1050-8830	Shriadh, (2000)
Arabian Gulf	400-44000	DouAbul,*et al*., (1984)
Gulf of Aden/Arabian Sea	120-2100	EPC,(1996)
Mina al Fahal coastal waters,Oman	4100-6800	Bady *et al*.,(1993)
Monteevideo, Uruguay	100-4940	Moyano *et al*.,(1993)
Dar el Salaam Harbor (Tanzania)	Up to164000	Machiwa,(1992)
Port of Spain Harbour, Trinidad	2800-240500	Agard,(1985)
Coastal sediment of Aden city	9-45	Present study

Table (5) Sedimentalogical Parameters

Station	% Total Organic Carbon (TOC)	Sediment type
1	0.03	Medium sand
2	0.05	Very fine sand
3	0.04	Very fine sand
4	0.05	Medium sand
5	0.05	Medium sand
6	0.04	Fine sand to Medium
7	0.06	Very fine sand
8	0.05	Fine sand
9	0.03	Coarse sand

These results are within the range of values residual hydrocarbons in sea water from Gulf of Aden, as reported previously Table (3).

And hydrograhpic parameter in conjunction with factors such as air, water temperature salinity, dissolved oxygen, pH, and total alkalinity

Goldberg (1975) has reported that the unpolluted open ocean sediments contain 1-4 ug/g dry weight hydrocarbons. Less than 100µg/g in coastal sediments and up to 1000 ug/g in highly polluted areas comparing the concentration of residual petroleum hydrocarbons of the present study with the values reported by other workers Table (4)

However it should be borne in mind that the efficiency of hydrocarbons adsorbance onto sediments particles are governed mainly by its grain size and total organic matter content. The sandy nature of the Gulf of Aden coasts of Yemen

sediments coupled with its very low content of organic matter (< 0.1%TOC, EPC 1996) render its low capacity for hydrocarbons adsorption Table (5). The complexity of hydrocarbon composition of environmental samples made the relative ranking of importance of various diffuse urban/ industrial sources difficult. Thus the differences in hydrocarbon composition between various ecosystem components imply the importance of biogeochemical processes acting on hydrocarbons discharged to coastal water. Nevertheless we shall discuss below the most probable source of hydrocarbons encountered to environmental samples from the Aden city. Our dates show that the concentration of oil in sediment increase in the eraes of various human activity. Another factor for concentration of oil in sediments are related to the physical properties of the sediments, the increase of oil inputs is caused by major oil spills, generally from errant oil tankers, blowouts, oil pipelines and storage tanks as a result of operational errors or caused by erroneous deliberate acts of war (Johnson, 1985). The concentrations of toxic components are generally low, localized and shot lived particularly in water, which is subjected to high physical energy (wave action). Area most susceptible to toxic effect from oil are sheltered bodies of water and stable benthic sediment (EPC, 1996). Although there is controversy about the ecological effects of oil in the marine environment, many of the world's major oil spills have had long- term consequences (Vandermeulen, 1982). The effects of oil on biological system arise from mechanical smothering and from the presence of toxic substances. The overall severity of the effects depends on the nature and quantity of oil spilled.

Table (6) The Results of air, water temperature, salinity, pH and total alkalinity at each station

No.	Location	Air temp. C^o	Water Temp.C^o	Salinity ‰	Dissolved O_2 mg/l	PH	Total Alkalinity
1	Al-Omal	29.5	30.0	38.0	4.60	8.01	2.330
2	Abian Coast (1)	28.3	29.3	37.9	4.50	8.00	2.293
3	Abian Coast (2)	28.1	29.1	37.9	4.45	7.99	2.300
4	Syra'a	28.3	28.5	37.5	4.43	8.00	2.291
5	Free area	30.0	30.5	38.3	4.20	7.90	2.230
6	Algader area	27.8	28.2	37.8	4.70	8.01	2.329
7	Infront of refinery	28.4	29.0	38.0	4.40	7.90	2.285
8	Steam station	29.0	29.7	38.1	4.37	7.87	2.280
9	Gold moor	27.0	27.3	37.3	4.80	8.02	2.350
	Mean	28.49	29.07	37.87	4.49	7.97	2.299
	Range	27.0-30.0	27.3-30.5	37.3-38.3	4.20-4.80	7.87-8.02	2.230-2350

1- *Air temperature:*

It ranged between 27.0 and 30.C^{o} with mean of 28.44C^{o}. The minimum value is at station (9), while maximum value is at station (5). The difference between the air temperature due to a strong decrease in mean surface temperature in the eastern Gulf of Aden during these months, the result of upwelling current off Somalia (Naval Oceanography Command Detachment, 1982).

2- *Water temperature*:

It ranged between 27.3 and 30.5C^{o} with mean of 29.07C^{o}. The minimum value is at station (9), while the maximum value is at station (5). Such distribution reflects that the inshore surface waters are caused by upwelling current off somalia. Both air and sea surface temperature peak in June but dip in July and August relative to the June and September temperature (Ba- Sumaidi, 1997).

3- *Salinity (S‰= ppt):*

The salinity readings were not more difference, the lowest value is at station (9), while the highest value at station (5). These variations may be mainly due to local hydrographic condition foe example semi-isolated, intensive evaporation and restricted circulation, (GEF, 1999 and Al-Shwafi, 2001).

4- *Dissolved Oxygen (D.O):*

The surface water of the Gulf of Aden and Arabian sea along the coast of Yemen contained sufficient amount of dissolved oxygen (Table, 6). Dissolved oxygen values ranged between 4.20 mg/l at station (5) to 4.80 mg/l at station (9) (Fig.4). These fluctuations may be attributed to several hydrographic and biological condition prevails at various locations.

5- *Hydrogen Ion concentration (pH) variations*:

The surface horizontal (geographical) distribution of pH is shown in (Table, 6). The general pattern of the surface horizontal distribution of pH gave local variation mainly due to the climate conditions, and consequently water temperature, as well as dissolved oxygen content and biological activity, (Hanna, *et al*., 1988).

6- *Total alkalinity (T.A)*

The total alkalinity value varies from 2.280 at Free area to 2.350 at Gold moor and the mean value is 2.366 meq1^{-1}. These values of total alkalinity are possibly

caused by the mixing processes and temperature increase which both expel Co_2; and photosynthesis processes. The photosynthesis processes is confirmed by the high values of O_2 content of surface seawater (Rushdi, 1994).

Conclusions and recommendations

In the light of the above reasoning we may thus is conclude that the Aden city-Gulf of Aden environment is subjected to the same point- sources pollution of oil contamination. This pollution is a consequences of localized oil operations (Rushdi *et al*., 1991) and/ or heavy ship traffic {Currently 100 million tons of oil transit the Red Sea annually (PERSGA, 1995)}.

The study recommends that continuos monitoring programme for the Aden city-Gulf of Aden coast of Yemen should be formulated and conducted to ensure that the concentrations of petroleum hydrocarbons are within the baseline levels established during the present survey.

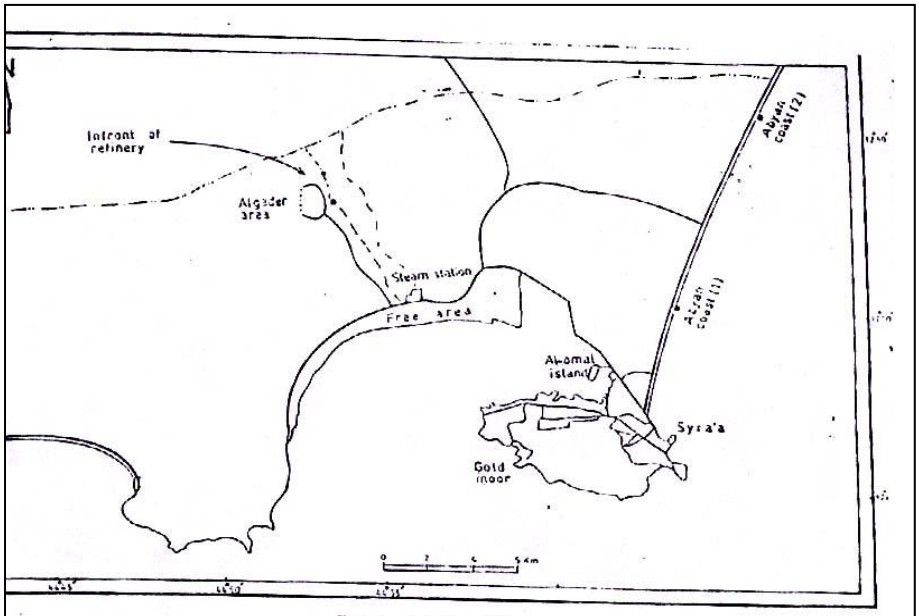

Fig.1 Study area

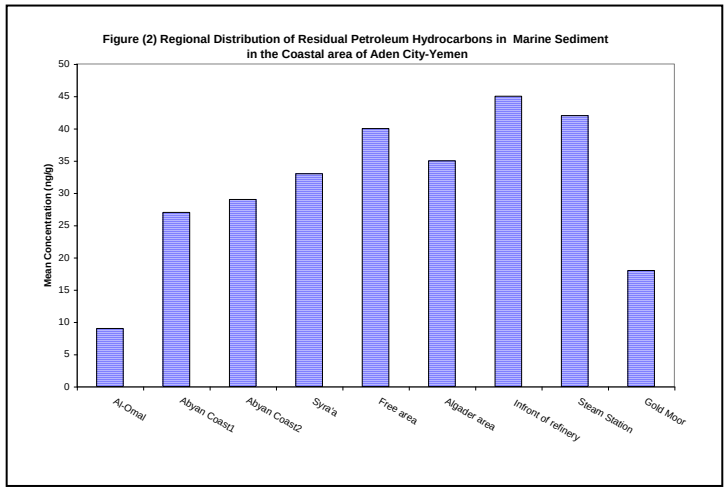
Figure (2) Regional Distribution of Residual Petroleum Hydrocarbons in Marine Sediment in the Coastal area of Aden City-Yemen
Mean Concentration (ng/g)
50
45
40
35
30
25
20
15
10
5
0
Al-Omal
Abyan Coast1
Abyan Coast2
Syra'a
Free area
Algader area
Infront of refinery
Steam Station
Gold Moor

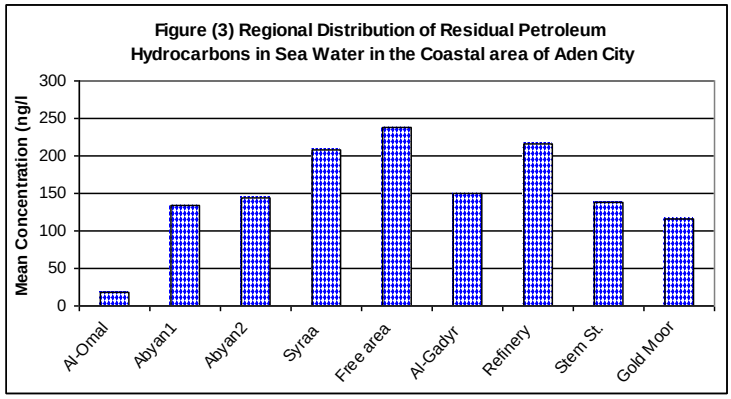
Figure (3) Regional Distribution of Residual Petroleum Hydrocarbons in Sea Water in the Coastal area of Aden City
Mean Concentration (ng/l
300
250
200
150
100
50
0
Al-Omal
Abyan1
Abyan2
Syraa
Free area
Al-Gadyr
Refinery
Stem St.
Gold Moor

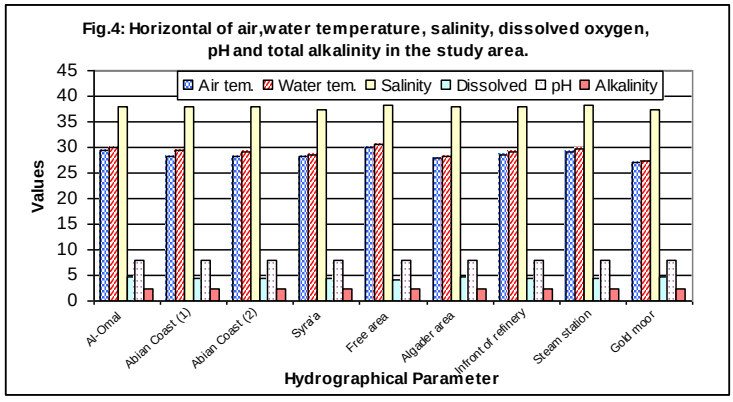
Fig.4: Horizontal of air,water temperature, salinity, dissolved oxygen, pH and total alkalinity in the study area.
Air tem.
Water tem.
Salinity
Dissolved
pH
Alkalinity
Values
45
40
35
30
25
20
15
10
5
0
Al-Omal
Abian Coast (1)
Abian Coast (2)
Syra'a
Free area
Algader area
Infront of refinery
Steam station
Gold moor
Hydrographical Parameter

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الملخص العربي

تقدير الهيدروكربونات النفطية المتبقية في المياه البحرية والرسوبيات لساحل مدينة عدن – اليمن

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تختص الدراسة الحالية بجمع وتحليل عينات من ماء البحر والرسوبيات من السواحل اليمنية (منطقة عدن).

أظهرت نتائج التحليل الكيمائية إن معدلات الهيدروكربونية النفطية المتبقية في عينات ماء البحر تقع ضمن مثيلاتها في السواحل العالمية والتي تعتبر غير ملوثة بالنفط.

تراكيز بقايا الهيدركربونية النفطية في عينات ماء البحر كانت من 9ng/l فى جزيرة العمال إلى 239ng/l فى المنطقة الحرة وفي الرسوبيات من 9 ng/g في جزيرة العمال الى45 ng/g أمام مصافي عدن بالإضافة تم قياس وتعين كل من درجة حرارة الجو والماء ،الملوحة،الأكسيجن المذاب،ألاس الهيدروجيني والقلوية الكلية.