

## **Status of oil pollution in water and sediment from Shatt Al-Arab Estuary and North-West Arabian Gulf**

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**Abstract** - In the present study, an attempt was made to study the levels of petroleum hydrocarbon residues in Water and Sediment samples collected from ten stations at the region of Shatt Al-Arab Estuary and North-West of the Arabian Gulf. The water and sediment samples were analyzed by utilizing the spectrofluorometry technique. The results of the present research obviously indicated a degree of oil pollution, however, still lower in magnitude when compared with status in some other regional sites. The obtained levels of petroleum hydrocarbons in water was ranged from (3.09 µg/l) at station 7 to (30.87 µg/l) at station 9 while in sediment ranged from (19.43 µg/g) dry weight at station 4 to (49.09 µg/g) dry weight at station 9. In order to give a better evaluation of the petroleum hydrocarbon levels in the sediments, the Total Organic Carbon (TOC) percentage and grain size analyses were done by granulometry technique on selected slips of the bulk sediments obtained for this purpose. The mean percent TOC estimations ranged from 0.1 at Station 4 to 1.34 at Station 9. The mean values obtained in this study indicated that the petroleum hydrocarbon levels in the sediment samples are lower than the levels in sediment samples obtained with similar methodology and analyses in some of the Gulf States and countries in close vicinity of the region. The major pollution sources may involve tanker and boat discharges and activities, municipal sewage and rural run-off from land. Also, discharges and effluents from oil refineries, electricity generating station and industrial activities into the Basrah city are obvious.

**Keywords:** Petroleum hydrocarbons, Spectrofluorometry, Granulometry, Total organic carbon and Pollution sources.

### **Introduction**

The Gulf is located in a subtropical region. It is shallow, semi-enclosed sea with a surface area of approximately 239,000 km<sup>2</sup> and an average depth of only 35 m. The Arabian Gulf has suffered from many oil spills due to military action (UNEP, 1991). In addition to oil spills, environmental pollution in the area arises from local exploration, refining, pipelines and tankers and natural seeps. In 1978, the spills from transported oil in the Arabian Gulf were as limited to range 0.95-1.27 million barrels (Oostdam and Anderlini, 1978). Oil spilled from the Iranian Nowruz oil field in 1983 was estimated to be about 400,000 barrels. The oil released as a result of the 1991 Gulf war was the largest oil spill in history, with an estimated release of 6-8 million barrels of Kuwait crude oil (Literathy, 1993). Al-Saad and Salman (2012)

presented a review which brings the valuable sources of information regarding oil pollution status in the Arabian Gulf and the adjacent areas of Shatt Al-Arab Estuary and Mesopotamian marshes. Freije (2015) gave a review on heavy metals and trace element and total hydrocarbons in the Arabian Gulf. The review has revealed the different concentrations of pollutants *viz.*, low, moderately and chronically contaminated areas from oil and metals. It has also outlined effective sustainable management measures and goals as a first step in the evaluation of coastal, marine, soil and air environment in the Arabian Gulf area.

Shatt Al-Arab river, Khor Al-Zubair and the Iraqi coastal waters are liable to contain small amounts of oil spills. There are different sources such as loading and transportation facilities, effluent discharges from oil refiners, atmospheric fallout etc. (Al-Saad, 1995).

Khor Al-Zubair is an extension of the Gulf waters in the lower reaches of Mesopotamia (Fig. 1). The upper north is connected with Shatt Al-Basrah canal and receives freshwater influx of average 700 m<sup>3</sup>/sec throughout the tidal which changing the environment of the Khor from hypersaline lagoon to an estuary one (Hussain and Ahmed, 1999). While, the lower (south) end is connected with Khor Abdullh. The current in the Khor is characterized by one directional throughout the tidal cycle towards the southern end (Arabian Gulf).

The place of origin of the Shatt Al-Arab river starts when Euphrates river (after it flows through the Hor Al-Hammar unit) joins the Tigris river at Garmat Ali. Karun river, the only tributary of the Shatt Al-Arab river, joins its eastern bank south of Basrah city. The Shatt Al-Arab unit end when reaching the Arabian Gulf. The length of the Shatt Al-Arab river from Garmat Ali to its mouth in the Arabian Gulf extends about 110 km. It's width varies at different points, ranging from 0.4 km at Basra city to 1.5 km at it's mouth. The water depth increase in general towards the Gulf with maximum of 12.5 m. the water level is affected by the high and low tides of the Arabian Gulf. Pollution of waterways of Shatt Al Arab River and Khor Al-Zubair Channel by petroleum hydrocarbons are expected to occur through different operations: land based discharges, atmospheric and natural inputs (GESAMP, 1993), as well as activities of human beings which comprise discharged through municipal and industrial wastes, urban and river runoffs. Moreover, the area of Khor Al Zubair channel was affected by petroleum hydrocarbons pollutants from fixed installations like Al-Shuaibah refineries, offshore production facilities, loading terminals, etc. (GESAMP, 1993). Untreated municipal and industrial wastes discharged into the lagoons, along with inputs of petroleum from oil spills and motorboat traffic have severely compromised the relative health of the Venice ecosystem in Shatt Al Arab River and Khor Al Zubair Channel (Al-Saad, 1995), and (Al-Saad *et al.*, 1995). Petroleum hydrocarbon inputs into the marine water were computed based on various databases into four categories. These includes: a) natural seeps, b) extraction of petroleum, c) transportation of petroleum and d) consumption of petroleum (Al-Saad, 1998). Oil refinery effluents and losses during loading operations have been identified as the major sources of oil contamination in the waters of Shatt Al-Arab River and Khor Al-Zubair channel which discharge into the North-West Arabian Gulf. The Khor Al-Zubair was also an extremely busy shipping line or oil transports with accidental spilling being almost unavoidable. In combination, these sources provide a long-term input source of petroleum, some major spill, either unintentional or as consequence of military activities have added occasional dramatic pulses of oil contamination to long term background.

## Materials and Methods

### A. Water:

Sampling: Water samples of 5 liter volume from three depths of each sampling site, were collected by means of water sampler. Stored in glass bottles contain 50 ml of Non grade carbon tetrachloride ( $\text{CCl}_4$ ), cooled and transferred to the oil pollution lab of the Marine Science Centre and store there in fridges prior to analysis.

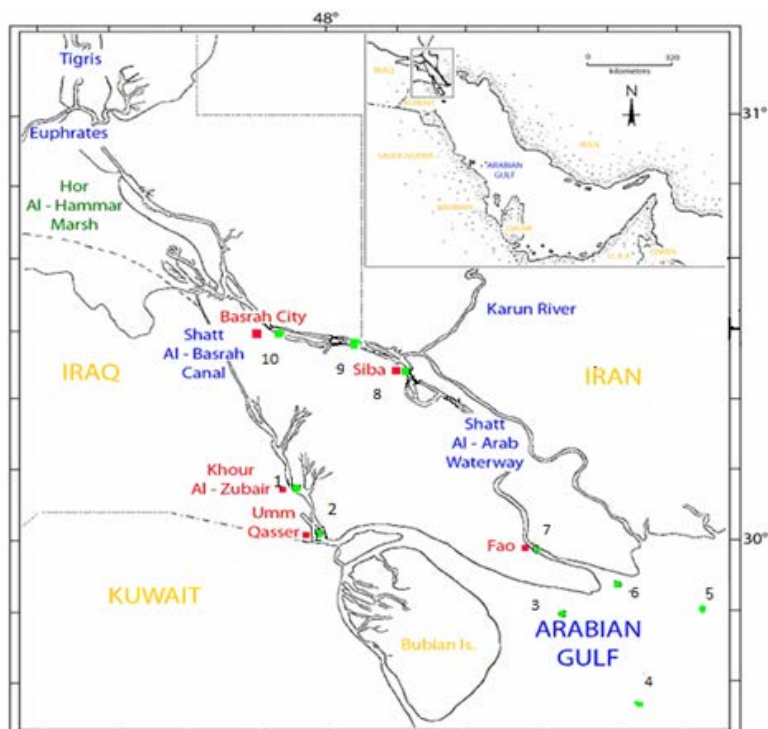


Figure 1. Sample collection locations

Extraction of Oil: Oil as a dissolved phase in water samples were extracted by carbon tetrachloride. Oil in water was solvent extracted following the procedure of (UNESCO, 1976). In this procedure, 100 ml of  $\text{CCl}_4$  was used in two successive 50 ml extractions and the extracts were combined. The mixture was vigorously shaken to disperse the  $\text{CCl}_4$  thoroughly throughout the water samples. The shaken condition was repeated several times before decanting the  $\text{CCl}_4$ . To these extract, a small amount of anhydrous sodium sulfate was added to break any emulsion and to remove excess water. The  $\text{CCl}_4$  extracts were reduced in volume to less than 5 ml by using a rotary evaporator. The reduce extract was carefully pipetted into a pre-cleaned 10 ml volumetric flask, making sure any residual particles of sodium sulfate were excluded and evaporated to dryness by a stream of pure nitrogen. Although,  $\text{CCl}_4$  is ideal solvent for the extraction process. It is not suitable for spectrofluorescence analysis, therefore  $\text{CCl}_4$  must be replaced by a solvent, such as n-hexane which dose not absorb light in the spectral rang of 300-400 nm. The flask was then rinsed with fresh hexane and the rinsing used to make the samples volume up to exactly 5 ml prior to ultraviolet fluorescence (UVF) analysis.

### B. Sediments:

**Sampling:** Sediment samples were also collected from the same locations within the study area by means of a Van Veen grab sampler. Undistributed, triplicate samples were taken. After retrieval of the sampler, the water was allowed to drain off, avoiding disturbing the surface layer of the samples. As soon as the samples were retrieved, they were wrapped in aluminum foil and immediately frozen  $-20^{\circ}\text{C}$ . Before analysis, sediment samples were freeze-dried, ground finely in agate mortar and sieved through a  $62\ \mu\text{m}$  metal (stainless-steel) sieve.

**Extraction of Oil:** Before extracting sediment, samples were kept in a freeze-dryer. The extraction and clean-up procedure for the determination of petroleum hydrocarbons in the sediment was based upon that of (UNESCO, 1976). Sediment was placed in a pre-extracted cellulose thimble and Soxhlet extracted with 150 ml methanol: benzene (1:1) mixture for 24 hours. At the end of this period, the extract was transferred to a storage flask and the samples were further extracted with a fresh solvent.

The combined extracts were reduced in volume to about 10 ml in a rotary evaporator. It was then saponified for 2 hours with a solution of 4N KOH in 1:1 methanol: benzene. After extracting the unsaponified matter with hexane, the extract was dried over anhydrous sodium sulfate, concentrated by a stream of  $\text{N}_2$  for UVF analysis.

### Total Organic Carbon (TOC):

TOC was determined by treating subsamples with phosphoric acids to remove carbonates, then dried at  $60^{\circ}\text{C}$  to constant weight and combusted using a Perkin-Elmer model 240B Elemental analyzer. Grain size analysis was done by using 15 g dried sample. Then, wet sieving was carried by sieve of 230 mesh (Folk, 1974). The fine grain ( $<63\ \mu\text{m}$ ) were passed through the Sedigraph ET-5000 instrument

## Results and Discussion

The oil pollution is one of the most serious pollutants and the most common problems related to the discovery and have since spread through all stages of production, transportation, refining, processing, storage, marketing and even the disposal of waste products.

The oil contents of the water particularly the toxic aromatic fraction was determined by fluorescence spectrophotometer. Water and sediment samples were collected from Shatt Al-Arab, Shatt Al-Arab Estuary, Umm Qasr, Khor Abdullah and Regional Iraqi Waters in July 2012. Figure (1) and Table (1) are illustrated sampling area and the stations location.

Maximum values recorded at station 9 ( $30.81\ \mu\text{g/l}$ ) and ( $49.09\ \mu\text{g/g}$ ) for water and sediment samples respectively, while lower values observed in water collected from station 7 ( $3.09\ \mu\text{g/l}$ ) and in sediment collected from station 9 ( $19.43\ \mu\text{g/g}$ ) (Table 1) and (Fig. 2).

Total organic carbon (TOC), concentration of different classes of hydrocarbons and diagnostic parameters at each sediment sampling site are summarized in Table (1). A significant correlation was observed between the percent of Total Organic Carbon and total petroleum Hydrocarbons ( $r = 0.97$ ). This finding is in agreement with the reports of Al-Muhana *et al.* (2015). The grain size of the sediment samples were mostly muddy sand.

Table 1. Concentrations of Petroleum Hydrocarbons in the study area.

Stations	Water $\mu\text{g/l}$	Sediment $\mu\text{g/g}$	TOC %	N	E
1	24.90	41.92	1.29	30° 11' 38.81"	47° 53' 21.67"
2	16.53	26.27	0.42	30° 01' 19.67"	47° 57' 24.02"
3	19.34	22.74	0.34	29° 53' 03.60"	48° 14' 14.61"
4	20.24	19.43	0.10	29° 40' 44.70"	48° 48' 37.90"
5	5.92	33.12	0.80	29° 47' 15.14"	48° 48' 09.68"
6	4.18	28.02	0.78	29° 52' 16.56"	48° 41' 07.34"
7	3.09	30.73	0.73	29° 58' 22.23"	48° 29' 25.19"
8	14.51	38.98	1.10	30° 23' 15.61"	48° 10' 49.44"
9	30.81	49.09	1.34	30° 27' 46.44"	48° 01' 25.35"
10	18.20	43.16	1.33	30° 28' 17.63"	47° 55' 23.64"

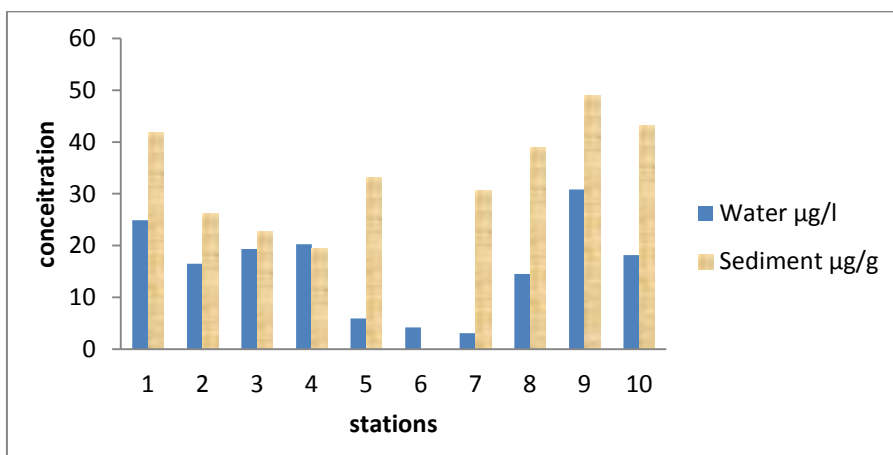


Figure 2. Variation of oil concentration in study area.

Table (2) and Figures (3 and 4) represent the variation of hydrocarbons concentrations in water during the period 1980 to 2012. Petroleum is a mixture of hundreds of different hydrocarbon compounds, the composition of petroleum products released into the environment is variable. Al-Saad (1995) also reported that many aquatic organisms of Shatt Al-Arab river including plants, algae, zooplankton, bacteria and fish were capable to synthesize biogenic hydrocarbons. While in the environment, petroleum composition was further influenced by volatilization, leaching and biological degradation. Traditional shipping and oil transportation routes are more exposed to the impacts of oil-polluted discharges from tankers and other vessels than other areas. Water containing  $1 \mu\text{g/l}$  of oil residue was considered not polluted, that containing between 1 and  $100 \mu\text{g/l}$  was moderately polluted, while that containing  $100 \mu\text{g/l}$  was highly polluted. The study area lies in the second category. The recorded levels of hydrocarbons in water samples in this study are lower than most reported hydrocarbons levels by the previous studies.

Table 2. Concentration of Petroleum Hydrocarbons in water ( $\mu\text{g/l}$ ) from southern Iraqi water.

Shatt Al-Arab River	Shatt Al-Arab Estuary	Khor Al-Zubair & Umm Qasr	Khor Abdullah	Regional Iraqi Waters	References
12.0-86.7				2.7- 68.0	DouAbul, 1984
5.2-14.2					DouAbul & Al-Saad, 1985
6.5-23.5					Al-Saad & Bedair, 1989
4.0-14.0	6.0-7.0			2.6-3.7	Al-Saad <i>et al.</i> , 1995
			0.9-9.6	1.0-6.0	Al-Imarah <i>et al.</i> , 1995
1.3-35.0					Al-Saad, 1998
4.2-8.6					Talal, 1999
		4.6-22.6		1.0-15.0	Al-Timari <i>et al.</i> , 2002
2.5-47.0	31.0	0.99-23.0	44.0-75.0		Al-Timari <i>et al.</i> , 2003
		15.0-46.0	12.0-51.0	5.0-40.0	Nasir, 2005
3.97-11.72					Ibraheem, 2004
5.67- 9.48					Ali, 2006
2.3-50.2					Hantoush, 2007
		36.8-47.82			Al-Saad <i>et al.</i> , 2008
14.51-30.81	3.09-14.51	24.90-16.53	19.34	3.09-4.18	Present Study

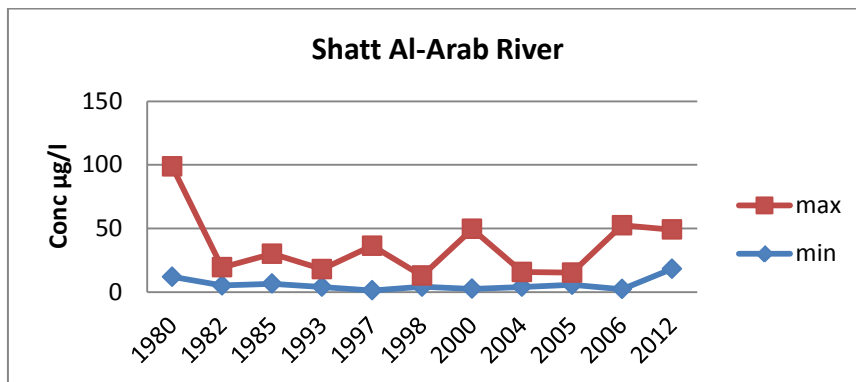


Figure 3. Variation of oil concentration in water from Shatt Al-Arab River, 1980-2012.

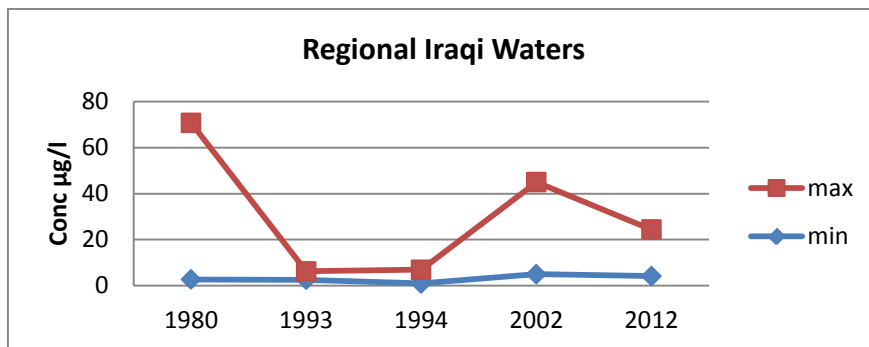


Figure 4. Variation of oil concentration in water from Regional Iraqi Waters, 1980-2012.

Hydrocarbons in the water body could be associated with sediments and accumulated in the bottom soil along the water body. In the present case, both dilution and adsorption of the hydrocarbons to suspended particulate matter with the subsequent sinking or over bank deposition are the main factors. Particulate material plus the adsorbed oil, will be deposited at the sediment bank or sink to the bottom as the water current was reduced in speed. Such considerable movement of water masses have a great transport capacity, resulting in dilution oil with the speed from the source of spill to a negligible amount down the North West Arabian Gulf. Hydrocarbons may be carried to the sediments adsorbed onto clay and silts which settle out of the water column (Al-Saad and Al-Timari, 1989).

The Table (3) and Figures (5 and 6) represent the variations of oil concentration in sediment samples during the same period 1980-2012. Total hydrocarbon concentrations >500 µg/g are generally indicative of significant pollution, values <10 µg/g are considered to denote unpolluted sediments (Volkman *et al.*, 1992). The TPH levels in sediments reported here are relatively low compared to those at worldwide locations classified to be chronically contaminated by oil, such as 60-646 µg/g in Hong Kong's Victoria Harbour (Hong *et al.*, 1995).

Table 3. Concentration rang of Petroleum Hydrocarbons in sediment from southern of Iraq.

Concentrations of Petroleum Hydrocarbons in Sediments (µg/g)					Reference
Shatt Al-Arab River	Shatt Al-Arab Estuary	Khor Al-Zubair & Umm Qasr	Khor Abdullah	Regional Iraqi Waters	
2.6-44.0	26.0-40.0	3.5-5.0	3.6-22	0.4-24	DouAbul <i>et al.</i> , 1984
		3.7-26.0			Al-Hamdi, 1989
			1.4-1.7	0.2-2.8	Al-Imarah <i>et al.</i> , 1995
9.7-38.0	10.7-23.0			5.7	A-Saad <i>et al.</i> , 1995
				2.4-5.8	Al-Saad <i>et al.</i> , 2000
		21.0-178.0			Al-Timari <i>et al.</i> , 2002
		99.0-192.0	51.0-191.0	16.3-139.0	Nasir, 2005
59.5-148.4					Al-Imarah <i>et al.</i> , 2007
28.8-275.4					Hantoush, 2007
		2.1-45.9			A-Saad <i>et al.</i> , 2008
43.16-49.09	30.73-38.98	41.92	22.74-26.27	19-43-33.12	Present Study

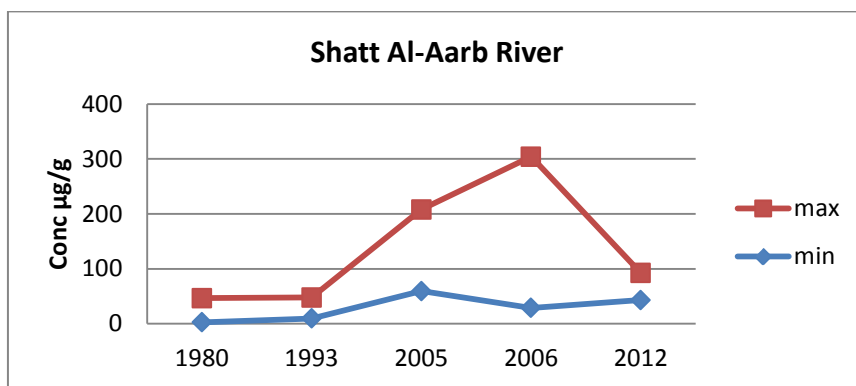


Figure 5. Variation of oil concentration in sediment from Shatt Al-Arab River, 1980-2012.

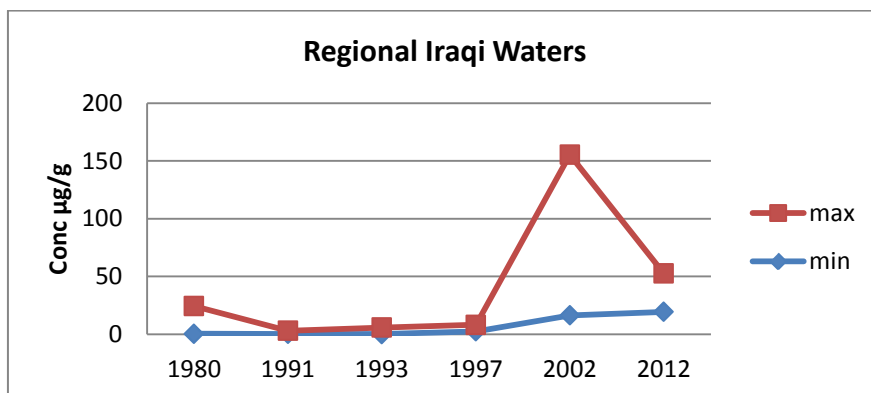


Figure 6. Variation of oil concentration in sediment from Regional Iraqi Waters, 1980-2012.

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## واقع التلوث النفطي في مياه ورواسب مصب شط العرب وشمال غرب الخليج العربي

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**المستخلص** - تم تحليل عينات مياه ورواسب من عشر محطات على طول مصب شط العرب والشمال الغربي من الخليج العربي، وذلك باستخدام تقنية الفلورة في محاولة لتقدير مستويات المخلفات الهيدروكربونية النفطية. أشارت النتائج إلى وجود درجة من التلوث النفطي، ومع ذلك، لا يزال أقل في القيم مقارنة مع الوضع في بعض المواقع الإقليمية الأخرى. المستويات التي تم الحصول عليها من الهيدروكربونات النفطية في المياه تراوحت من 3.09 مايكروغرام/لتر في محطة (7) إلى 30.87 مايكروغرام/لتر في محطة (9) في حين تراوحت في الرواسب من 19.43 مايكروغرام/غم وزن جاف في محطة (4) إلى 49.09 مايكروغرام/غم وزن جاف في محطة (9). لإعطاء تقييم أفضل لمستويات الهيدروكربونات النفطية في الرواسب، تم تحليل الكربون العضوي الكلي والحجم الحبيبي للرواسب. تراوحت تقديرات الكربون العضوي الكلي من 0.01% في محطة (4) إلى 1.34% في محطة (9). أشارت متوسطات القيم التي تم الحصول عليها في هذه الدراسة إلى أن مستويات المواد الهيدروكربونية في عينات الرواسب هي أقل من المستويات التي تم الحصول عليها مع منهجية مماثلة وتحليلات في بعض من دول الخليج والدول في المنطقة القريبة من منطقة الدراسة. قد تأتي مصادر التلوث من الناقلات النفطية والسفن، التصريفات والنفايات السائلة من مصافي النفط ومحطات توليد الكهرباء والأنشطة الصناعية والبشرية في مدينة البصرة.