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Seasonal Distribution of Dissolved and Particulate Hydrocarbons in Shatt Al-Arab Estuary and the North-West Arabian Gulf

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Oil refinery effluent and losses during loading operations have been identified as the major sources of oil contamination in the waters of the Shatt Al-Arab River which empties into the North-West Arabian Gulf (Bedair and Al-Saad, 1992; Al-Saad *et al.*, 1995). Estimates suggest that this river transports about 48 tonnes of oil residues to the Arabian Gulf annually (DouAbul and Al-Saad, 1985). The Arabian Gulf is also an extremely busy shipping route for oil, with accidental spillage being almost unavoidable. In combination, these sources provide a long-term input of petroleum. Major spills, these being either unintentional or as a consequence of military activities, have added occasional dramatic pulses of oil contamination to the long-term background (Price *et al.*, 1993).

This study constitutes part of an ongoing project on the origin, role and fate of hydrocarbons in the Shatt Al-Arab estuary and North-West Arabian Gulf, and also provides data on seasonal changes of dissolved and particulate hydrocarbons. The study also provides a baseline against which any changes that might occur in the future may be determined.

Seven sampling sites were selected in the Shatt Al-Arab estuary and North-West Arabian Gulf for the study of hydrocarbons in the dissolved and particulate phases. The sampling programme was completed between June 1993 and July 1994. Water samples (50 l) were obtained from a depth of approximately 1 m in well-washed glass aspirator bottles, at each station (Fig. 1).

The water samples were suction filtered within 5 h through pre-weighed 0.45 μm pore Whatman GF/F glass fibre filters. Material passing through the filter was considered as 'dissolved', that retained being assigned to the 'particulate' phase. Dissolved hydrocarbons were solvent extracted following the procedure of UNESCO (1976). A total of 100 ml of nanograde carbon tetrachloride (CCl_4) was used in two successive

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50 ml extractions, and the extracts were combined. To the combined extracts, anydrous sodium sulphate was added to break up any emulsion and remove excess water. The CCl_4 extracts were reduced in volume to less than 5 ml using a rotary evaporator. The reduced extracts were pipetted carefully into precleaned 10 ml volumetric flasks, making sure any residual particles of sodium sulphate were excluded, and evaporated to dryness in a stream of pure nitrogen. The flasks were then rinsed with fresh hexane and made up to 5 ml prior to ultraviolet fluorescence (UVF) analysis.

Before extracting suspended particulate matter, the particulate fractions retained on the filters were freeze-dried. The extraction procedure was based upon that of Goutx and Saliot (1980). The filters were placed in a pre-extracted cellulose thimble and soxhlet extracted with 150 ml of methanol:benzene (1:1) for 24 h. The extract was then transferred to a storage flask and the sample was further extracted with fresh solvent. The combined extracts were reduced in volume to about 10 ml in a rotary evaporator, and saponified for 2 h with a solution of 4 N KOH in 1:1 methanol:benzene. After removing the unsaponified matter with hexane, the extracts were dried over anhydrous Na_2SO_4 and concentrated by a stream of pure N_2 for UVF analysis.

Quantitative measurement for total petroleum hydrocarbons were made by measuring the emission intensity at 360 nm, with excitation at 310 nm. All blanks, standards and samples were run at identical instrument conditions on a Shimadzu RF-540 spectrofluorometer equipped with a DR-3 data recorder. Procedural blanks consisting of all reagents and glassware used during the analysis were analysed periodically.

The concentrations of petroleum residues dissolved in the waters of the Shatt Al-Arab estuary and North-West Arabian Gulf (expressed as Kuwait crude oil equivalents) are listed in Table 1, with average seasonal and regional concentrations presented in Fig. 2.

The average concentrations of dissolved hydrocarbons varied from 2.60 $\mu g\,l^{-1}$ at station 7 to

TABLE 1

Concentrations of total petroleum hydrocarbons ($\mu g\,l^{-1}$ as Kuwait crude oil equivalents) in the dissolved phase of the Shatt Al-Arab estuary and North-West Arabian Gulf.

Date	Station samples	No. of samples	Concentration Range	Mean	SD
Summer 1993	1	8	3.18–5.62	4.24	0.748
	2	8	10.39–18.92	14.13	2.880
	3	8	4.19–7.20	5.55	1.120
	4	8	5.72–8.01	6.54	0.992
	5	8	2.80–4.86	3.72	0.742
	6	8	2.90–4.10	3.33	0.723
	7	8	1.76–3.42	2.60	0.578
Autumn 1993	1	5	4.84–8.62	6.85	1.662
	2	5	24.91–29.48	26.56	2.058
	3	5	7.40–10.38	9.30	1.425
	4	5	7.31–9.00	8.16	0.807
	5	5	4.11–6.32	5.27	1.010
	6	5	3.16–4.88	3.91	0.717
	7	5	3.00–4.20	3.58	0.526
Winter 1993	1	5	6.26–8.94	7.34	1.012
	2	5	34.60–42.76	38.29	3.442
	3	5	11.82–13.85	12.82	0.965
	4	5	8.20–11.53	9.80	1.226
	5	5	5.29–7.21	6.10	0.715
	6	5	3.69–5.30	4.24	0.631
	7	5	2.93–5.28	3.94	0.973
Spring 1994	1	10	3.40–7.20	5.15	1.317
	2	10	18.89–26.72	22.37	2.697
	3	10	6.00–9.01	7.25	1.246
	4	10	4.82–7.46	6.27	1.108
	5	10	2.18–5.03	3.53	0.795
	6	10	2.45–4.12	3.06	0.639
	7	10	1.98–3.98	2.90	0.611
Summer 1994	1	5	2.96–5.62	3.97	1.020
	2	5	11.16–14.62	12.89	1.246
	3	5	4.85–6.29	5.52	0.662
	4	5	3.93–5.90	4.91	0.752
	5	5	2.75–4.10	3.08	0.693
	6	5	2.10–3.76	2.96	0.788
	7	5	2.18–3.51	2.88	0.501

TABLE 2

Concentrations of total petroleum hydrocarbons ($\mu g\,l^{-1}$) in the particulate phase of the Shatt Al-Arab estuary and North-West Arabian Gulf.

Date	Station samples	No. of samples	Concentration Range	Mean	SD
Summer 1993	1	3	2.88–3.58	3.21	0.350
	2	3	8.88–10.97	9.83	1.056
	3	3	4.86–6.10	5.49	0.620
	4	3	3.10–5.11	4.28	1.063
	5	3	2.55–3.16	2.82	0.310
	6	3	1.98–3.82	2.60	1.053
	7	3	1.67–2.30	1.93	0.329
Autumn 1993	1	3	6.12–8.62	7.18	1.292
	2	3	21.08–24.00	22.37	1.489
	3	3	8.58–9.24	8.90	0.330
	4	3	7.97–8.53	8.32	0.305
	5	3	2.98–4.53	3.73	0.776
	6	3	2.56–3.28	2.94	0.361
	7	3	1.65–2.10	1.89	0.226
Winter 1993	1	3	6.93–10.29	8.25	1.789
	2	3	28.76–32.80	30.60	2.043
	3	3	9.68–11.30	10.65	0.857
	4	3	8.65–10.90	9.80	1.126
	5	3	3.89–5.83	4.84	0.970
	6	3	3.18–4.30	3.81	0.575
	7	3	2.01–3.84	2.93	0.915
Spring 1994	1	3	3.47–5.22	4.24	0.293
	2	3	17.65–20.89	18.83	1.787
	3	3	5.24–7.65	6.33	1.221
	4	3	4.32–6.18	5.19	0.935
	5	3	2.68–3.18	2.91	0.251
	6	3	2.36–3.08	2.64	0.385
	7	3	1.98–2.74	2.18	0.482
Summer 1994	1	3	2.16–4.50	3.07	1.251
	2	3	7.21–9.85	8.76	1.380
	3	3	4.80–5.18	4.97	0.193
	4	3	2.95–4.95	3.92	1.000
	5	3	2.10–3.15	2.61	0.525
	6	3	1.88–3.08	2.36	0.617
	7	3	0.98–1.52	1.22	0.274

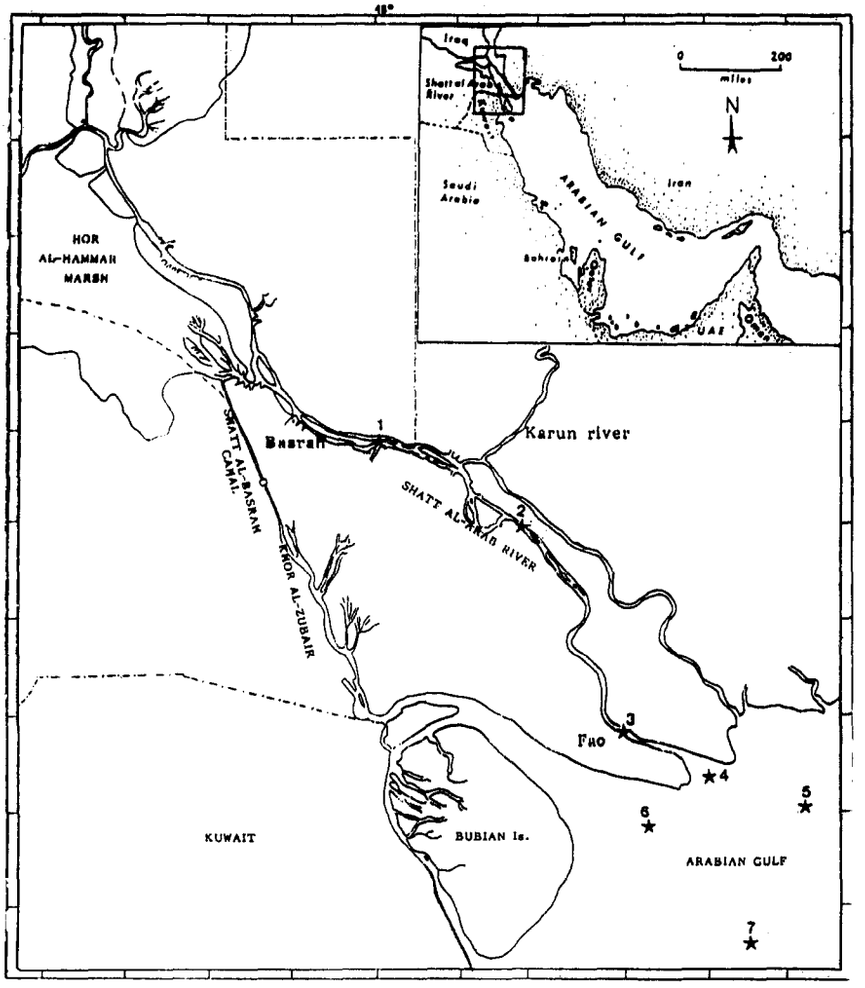

Fig. 1 The sampling locations for the present study.

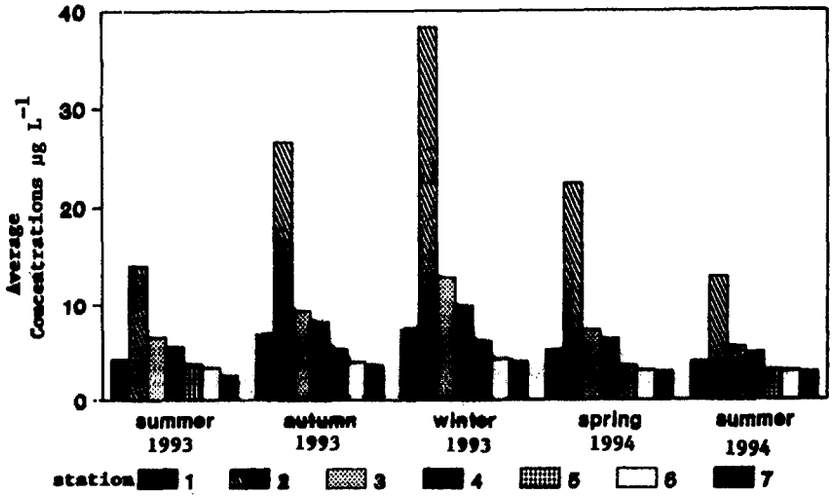

Fig. 2 Seasonal and regional variations of total petroleum hydrocarbons in the dissolved phase of the Shatt Al-Arab River and North-West Arabian Gulf.

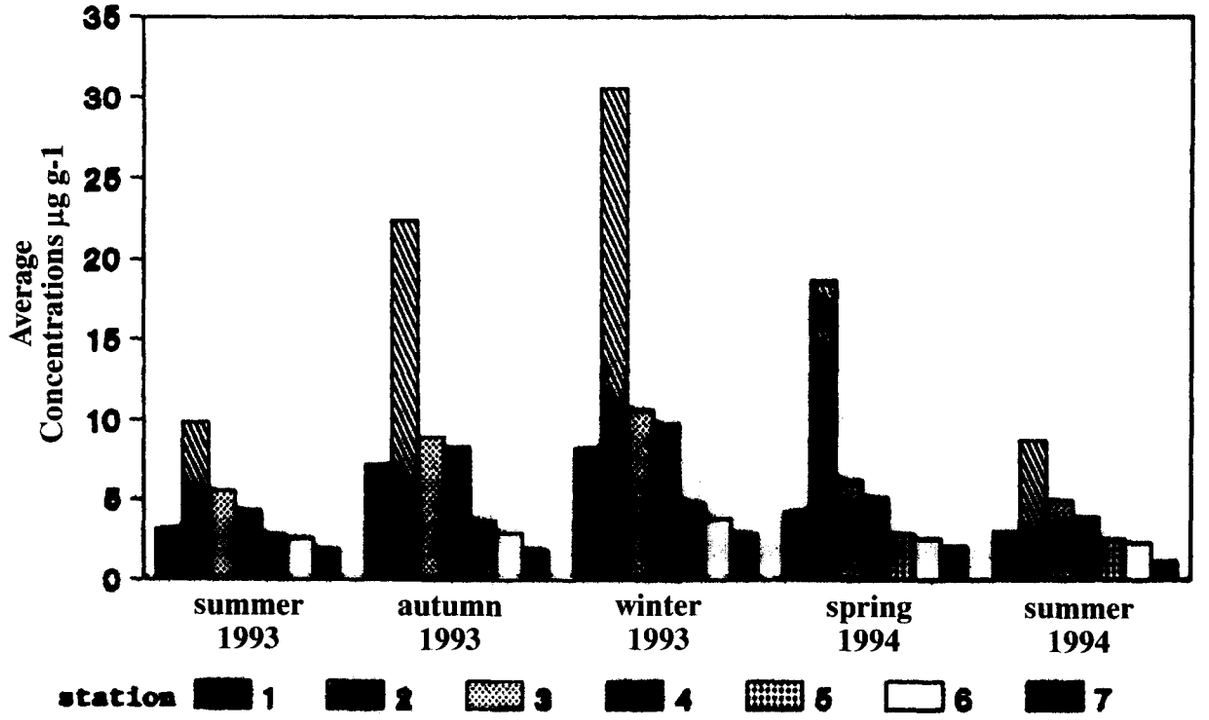

Fig. 3 Seasonal and regional variations of total petroleum hydrocarbons in the particulate phase of the Shatt Al-Arab River and North-West Arabian Gulf.

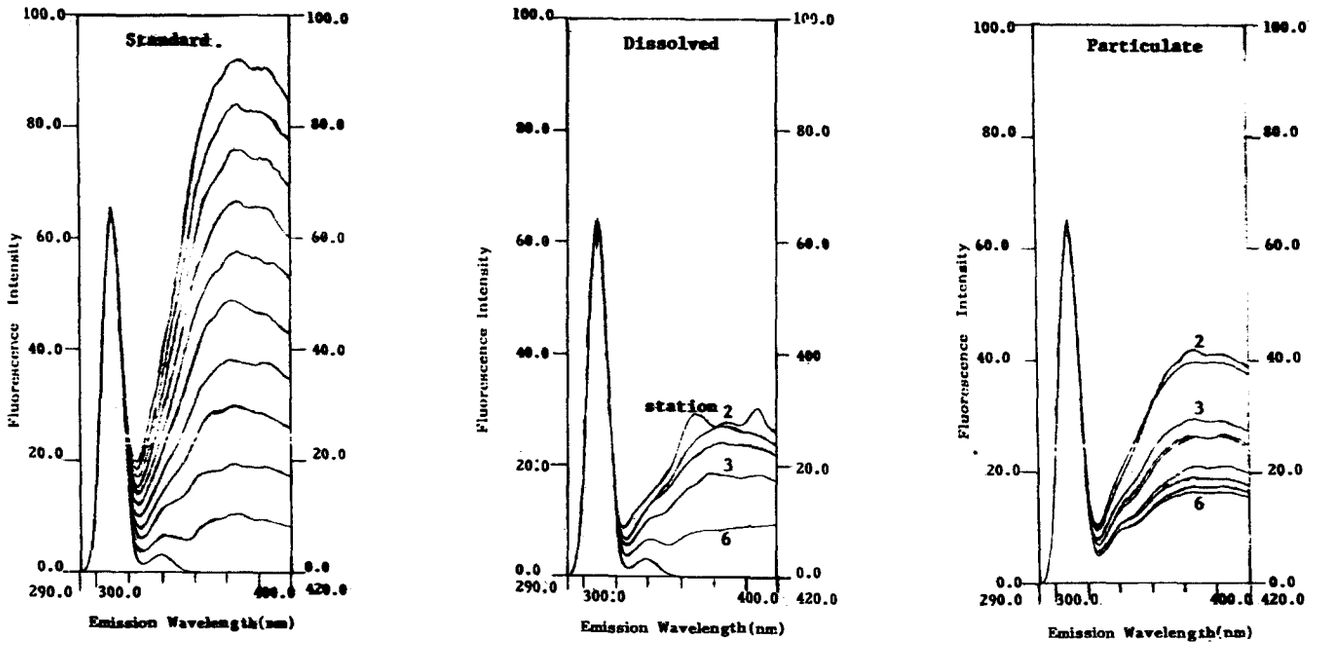

Fig. 4 Fluorescence spectra of the ROPME crude oil standard and of extracts of dissolved and particulate samples from stations 2, 3 and 6.

TABLE 3

Comparison of oil concentrations in coastal and open sea waters, as estimated by UVF spectroscopy.

Location	Concentration ($\mu g\ l^{-1}$)	Authors
Nova Scotia	0.8	Gordan *et al.* (1978)
Southern Baltic Sea	2.0–130	Law and Andrulewicz (1983)
Qatar	1.2–428	El-Samra *et al.* (1986)
Saudia Arabia	4.3–546	El-Samra *et al.* (1986)
Kuwait	2.1–3.6	El-Samra *et al.* (1986)
Winyah Bay (USA)	0.23–25	Bidleman *et al.* (1990)
Gulf of Thailand	1.9–72	Wattayakorn (1991)
UK estuaries	9.3–48	MAFF (1993)
Cortiou (France)	104	Marchand *et al.* (1988)
Gulf of Lyons	18–23	Marchand *et al.* (1988)
Arabian Sea	1.6–11.1	Sen Gupta *et al.* (1993)
Shatt Al-Arab River	5.6–14.2	Al-Saad (1985)
Shatt Al-Arab and NW Arabian Gulf	2.7–86.7	DouAbul (1984)
Arabian Gulf	3.5	Erhardt and Burns (1993)
Shatt Al-Arab and NW Arabian Gulf	3.25–25.33	Present study

14.13 $\mu g\ l^{-1}$ at station 2 during summer periods, and from 3.94 $\mu g\ l^{-1}$ at station 7 to 38.2 $\mu g\ l^{-1}$ at station 2 during winter. In general, the concentrations of petroleum residues in the dissolved phase were much higher in winter than in summer.

A summary of the concentrations of hydrocarbons in the particulate phase is shown in Table 2, with seasonal and regional averages being presented in Fig. 3. The average concentrations of hydrocarbons in the particulate fraction varied from 1.22 $\mu g\ l^{-1}$ at station 7 to 9.83 $\mu g\ l^{-1}$ at station 2 during summer, and from 2.93 μg^{-1} at station 7 to 30.60 $\mu g\ l^{-1}$ at station 2 during winter. The highest concentrations were found in all seasons at station 2 near the Abadan oil refinery, while the lowest concentrations were observed at station 7 in the open waters of the Arabian Gulf. Oil concentrations were substantially higher in winter than in spring or summer.

The higher concentrations of petroleum hydrocarbons in the Shatt Al-Arab River relative to those in the open water of the Arabian Gulf imply that the contribution of oil through shipping activities is less significant than that of the river discharges. Most samples exhibited their maximum fluorescence at 340–380 nm, and this region is believed to be derived primarily from diesel oil which is commonly used in small transport and fishing boats (Al-Saad and Al-Timari, 1993; Wattayakorn, 1991).

Spectra for the higher concentrations found near oil refineries on the banks of the Shatt Al-Arab River were similar to those of lubricating and fuel oils (Fig. 4). Petroleum hydrocarbons in the estuary are likely to originate from boating activities, runoff, and introduction via sewage outfalls (DouAbul and Al-Saad, 1985; Al-Mudaffar *et al.*, 1990; Bedair and Al-Saad, 1992).

Apart from station 2, the concentrations of hydrocarbons observed in this study were either lower than, or similar to, those found in other surface waters (Table 3).

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