Total Aromatic Hydrocarbon Content in the Muscle and Liver Lipid Extracts of Two Seabream Fishes from the Arabian Gulf

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ABSTRACT. Concentrations of total aromatic hydrocarbons (TAHC) were determined in two seabream fishes from the Family Sparidae, longspine seabream, Argyrops spinifer (Kofar) and Mylio bifasciatus (Faskar) from the eastern and southeastern coats of Qatar. Levels of total hydrocarbons in the muscle and liver tissues of Argyrops sp. were lower than those in Mylio sp. Variations in TAHC with size were very slight in Argyrops sp. while Mylio sp. showed significant variations in both muscle and liver tissues.

The hydrocarbons contained, in the two fishes displayed, high aromatic concentrations correlated with increasing fat content. Mean concentrations of total aromatic hydrocarbons in the muscle and liver tissues of *Argyrops* sp. were 24.7 and 23.7 μgg^{-1} while in *Mylio* sp. they were 53.4 and 41.1 μgg^{-1} wet weight chrysene equivalents respectively.

In recent years, particularly from the middle of the 6th decade, scientists, governments and international organizations have become more concerned as to the distribution, effects and fate of petroleum derived hydrocarbons in marine ecosystems. The estimated amounts of petroleum entering the waters of the Arabian Gulf in 1976 following successive losses were 2,000,000 tons (Research Planning Institute, 1977), while the input into the Mediterranean Sea was in the order of 1,000,000 tons (UNEP, 1982). Recent investigations in the northwestern part of the Arabian Gulf showed high pollution by total hydrocarbons compared to other marine environments with mean concentrations of 2.95, 4.14 and 3.29 µgl⁻¹ in the sea water off Kuwait, Saudi Arabia and Qatar respectively (El Samra *et al.* 1986). High concentrations of 546.4 and 428 µgl⁻¹ were recoded adjacent to pipelines of oil production and exportation centres at Ras Tanura and Ju'aymah, Saudi Arabia and at the northern coast of Qatar.

Natural hydrocarbons are found in most plants and animals in both terrestrial and aquatic environments. Marine organisms absorb oil, but there is a little evidence of concentration or accumulation in their tissues. Some analytical methods are unable to distinguish between hydrocarbons from the two sources. Aromatic hydrocarbons were found to be taken up *via* the gills of the marine fishes tested by Lee *et al.* (1972). Metabolism in the liver is followed by transfer of hydrocarbons and metabolites to the bile and finally excretion. When petroleum hydrocarbons are taken up by an organism they may be extracted unchanged, metabolised, or stored with possible elimination at a later date (UNEP, 1977).

The oil pollution problem in the Gulf region and the absence of a data base for petroleum residues in fish tissues gives a high priority to serious investigations about the levels of petroleum-derived hydrocarbons in the fish meal.

Dispersant chemicals, which remove oil and treat minor oil spills from the sea surface are rarely used in Qatar (Table 1). This is important, since the use of dispersants facilities the uptake of oil by organisms (UNEP, 1982).

Table 1. Dispersant usage in Qatar offshore waters, 1979-1984, with number of incidents, amount and type of dispersant used.

Year	Number of incidents	Frequency of dispersant used	Type of dispersant	Amount used	
1979	7	3	LTX	6200 liters	
1980	5	4	LTX plus unknown made from ship	400 litres	
1981	3	1	LTX	2600 litres	
1982	1	Nil	Nil	Nil	
1983	3	2	Servo 2000 plus LTX	600 litres	
1984	Nil	Nil	Nil	Nil	

^{*} Data obtained from Qatar General Petroleum Corporation (offshore operations) Doha, 1985.

The present, preliminary, work describes laboratory investigations undertaken to estimate current levels of total aromatic hydrocarbons (TAHs) in the muscle and liver tissues of two commercially consumable fishes belonging to Family Sparidae from Qatari waters in the Arabian Gulf. Such a study could enable us to assess the effects of oil pollution on the biochemical composition of the edible fishes in the Gulf.

Materials and Methods

Two species of edible fishes from the Family Sparidae were selected for this study. Sixty eight specimens, comprising forty Argyrops spinifer (Kofar) and twenty eight Mylio bifasciatus (Faskar), were obtained fresh from Doha City Fish Market during the winter season (December 1985, January-February 1986). Total body weight and standard length for each individual were recorded. Specimens of mixed sex and sizes, were grouped into five size classes according to their mean standard length, and the reproductive status of the fish groups, particularly females were shown in Tables 2 and 3. Samples, washed, dried at 80°C, were finely ground and stored in clean dry glass tubes with covers. Moisture percentage of every specimen was calculated. One gram dry weight of the muscle and liver samples was used in the analysis and then recalculated for the corresponding wet weight. The lipid and total aromatic hydrocarbon content were extracted from each sample following the method described by Warner (1974) with the modification that Soxhlet extraction apparatus was used in place of the cold extraction procedure described by Warner. The same solvent (aromatic free diethylether) was used in the extraction steps which continued for more than 16 hrs. The diethylether lipid extract was then evaporated to a volume Ca. 30 ml. in a rotary evaporator. Clean up procedure of the hydrocarbon extract was facilitated by eluting the lipid extract from the top of a 0.9 cm diameter glass column, containing 20 cm deactivated silica gel covered with 1g. anhydrous sodium sulphate. Lipid content was determined gravimetrically by evaporating a half portion of the extract to dryness. The purified, moisture free, hydrocarbon extract was reevaporated to dryness in a rotary evaporator then dissolved in 10 ml of n-hexane (pesticide grade). The intensity of fluorescence of hydrocarbon extracts was measured using a Spectrofluorometer (Turner model 430). Calculation of TAHC in the samples as chrysene, Kuwait and Qatar (Dukhann) crude oil equivalents was achieved using the formula:

$$TAHC = \frac{C X V}{W}$$

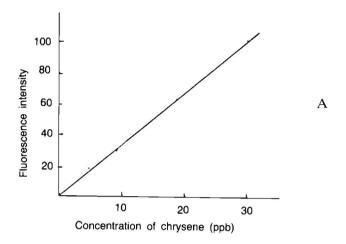
where TAHC = concentration of total aromatic hydrocarbons in the fish tissue in μgg^{-1} wet weight.

 $C = Concentration of hydrocarbons in sample extracts in <math>\mu g ml^{-1}$

V = volume of extract in ml.

W = weight of the wet fish tissue in gram.

The standard curves used for chrysene, Kuwait and Qatar crude oils are shown in Fig. 1.



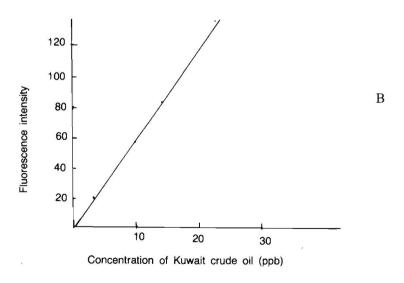
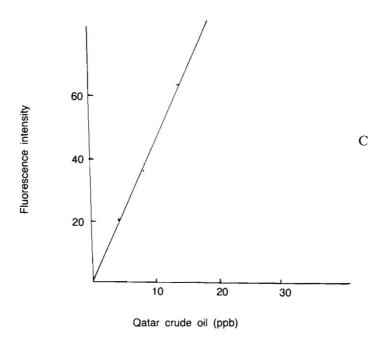


Fig. 1. Standard curves



Results and Discussion

Concentration of total aromatic hydrocarbons (both natural and petroleumderived), and the diethylether - extractable lipid content of muscle and liver tissues in each fish species are presented in Tables 2 and 3 respectively. Linear regression equations were applied to investigate the possible relationships between TAHC and both body length and weight of the two fish species grouped as five size classes. The correlation between standard body length and mean total weight of Argyrops spinifer & Mylio bifasciatus is r = 0.98 and r = 0.97 respectively, a figure considered high for the two species. Correlation between standard body length and TAHC was attempted since the previous parameter rarely changes according to the condition of individual fish. Table 4 presents the correlation coefficients between TAHC in muscle and liver tissues and both standard length, and lipid content. Such correlation revealed significant differences in TAHC between different tissues and sizes of the two fishes. This content appeared high in the muscle and liver of Mylio bifasciatus while it was comparably low in the tissues of Argyrops spinifer, as is indicated by the higher correlation with both the size and lipid content of the former species. The increased concentration of TAHs in the liver of Mylio bifasciatus may be related to their increased lipid content. This agrees with the suggestion of Lee and Benson (1973) which indicated the accumulation of hydrocarbons in the liver of marine fish, since it is generally high in lipid but Argyrops spinifer does not agree.

Table 2. Total aromatic hydrocarbon content in the muscle and liver of five size classes of Kofar, Argyrops spinifer and Faskar, Mylio bifasciatus, from Doha Fish Market; values in μgg⁻¹ wet weight chrysene, Kuwait and Qatar crude (Dukhann) oil equivalents

Argyrops spinifer					
tissue	size & group cm	b weight g	ТАНС		
			chrysene equivalents	Qatar crude oil	Kuwait crude oil
M L	I (15-20)	111 (8)	31.7 28.1	25.1 22.3	19.8 17.6
M L	II (21–25)	232 (10)	16.5 20.2	13.1 16.0	10.3 12.6
M L	III (26-30)	389 (12)	40.3 35.2	32.5 27.9	25.6 22.0
M L	IV (31-35)	610 (6)	13.1 18.9	10.4 15.0	8.2 11.8
M L	V (36-50)	1250 (4)	21.7 16.3	17.2 12.9	13.6 10.2

Mylio bifasciatus

tissue	a size & group cm	b weight g	ТАНС		
			chrysene equivalents	Qatar crude oil	Kuwait crude oil
M L	I (15-20)	109 (6)	55.1 135.3	43.7 107.2	34.5 84.6
M L	II (21-25)	241 (7)	38.6 15.1	30.6 12.0	24.1 9.4
M L	III (26-30)	422 (10)	35.6 19.7	28.3 15.5	22.3 12.2
M L	IV (31-35)	591 (3)	22.8 19.8	18.1 15.7	14.3 12.4
M L	V (36-45)	1330 (2)	114.9 15.6	91.1 12.4	71.9 9.8

a) Size: standard length.

b) Mean total wet weight.

M = Muscle.

L = liver.

^{() =} the number of fish individual in the sample.

Table 3. Lipid content in the muscle and liver of five size classes of Kofar, Argyrops spinifer and Faskar, Mylio bifasciatus in mgg⁻¹ wet weight

Argyrops spinifer				Mylio bifasciatus			
Size and group cm	Tissue	Lipid content	%	Size and group cm	Tissue	Lipid content	%
I (15-20)	M I & II L	8.0 28.0	0.83	I (15-20)	M I & II	13.0	1.29
II (21-25)	M III	9.0	0.94	II (21-25)	M III	6.0	0.65
	L M	27.0	2.71	11 (21 25)	L M	33.0 25.0	3.29 2.51
III (26-30)	L IV	27.0	2.68	III (26-30)	L IV	25.0	2.48
IV (31-35)	M L	18.0 20.0	1.83 1.97	IV (31-35)	M V L	2.0 21.0	0.19
V (36-50)	M V	3.0	0.28	V (36-45)	M V	13.0	1.34
. (20 30)	L ·	26.0	2.64		L A	35.0	3.45
reproductive status			l reproductive status				

Table 4. Correlation coefficients for the linear regression equations of the relationship between wet weight TAHC in muscle and liver tissues and both standard length and lipid content of the two fish species

		Correlation coefficient		
Fish species	Tissue	With standard length	With lipid content	
Argyrops sp.	muscle	-0.31	-0.38	
	liver	-0.54	-0.46	
Mylio sp.	muscle	0.52	0.20	
	liver	0.00	0.60	

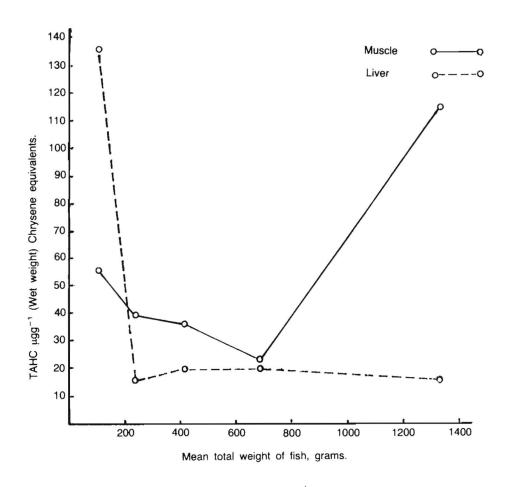


Fig. 2. Variations of hydrocarbon content with weight in muscle and liver tissues of Faskar, Mylio bifasciatus.

It is noticeably that variations in hydrocarbon content with weight in muscle and liver tissues of Argyrops sp. were very slight, ranging between 13.1-40.3 and $16.3-35.2~\mu gg^{-1}$ wet weight chrysene equivalents respectively. On the other hand, values between 22.8-114.9 and $15.1-135.3~\mu gg^{-1}$ wet weight chrysene equivalents were determined in the tissues of Mylio sp. (Fig. 2). In spite of these apparently high levels they are lower than those recorded for the flesh of the estuarine fish Callionymus~lyra~found in European and North American waters, which were

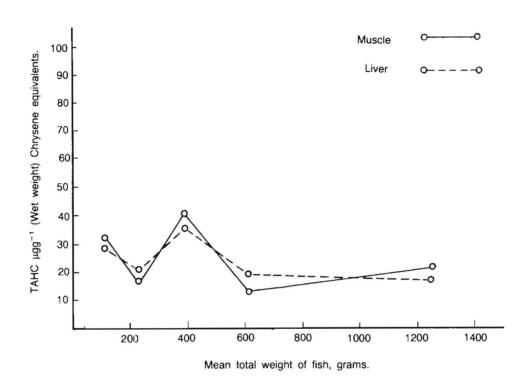


Fig. 3. Variations of hydrocarbon content with weight in muscle and liver tissues of Kofar, *Argyrops spinifer*.

123.1–216.0 ppm wet weight respectively (Burns and Teal 1973 and Farrington *et al.* 1972, 1973). The pelagic Tuna fish showed lower hydrocarbon concentrations $(45.5-51.5) \, \mu gg^{-1}$ wet weight (Farrington *et al.* 1974) than *Callionymus lyra* and higher than that of *Argyrops spinifer* in the present study.

The accumulation of petroleum hydrocarbons in soft tissues of marine organisms is prominent in benthic invertebrates, particularly oysters. Blue Mussels (*Mytilus edulis*) from a location in the Kiel Bight (Federal Republic of Germany), contain fossil hydrocarbons in concentrations somewhat above the natural background of recent biogenic hydrocarbons, at 2.18 mgg⁻¹ of wet tissue (Ehrhardt and Heinemann, 1974). This is probably due to relatively little obvious pollution by fossil fuels. Lower maximum concentrations of 300 µgg⁻¹ dry weight

were recorded in the oyster, *Pinctada margaratifera* by Anderlini and Co-workers (Anderlini *et al.* 1981). However, there was no discussion as to whether or not the levels in *Mytilus* might be considered high.

The concentration of total aromatic hydrocarbons in the muscles of Mylio sp. showed the highest level in group V (114.9 µgg⁻¹) and the lowest in group IV (22.8 µgg⁻¹) indicating very high correlation with the lipid content, where it was 13 & 2 mg/g wet weight respectively. The same trend was observed in liver tissues where the maximum (135.3 µgg⁻¹) was determined in group I (lipid content 39 mg/g wet weight). Biochemical composition of fishes generally shows that lipid concentration ranges between 0.1 and 24% (Mohammed et al. 1967). The concentration of lipids in the two species studied varies between 0.19 and 2.51% for the muscle tissues and 1.97 and 3.89% for the liver, indicating that both fishes are generally "nonfatty". The accumulation of hydrocarbons in the liver of Mylio sp. group I, may be due to its high fat content, and to physiological characters of juvenile fishes, which leads to active metabolism of hydrocarbons in the liver and then elimination with increasing size and age of the fish. The high levels in the muscle of group V in this species might be due to a tendency to concentrate aromatic hydrocarbons during this period of growth, although it is below the concentrations estimated for other fishes by Burns and Teal (1973) and Farrington et al. (1972-1973). This seems to emphasise the view (Whittle et al. 1974) that in fish with a herring-type lipid metabolism some dietary hydrocarbons are quickly assimilated in the muscle, whilst fish with a cod-type lipid metabolism do not assimilate dietary hydrocarbons in the muscle but, instead, in the liver, and with some discrimination.

Marine fish and shellfish tend to concentrate polynuclear aromatic hydrocarbons within their tissues when exposed to oil, but do not retain these levels indefinitely (GESAMP, 1982).

The present preliminary data in this study indicate that levels of hydrocarbon concentrations in the tissues of the two fish species are rather low (Fig. 3). The relatively high values found in *Mylio bifasciatus* compared to *Argyrops spinifer* could be attributed to its relatively higher lipid content and possibly to other physiological characters. The activities and extensive operations for combating oil pollution in the Gulf region, particularly in Kuwait and Saudi Arabia, in spite of using measures which facilitate the uptake of oil by marine organisms, keeps the concentration of dissolved and dispersed hydrocarbons in sea water to levels comparable to the minimum rate of uptake by pelagic fish.

Conclusion

Seabream of the Family Sparidae, Argyrops spinifer and Mylio bifasciatus from eastern and southeastern Qatari waters contain low concentrations of total aromatic hydrocarbons. The coincidence of TAHC levels in muscle and liver tissues of Argyrops spinifer indicated that its origin is likely to be biogenic. The technique and method of analysis does not allow differentiation between biogenic and petroleum-derived hydrocarbons, and it is recommended that investigations continue using other techniques in order to achieve the values for non-biogenic hydrocarbons.

At present, it can be concluded that our tested fishes contain lower levels of total hydrocarbons than those reported in other marine fishes from European and North American waters.

References

- Anderlini, V.C., Al-Harmi, L., De Lappe, B.W., Riesenbrough, R.W., Walker II, W., Simineit, B.R. and Newotons, A. (1981) Distribution of hydrocarbons in the oyster *Pinctada margaratifera*, along the coast of Kuwait. *Marine Poll. Bull.* 12: pp. 57-62.
- Burns, K.A. and Teal, J.M., (1973) Hydrocarbons in the pelagic sargassum community. Deep Sea Research 20: 207-211.
- Ehrhardt, M. and Heinemann, J. (1974) Hydrocarbons in blue mussels from the Kiel Bight. National Bureau of Standards (U.S.A.) Spec. Publi. 409, Marine Pollution Monitoring (petroleum), pp. 221-225.
- El Samra, M.I., Emara, H.I., and Shunbo, F. (1986) Dissolved petroleum hydrocarbons in the Northwestern Arabian Gulf. *Marin Poll. Bull.* 17(2): 65-68.
- Farrington, J.W., Giam, C.S., Harvey, G.R., Parker, P. and Teal, J. (1972) Analytical techniques for selected organic compounds. Marine Pollution Monitoring, Strategy for a National Program. NOAA, U.S. Department of Commerce, Washington, DC, pp. 152-176.
- Farrington, J.W., Teal, J.M., Quinn, J.G., Wade, T., and Burns, K. (1973) Intercalibration analysis of recently biosynthesized hydrocarbons and petroleum hydrocarbons in some lipids, Bull. Environ. Contam. Toxicol. 10: 129-136.
- Farrington, J.W., Teal, J.M., Quinn, J.G., Parker, P.L., Winters, J.K., Wade, T.L., and Burns, K. (1974) Analysis of hydrocarbons in marine organisms: Results of Iode Intercalibration exercises. National Bureau of Standards Spec. Publi. 409, Marine Pollution Monitoring (petroleum), (U.S.A.), pp. 163-166.
- GESAMP (1982) The health of the oceans. In UNEP Regional Seas Reports and Studies, No. 16: 111p. Lee, R.F., Saureheber, R. and Benson, A.A. (1972) Petroleum hydrocarbons: uptake and discharge by the marine mussel, Mytilus edulis, Science, N.Y. 177: 344-346.
- Lee, R.F. and Benson, A.A. (1973) In Preliminary report on the state of pollution of the Mediterranean Sea. UNEP/IG.11/INF. 4,9 December 1977. Restricted distribution, pp. 95-168.
- Mohammed, M.S., Hussien, M.F. and Hassan, Y.M. (1967) Fish Technology Textbook. Scientific Library, Dar Al-Maaref, Cairo. "in Arabic". 569 p.
- Qatar General Petroleum Corporation (1985) Report submitted to ROPME Working Group Meeting, Kuwait 4th March, 1985. Unpublished report.

- Research Planning Institute (1977) Oil pollution in the Arabian Gulf. A preliminary summary. Plivate study undertaken for Kuwait Engineering Operation and Management Co., Kuwait.
- UNEP (1977) Preliminary report on the state of pollution of the Mediterranean Sea, UNEP/IG.11/INF. 4: 209 p.
- Warner, J.S. (1974) Quantitative determination of hydrocarbons in marine organisms, National Bureau of Standards (U.S.A.) Spec. Publi. 409, Marine Pollution Monitoring (petroleum), pp. 195-196.
- Whittle, K.J., Mackie, P.R., Hardy, R. and McIntre, A.D. (1974) The fate of n-alkanes in marine organisms, ICES CM 1974/E: 33.(mimeo).

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المحتوى الكلي للهيدروكربونات الأروماتية في مستخلصات الليبيدات من لحم وكبد نوعين من أساك عائلة السباريدي من الخليج العربي

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تم تقدير تركيزات الهيدروكربونات الأروماتية الكلية في نوعين من أسماك عائلة السباريدي، ذات القيمة التسويقية العالية هما سمكة الكوفر «سمكة البريم البحرية ذات الشوكة الطويلة» وسمكة الفسكر المصادة من السواحل الشرقية والشمالية المشرقية لدولة قطر. وقد تبين أن مستوى الهيدروكربونات الكلية المخزونة في لحم وكبد سمك الكوفر أقل مما في أسماك الفسكر، وأن التغيرات «التنوع» في المحتوى الكلي للهيدروكربونات الأروماتية مع اختلاف حجم الأسماك طفيفاً جداً في سمكة الكوفر، بينها أظهرت سمكة الفسكر تغيرات ذات دلالة خاصة في كل من أنسجة اللحم والكبد.

وقد تبين من الدراسة أن تركيز الهيدروكربونات الأروماتية الموجودة في كلا النوعين من أسهاك هذه العائلة يزداد بزيادة محتوى الدهن لأنسجة هذه الأسهاك. كها تبين أن مستوى تركيز الهيدروكربونات الكلية في هذين النوعين أقبل من تركيزه في بعض أنواع الأسهاك الموجودة في المياه البحرية لبعض الدول الأوروبية وشهال أمريكا. وقد بلغ متوسط تركيز الهيدروكربونات الأروماتية الكلية في أنسجة لحم وكبد أسهاك الكوفر ٧٤,٧ و ٢٣,٧ ميكروجرام / جم على أساس الوزن الرطب، بينها وصل هذا المتوسط في أنسجة سمكة الفسكر إلى ٤١,٥ و ١,١١ ميكروجرام / جم على أساس الوزن الرطب، وقد جم على أساس الوزن الرطب، ينها القولي. وقد حسبت هذه التركيزات على أساس ما يكافئها من مادة الكريسين الأروماتية ذات الوزن الجزيئي العالى.