BIOACCUMULATION OF CHROMIUM, NICKEL, LEAD AND VANADIUM IN SOME COMMERCIAL FISH AND PRAWN FROM QATARI WATERS

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تراكم بعض الفلزات الثقيلة في الأسماك الاقتصادية والروبيان من المياه القطرية

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تم عن طريق الصيد التجريبي جمع ١٥٠ عينة من الأسماك الاقتصادية والروبيان خلال عامي ١٩٩٥/١٩٩٦ من ثلاثة مواقع تغطي المياه القطرية (السافلية – رأس لفان ومسيعيد) وتحليل محتواها من فلزات الكروم، الرصاص، النيكل والفانديوم. أوضحت النتائج ارتفاع طفيف في تركيز تلك الفلزات في الروبيان عنها في الأسماك خاصة في المواقع المتأثرة بالصرف الصناعي. أظهرت الاسماك القاعية قدرة مرتفعة على مراكمة الفلزات عن الأنواع البلاحية، بينما كانت معظم القيم التي تم الحصول عليها مقاربة لتلك المسجلة سابقا في المياه القطرية والخليج العربي. ارتفعت قيم الرصاص في بعض العينات لتقترب من الحد المسموح به دولياً لتناول المأكولات البحرية. رغم التحقق من أن معدل الاستهلاك الحالي للاسماك والروبيان في قطر لايزال أمن لمستهلكي المأكولات البحرية فان وضع برنامج رصد دائم لمتابعة تركيزات الفلزات في تلك الكائنات أصبح ضرورة .

Key Words **:** Heavy metals, bioaccumulation, fish, prawn, Qatar, Arabian Gulf.

ABSTRACT

A total of 150 fish and prawn samples representing the most economically important species were collected during 1995-96 by experimental fishing in three locations covering the coastal waters of Qatar. Fish and prawn flesh were analysed for the oil related metals Cr, Pb, Ni and V. The range of levels in fish were 0.06 - 0.64 $\mu g\ g^{-1}$, 0.03 - 0.93 $\mu g\ g^{-1}$, 0.31 - 2.23 $\mu g\ g^{-1}$ and ND - 0.232 $\mu g\ g^{-1}$ for Cr, Pb, Ni and V, respectively;

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while in prawn corresponding values were 0.3 - 0.99 $\mu g\ g^{-1}$ for Cr, 0.88 - 2.9 $\mu g\ g^{-1}$ for Pb, 1.14 - 3.63 $\mu g\ g^{-1}$ for Ni and 0.071 - 0.531 $\mu g\ g^{-1}$ for V indicating higher concentration of metals in prawn compared to fish. Fish and prawn collected opposite to the industrial area of the country sustained high metal concentrations. *E. tauvina* revealed a higher tendency for accumulating metals than other fish species. Levels of Pb in some fish specially those collected opposite to the industrial area are close to the upper permissible limit published by International organisations, concerned with healthy food. Based on the provisional tolerable weekly intake of metals, an increased number of fish meals/week (more than 8) could lead to a problem with vanadium.

INTRODUCTION

Marine fish, in particular bony fishes, are of high economic importance and a major source of protein in the Arabian Gulf. About 35,000 tonnes of fish by catch are available in the Arabian Gulf [1]. Being a peninsula, surrounded by seawater along it extended coasts fisheries are of prime importance to Qatar's economy.

The Department of Fisheries in Qatar [2] reported that the total catch in Qatar during 1995 was 4271 tonnes. About 150 fish species, belonging to 50 families are currently recorded in Qatari waters [3]. According to the annual fish landing the most important fishes belong to family Clupeldae (25%), Chirocentridae (11.6%), Sparidae (9.6%), Gerreidae (8%), Lethrinidae (6.4%), Carangidae (5%), Platycephalidae (2%), Sphyraenidae (2%), Serranidae (2%) and Siganidae (2%). The prawn *Penaeus semisulcatus* is also considered among the most common market economic crustacean species landed in Qatar and the Arabian Gulf.

Metals are common contaminants in the nearshore marine environment adjacent to industrial activities and are ubiquitous in sewage discharge from urban areas. Among the most important oil activity related metals are Cr, Ni, V and Pb. The existence of metal sources in a certain area may lead to the contamination of seafood leading to human health hazard consequences. Marine organisms concentrate many metals from sea water or sediments which are particularly toxic to man. In recent years the concentrations of heavy metals in marine fishes has received much interest. This interest has been directed towards the commercial fishes since high levels of heavy metals in these species represent a potential human health hazard. Other, commercially unimportant species have received little attention.

The present work aims to determine the levels of some heavy metals in selected, economically important fish and prawn collected from the coastal waters of Qatar in an attempt to assess the impact of developing industrial areas as well as increasing oil exploration activities on the accumulation of metals in representative seafood species.

MATERIALS AND METHODS

Samples were collected to represent 3 different areas Ras Laffan, Al-Saflyah and Messaied along the eastern coast of the Qatari peninsula (Figure 1). The sampling areas represent quite different environments. The water of Al-Saflyah (located opposite to Doha city) is influenced by discharge of surface water from urban areas and desalination plant while Ras Laffan is a new developing industrial area subjected to dredging activities and marine traffic. Messaied marine area is located opposite the major industrial complex in Qatar, harbouring fertilisers, steel and petrochemical industrial activities as well as oil terminals through which the Qatari crude is transported all over the world. From these sources several pollutants are discharged to the marine environment, especially toxic heavy metals.

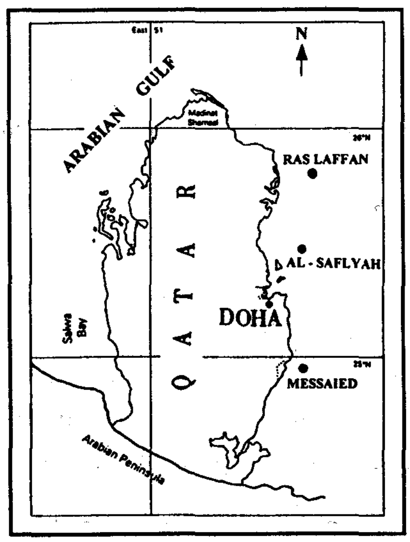

Figure 1. The coastal waters of Qatar showing sampling locations.

Specimens were collected through experimental fishing on board R/V Mukhtabar Al-Bihar during 1995-1996. Fish samples have been collected by gill nets while prawn were sampled using 2 m beam trawl. Targeted fish were mainly those contributing to more than 2% of the catch and are of considerable economic market value compared to other fish species.

Muscle samples were collected from tissues located below the dorsal fin. Dissection was performed using previously nitric acid cleaned plastic forceps and knife. Samples were stored frozen in plastic bags prior to analyses. The total weight and length of each organism were taken. Samples were freeze dried using Model-8 Lab Conco freeze dryer. Samples were identified and prepared for analysis according to the nitric acid wet ashing method described in MOOPAM [4]. Metals concentrations were measured using Model 2380 Perkin Elmer Atomic Absorption Spectrophotometer with HGA 400 Graphite Furnace.

To ensure quality control of metal analyses, the certified Reference Material TORT-2 Lobster Hepatopancreas Reference Material for trace metals from NRC of Canada was analysed with each batch of samples (Table 1). The detection limit of the technique used was 5 ng/g (Pb) 10 ng/g (Cr) 25 ng/g (Ni) and 5 ng/g (V). Recoveries of standard metals addition to the flesh of *E. tauvina* were 92±7% (Pb), 95 ±9% (Cr), 91 ±4% (Ni) and 90 ± 7% (V). The coefficient of variation for replicate samples ranged between 4-11%. No detectable amounts of any metal were found in blanks.

Table 1.
Six Replicate Analyses of Standard Reference Material TORT-2 Lobster Hepatopancreas for Trace Metals

Metal	Certified value ($\mu g\ g^{-1}$ d wt)	Present results ($\mu g\ g^{-1}$ d wt)	Efficiency (%)
Ni	2.5 ± 0.19	2.41 ± 0.34	96
V	1.64 ± 0.19	1.54 ± 0.08	94
Pb	0.35 ± 0.13	0.34 ± 0.09	97
Cr	0.77 ± 0.15	0.79 ± 0.4	102

RESULTS AND DISCUSSION

The mean and range of metals concentrations in the flesh (muscles) of different fish species and prawn collected from Al-Saflyah, Messaied and Ras Laffan areas around Qatar during 1995 - 1996 are presented in Table 2. The most common prawn *P. semisulcatus* appeared to contain the highest levels of metals in all locations compared to other fish species. Maximum mean concentrations reached 0.99 ±0.34 $\mu g\ g^{-1}$ Cr, 2.90 ± 1.14 $\mu g\ g^{-1}$ Pb, 3.63 ± 0.94 $\mu g\ g^{-1}$ Ni and 0.531 ± 0.104 $\mu g\ g^{-1}$ V, all recorded in front of Messaied industrial area. Comparing these levels with other areas, it is observed that values reported around Al-Saflyah were the least i.e. 0.3 ± 0.09 $\mu g\ g^{-1}$ Cr, 0.88 ± 0.13 $\mu g\ g^{-1}$ Pb, 1.14 ± 0.23 $\mu g\ g^{-1}$ Ni and 0.071 ± 0.009 $\mu g\ g^{-1}$ V. *P. semisulcatus* collected from Ras Laffan area showed levels in between those recorded for Al-Saflyah and Messaied (Table 2). Values observed in the present study were higher than those recorded for *P. semisulcatus* purchased from the retail market [5] during December 1989 - April 1990 (mean 0.237 ± 0.093 $\mu g\ g^{-1}$ Cr and 0.025 ± 0.009 $\mu g\ g^{-1}$ Pb) most of which could have been caught off the Iranian coast due to the cessation of commercial fishing of prawn in Qatari waters during this the period. or deteriorated during transfer to the market. On the other hand, in composite samples collected off Doha before, during and after the Gulf war oil spill crises , lead and nickel levels in prown ranged between 0.96 - 1.1 $\mu g\ g^{-1}$ for Pb and 0.62 - 0.97 $\mu g\ g^{-1}$ for Ni [6], with surprisingly higher values recorded in the pre-war period. Around Halul Island, where oil loading and storage activities are taking place Pb and Ni in *P. semisulcatus* reached 1.01 and 1.12 $\mu g\ g^{-1}$, respectively, during the same study [6]. Off Kuwait, Cr and Pb values in prawn before the war did not exceed 0.2 and 0.7 $\mu g\ g^{-1}$, respectively [7]. Off Bahrain, levels of Pb in prawn ranged between 0.008 and 0.15 $\mu g\ g^{-1}$ showing increased levels over other fish species [8] for October 1993 samples. This finding as well as that observed during the present study indicate that crustaceans in general appear to bio-concentrate metals to a much greater extent than fish, an observation which is consistent with published literature [9, 10, 11].

Table 2. Mean and standard deviation (±) for concentrations of trace metals in Qatari marine fish and prawn during 1995 - 1996

LOCATION / SPECIES	Weight range (g)	Length range (cm)	Trace metals ($\mu g\ g^{-1}$ dry wt) Chromium	Lead	Nickel	Vanadium
Al-Saflyah						
Lethrinus nebulosus	154 - 496	18 - 38	0.16 ± 0.02	0.24 ± 0.13	0.79 ± 0.24	0.021 ± 0.004
Sphraena jello	339 - 699	52 - 70	0.17 ± 0.03	0.16 ± 0.07	0.64 ± 0.19	0.032 ± 0.003
Rhaboosargus sarba	110 - 160	19 - 22	0.09 ± 0.02	0.08 ± 0.02	0.47 ± 0.17	0.029 ± 0.006
Siganus canaliculatus	80 - 130	10 - 17	0.11 ± 0.04	0.17 ± 0.11	0.51 ± 0.09	0.024 ± 0.004
Crenidens crenidens	120 - 145	17 - 26	0.16 ± 0.08	0.09 ± 0.03	0.53 ± 0.11	0.031 ± 0.005
Alepes mate	244 - 408	27 - 46	0.08 ± 0.03	0.04 ± 0.01	0.39 ± 0.16	ND
Gnathanodon speciosus	212 - 291	24 - 27	0.11 ± 0.03	0.13 ± 0.08	0.44 ± 0.12	0.042 ± 0.007
Epinephelus tauvina	314 - 630	29 - 57	0.23 ± 0.09	0.21 ± 0.14	0.63 ± 0.14	0.014 ± 0.004
Platycephalus indicus	504 - 549	44 - 51	0.19 ± 0.03	0.06 ± 0.02	0.31 ± 0.17	0.011 ± 0.003
Alepes melanoptera	131 - 206	13 - 18	0.06 ± 0.01	0.03 ± 0.01	0.59 ± 0.19	ND
Nematalosa nasus	16 - 36	80 - 130	0.19 ± 0.07	0.09 ± 0.02	0.43 ± 0.17	0.023 ± 0.006
Penaeus semisulcatus	10 - 31	11 - 18	0.30 ± 0.09	0.88 ± 0.13	1.14 ± 0.23	0.071 ± 0.009
Ras Laffan						
Lethrinus nebulosus	168 - 456	19 - 36	0.24 ± 0.13	0.35 ± 0.14	0.88 ± 0.24	0.053 ± 0.011
Epinephelus tauvina	414 - 981	53 - 72	0.31 ± 0.16	0.61 ± 0.21	0.97 ± 0.19	0.064 ± 0.006
Gnathanodon speciosus	192 - 238	25 - 27	0.14 ± 0.05	0.14 ± 0.07	0.64 ± 0.26	0.031 ± 0.006
Gerres oyena	140 - 165	21 - 25	0.23 ± 0.11	0.27 ± 0.04	0.90 ± 0.14	0.047 ± 0.009
Sphyraena jello	769 - 1598	58 - 74	0.18 ± 0.04	0.19 ± 0.11	0.83 ± 0.19	0.039 ± 0.014
Scomberoides commersonianus	526 - 904	41 - 51	0.26 ± 0.13	0.21 ± 0.09	0.66 ± 0.09	0.041 ± 0.009
Nematalosa nasus	92 - 156	19 - 39	0.26 ± 0.06	0.32 ± 0.13	0.73 ± 0.11	0.036 ± 0.011
Penaeus semisulcatus	9.5 - 31	11.8 - 17.3	0.59 ± 0.10	1.16 ± 0.40	1.93 ± 0.35	0.114 ± 0.017

Table 2. (Cont'd)

LOCATION SPECIES	Weight range (g)	Length range (cm)	Trace metals (µg g^{-1} dry wt)			
			Chromium	Lead	Nickel	Vanadium
Messaied						
Scomberoides commersonianus	490 - 887	39 - 49	0.36 ± 0.09	0.28 ± 0.11	1.96 ± 0.39	0.087 ± 0.022
Platycephalus indicus	491 - 527	42 - 47	0.29 ± 0.14	0.26 ± 0.13	1.58 ± 0.28	0.069 ± 0.017
Gerres oyena	163 - 175	24 - 27	0.13 ± 0.11	0.31 ± 0.14	1.47 ± 0.13	0.091 ± 0.014
Rhaboosargus sarba	86 - 105	17 - 21	0.16 ± 0.03	0.27 ± 0.09	1.26 ± 1.09	0.073 ± 0.019
Gnathanodon speciosus	215 - 311	26 - 30	0.17 ± 0.04	0.19 ± 0.07	1.01 ± 0.21	0.077 ± 0.023
Crenidens crenidens	117 - 151	19 - 27	0.19 ± 0.07	0.33 ± 0.13	0.97 ± 0.26	0.091 ± 0.013
Siganus canaliculatus	93 - 150	18 - 23	0.17 ± 0.06	0.21 ± 0.08	1.32 ± 0.17	0.083 ± 0.011
Lethrinus nebulosus	147 - 407	18 - 33	0.50 ± 0.10	0.43 ± 0.11	1.46 ± 0.33	0.135 ± 0.026
Epinephelus tauvina	593 - 1008	66 - 82	0.64 ± 0.11	0.93 ± 0.24	2.23 ± 0.18	0.232 ± 0.043
Penaeus semisulcatus	8 - 33	12 - 9	0.99 ± 0.34	2.90 ± 1.14	3.63 ± 0.94	0.531 ± 0.104

ND = Not detected

The highest average concentration of metals were observed in fish caught from Messaied area i.e. 0.31 ± 0.09 μg g^{-1} Cr, 0.36 ± 0.011 μg g^{-1} Pb, 1.47 ± 0.19 μg g^{-1} Ni and 0.109 ± 0.021 μg g^{-1} V. Statistical analyses indicated that all these means are significantly higher than fish caught from Ras Laffan and Al-Saflyah regions but with variable magnitudes i.e. $p < 0.001$ for Ni and V, $p < 0.01$ for Pb and $p < 0.05$ for Cr. The general trend of decreasing metals levels in fish is Messaied > Ras Laffan > Al-Saflyah (Table 2) matching the extent to which each of these regions is exposed to land-based or sea activities. On the other hand, apart from the increased metals concentrations in benthic fish species, statistical analysis did not reveal significant differences in between mean levels of fish species collected from the same area. Habashi et al. [12] found no significant differences in lead levels between species in the Western Gulf region.

The lowest individual metals concentrations recorded among different species were observed for *A. mate and A. melanoptera* (observed only in Al-Saflyah area) showing undetectable levels of vanadium. Concentrations for other metals for these species could be regarded as baseline levels for the area. *R. sarba* (common in Al-Saflyah and Messaied areas), *G. speciosus* (common in all areas) and *S. canaliculatus* (common in Al-Saflyah and Messaied areas) are among the fish showing lowest metals concentrations in coastal waters.

Generally speaking benthic fish showed increased affinity for metals accumulation than pelagic fish. This is evident from the increased metals levels recorded in *E. tauvina* specially for Messaied region where significantly higher values were recorded for Pb ($P < 0.001$), Ni ($p < 0.001$), Cr ($p < 0.05$) and V ($p < 0.01$). Increased concentrations off Messaied are related to increased industrial as well as other man-made activities in the region specially those related to oil loading, storage, exploration, exploitation and refining in addition to harbour activities and tankers traffic, all leading to elevated metals in water and sediments. The average total Pb in sediments from the area was 10.84 μg g^{-1} [13]. The same authors also mentioned higher levels in zooplankton (av. 3.66 μg g^{-1}) and phytoplankton (av. 2.07 μg g^{-1}).

Chromium is an anthropogenic pollutant reaching the gulf as a result of human activities associated with oil. Levels of Cr in marine fish were generally found to be less than 1 μg g^{-1}. Higher concentrations have been reported in plaice and herring i.e. 3.8 - 6.4 μg g^{-1} from the Irish Sea [14]. This metal was most concentrated in kidney followed by liver and muscle in fish from the coastal waters of Pakistan [15]. On the other hand, Pb can be accumulated in fish directly from seawater and sediments. The increase of salinity in the area i.e. average 45 psu could increase lead uptake by fish due to the tendency for formation of soluble Pb-chlorocomplexes. Nickel is an oil related metal, showing a nutrient type behaviour and can become toxic at high levels. Ni concentrations in marine fish are always less than 1 μg g^{-1} although in Liverpool bay and the Irish Sea, values > 2 μg g^{-1} have been observed [16]. V is also an oil related metal bearing a constant ratio to Ni. Little is known about V levels in marine fish in the area.

Our results demonstrate significant associations between metals in each area and the whole region specially Ni and V on the one hand ($r = 0.844$, $p < 0.0.003$), and Pb and Cr on the other ($r = 0.762$, $p < 0.004$), an observation suggesting that these metals enter the marine environment around Qatar from the same source.

Table 3 shows the concentrations of Cr, Pb, Ni and V in fish flesh observed from different locations in the Gulf (ROPME Sea Area) including those previously measured around Qatar compared to values measured during the present study. Values observed off Al-Saflyah and even those off Ras Laffan fall within levels recorded in the coastal waters of the different Gulf States; however, levels for fish caught off Messaied were always significantly higher. Sometimes these levels, example those for Pb, approach the upper limit recommended by WHO [9] for seafood while Ni concentrations in *E. tauvina* (Table 3) exceeded the guidelines and maximum permissible levels given by WHO [9]. When compared to previous data collected from other locations around Qatar [6], data for Pb are in the same general range, except those collected from Messaied area; whereas some values for Ni exceeded the maximum concentrations recorded. Levels for Cr were within the range of 1989/90 samples [5] while V is recorded for the first time in Qatari fish (Table 3).

Table 3 . Mean levels of trace metals (μg g^{-1} dry weight) in fish species from the present study and values obtained from other studies in the Gulf.

METAL	ROPME SEA AREA 1993 [17]	ROPME SEA AREA 1992 [12]	QATAR OFF DOHA 1991 [6]	QATAR AROUND HALUL ISLAND 1991 [6]	** QATAR (MARKET) 1989 / 90 [5]	*KUWAIT 6/91 [18]	*BAHRAIN 6/91 [18]	BAHRAIN 1993 [8]	*SAUDI ARABIA 8/91 - 92 [18]	*UAE 9 / 91 [18]	*OMAN 10 / 91 [18]	***WHO 1991 [19]	QATAR 1995 / 1996 PRESENT STUDY
Cr	0.2	-	-	-	0.33 - 0.6	0.14	ND - 0.53	-	0.05 - 0.21	0.16 - 0.4	0.03 - 0.38	2.0	0.06 - 0.64
Pb	0.29	0.01	0.58 - 11.18	0.52 - 3.82	0.09 - 0.11	0.27	ND - 0.13	0.006 - 1.04	0.008 - 0.04	0.01 - 0.08	ND - 0.07	1.3	0.03 - 0.93
Ni	0.31	0.36	0.6 - 2.24	0.58 - 2.08	-	-	ND - 0.49	-	0.023 - 0.38	0.06 - 0.18	0.02 - 0.32	2.0	0.31 - 2.23
V	0.02	0.94	-	-	-	0.07	0.01 - 0.1	-	0.02 - 0.05	0.01 - 0.02	ND - 0.03	1.0	ND - 0.232

ND = not detected

* Main fish species = E. tauvina, E. jayakan, E. suillus, L. nebulosus, A. thalassinus.

** E. tauvina and L. nebulosus.

*** Maximum permissible levels.

Based on the overall mean concentrations of metals in different fish species which are 0.226, 0.260, 0.93 and 0.058 $\mu g\ g^{-1}$ for Cr, Pb, Ni and V, respectively; and the average consumption of fish per person 20 kg in 1994 (54 gram/day), the daily intake of metals by Qatar marine fish consumers can be calculated as 12, 14, 50 and 3 µg/day for Cr, Pb, Ni and V respectively.The provisional tolerable weekly intake (PTWI) per 60 kg average person was calculated. Therefore, PWI values were 1.4, 1.6, 5.8, and 0.3 $\mu g\ kg^{-1}$ bw/wk for Cr, Pb, Ni and V respectively. The established PTWI for these metals (9, 10, 19, 20) are 20, 40, 75 and 5 $\mu g\ kg^{-1}$ bw/wk, respectively which indicate higher limits than those observed in the precent study. Thus, the current rate of fish consumption in Qatar is considered safe to seafood consumers. Vanadium could present a potential risk to persons eating prawn more than 8 meals per week as they can easily reach the PTWI. Therefore, a continuous monitoring programme for metal levels in fish species became a must.

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