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Estimation of heavy metals in some economic fishes in the Arabian Gulf, Saudi Arabia

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Abstract
The aim of the present study was to investigate the concentration and the pollution status of heavy metals; Cadmium, Copper, Lead, Zinc, Nickel, Iron, Chromium, Arsenic and Mercury in the water, sediment and some economic fishes in the Arabian Gulf, Saudi Arabia. The Cd, Cu, Pb, Zn, Ni, Fe, Cr, As and Hg were determined monthly in water, sediment and in the muscles, gills and livers of six fish species; Orange-spotted grouper (*Epinephelus coioides*), Spangled emperor (*Lethrinus nebulosus*), White-spotted rabbitfish (*Siganus canaliculatus*), Silver sillago (*Sillago sihama*), Twobar seabream (*Acanthopagrus bifasciatus*) and Haffara seabream (*Rhabdosargus haffara*) collected from the coastal water of Arabian Gulf, during the period (July–December 2015). The results declared that, the average concentrations of heavy metals were in trend water < soil< livers of fishes, the liver recoded the highest heavy metals values in all fish species, but the muscles (edible parts) of all species recorded the lowest values of heavy metals and less the permissible limit of national and international standers. The accumulation of heavy metals in the edible muscles of the investigated species and other organs may be considered as an important warning signal for water quality, fish health and human consumption.

Keywords: Arabian Gulf, water quality, sediment, fish, heavy metals

1. Introduction

Arabian Gulf is set as an extremely arid region of a shallow semi-closed water basin, it receives a huge amount of waste water and other pollutants from global oil transportation, drilling oil, industrial and human activities, it leads to disturbance to the coastal environment [1, 2]. The water and sediment quality is considered the main factor controlling the state of the aquatic environmental health. Pollution of the organic and inorganic chemicals play an important role in threatens to marine organisms, including fish [3]. The concentration of metals in the animal organs can be a reasonable guide to public health standards and for the organism condition as well. Each environmental pollutants, and metals are of particular concern, to their potential toxic effect and ability to bio accumulate in aquatic ecosystems [4]. Heavy metal concentrations monitor in aquatic ecosystems by measuring their concentrations in water, sediments and biota [5, 2], which exist in low levels in water and attain considerable concentration in sediments and biota [6]. Sediments are important substrates for various pollutants like pesticides and heavy metals and play a significant role in the demobilization of contaminants in aquatic systems under favorable conditions and in interactions between water and sediment. Fish samples can be considered as one of the most significant indicators for the estimation of metal pollution level [7]. The bioaccumulation of heavy metals varied between species, ages, sex and organs. In general, the target tissues of heavy metals are metabolic active ones which accumulate high levels of metal in fish such as liver and gills, whereas in muscles where the metabolic activity is relatively low accumulates less level of heavy metals. The measurement of heavy metal concentrations in bio-indicators have been recognized as highly relevant in Eco toxicological terms because of the reflection of bioavailability in the ecosystem that reasonable measurement for public health standards from the animal health point of view [8, 9].
Therefore, the objective of the present study is to determine levels of certain heavy metals in water, sediment and fishes from the Arabian Gulf.

2. Materials and Methods

Collection and preparation of samples

Water temperature, salinity, dissolved oxygen (DO) and pH were measured in situ at each site using a pH/ISE/ conductivity/RDO/DO Meter Thermo Scientific Orion Star A329 Portable. Duplicate water samples for water quality variables were collected (surface samples) 0.25-m depth (below surface water) and above sediment 0.25-m (bottom samples), using a PVC Niskin bottle. NH_3-N concentrations were determined according to 10-IOC (1983). NO_2-N, NO_3-N, PO_4-P Total-P and SiO_4-Si concentrations were determined in pre-filtered seawater samples, (Whatman GF=C) following the techniques described by 11- IOC (1993) and 12-Strickland and Parsons (1972). For chlorophyll-a (Chl-a) determination, additional water samples were collected and filtered on 0.45 mm filters. Chl-a was extracted by using 90% acetone and measured spectrophotometrically according to 12- Strickland and Parsons (1972).

Two water samples were taken monthly from three sites in 500 ml polyethylene container, and acidified by addition of 2 ml of HNO_3 in 1 liter of water, to prevent the absorption of heavy metals on the vessel walls. The samples were then transported to the laboratory at (4 °C) and heavy metals were analyzed on the same day.

Three samples of sediment were taken monthly from the surface of sediment during the period (Jul – December 2015) deep (0-10 cm) of each site and dried in air, then dried in an oven at (60 °C for 72 h) grinding and sieve through a mesh size 120 (125µ) screen, take part for chemical analyses and other part (0.5 g) digests with 8 ml HNO_3 (65%) in a microwave mineralization (microwave Milestone Ethos one) (ISO 16729:2013, Soil quality - Digestion of soluble elements in nitric acid), the heavy metals were measured.

Six fish species; Orange-spotted grouper (*Epinephelus coioides*), Spangled emperor (*Lethrinus nebulosus*), White-spotted rabbit fish (*Siganus canaliculatus*), Silver sillago (*Sillago sihama*), Two bar seabream (*Acanthopagrus bifasciatus*) and Haffara seabream (*Rhabdosargus haffara*) were captured monthly from three sites in the Arabian Gulf by using cast nets. Immediately, after sampling fishes were stored in a container, preserved in crushed ice and transferred to the laboratory. We used 5 same size from every species of fishes and frozen at -20 °C until analyzed. Before analysis, muscle, liver, and gills were removed. They were dried in an oven (60 °C for 72 h) until fixed weight and calculated the moisture %, grind and sieve through a 2 mm screen for subsequent, the 500-600 mg of dried samples was accurately weighed into polyethylene tube, then digests with 8 ml HNO_3 (65%) in a microwave mineralization (microwave Milestone Ethos one). The heavy metals in all samples (water, sediment and fish) were determined by the Inductively Coupled Plasma-Optical Emission Spectrometer (ICP-OES; Varian 720-ES) and mercury was measured by the Direct mercury analyzer (DMA-80), Milestone Company, Model Unit DMA-80ICP (13- Clesceri *et al*., 1998) and they were quantified in ppm.

The bioconcentration factor (BCF) was calculated for each metals, as the relation between the metal concentrations in whole fish (FC) and its concentration in the water (WC) according to the equation: BCF = FC/WC [14].

Statistical analysis was run using the Statistical Package for the Social Science (SPSS 22). One and two ways ANOVA were employed to find the significant differences of physicochemical parameters and heavy metals between sites and months, also, means ± standard errors and Duncan were derived from all data.

3. Results

The average mean values for water quality of the study area are illustrated in Table 1; the Chlorophyll "a" (µg/l) (25.84±6.36), NH_3mg/l (0.020±0.004), NO_2mg/l (0.004±0.001), NO_3 mg/l(0.49 ± 0.05), Sulfide (µg/l) (5.08 ± 1.56), Active phosphorus mg/l (0.42±0.17), Total phosphorus mg/l (0.58±0.16), Total chloride mg/l (0.02±0.00),Total Alkalinity mg/l (156.00±5.53), CO_2 mg/l (1.94±0.19), DO. mg/l (6.53±0.32), DO % (79.70±4.75), pH (8.10±0.02), Electric conductivity mg/l (64.69±0.41), Salinity %º (43.64±0.29), Total dissolves salts (31.72±0.19) and heavy metals were Hg (ppm) (0.00828±0.00527), As (ppm) (0.00277±0.00070), Zn (ppm) (0.01940±0.00722), Ni (ppm) (0.00085±0.00025), Fe (ppm) (0.00010±0.00004), Cr (ppm) (0.00528±0.00184).

In another hand the chemical characters of sediment recorded these values in Table 1; pH (8.180±0.063), NH_3 mg/l(0.403±0.163), NO_2 mg/l (0.087 ± 0.060), NO_3 mg/l (2.890 ± 0.680), Sulfide (µg/l) (27.000±5.947), Active phosphorus mg/l (1.330±0.093), Organic Carbon (3.4733±0.5133) and heavy metals were Hg (ppm) (0.0453±0.0297), As (ppm) (3.2757±2.8585), Zn (ppm) (3.4021±0.6193), Ni (ppm) (1.6122±0.5051), Fe (ppm) (1.1125±0.4031), Cr (ppm) (6.0351±1.3139).

In the present study, gills and liver are chosen as target organs for assessing metal accumulation higher than the muscle tissue, which usually exhibits a low accumulation potential of metals. Therefore, muscle is chosen because it is the part consumed. The liver recoded the highest heavy metals values in all fish species, but the muscle (edible parts) of all species recorded the lowest values of heavy metals.

Heavy metal concentrations in the muscles, gills and liver of fish species collected from coastal Arabian Gulf waters was illustrated in Table 2 and Fig 1. The Hg, Ni, Fe, Zn As and Cr (ppm) recorded values in gills, livers and tissues in all species of fishes. The results indicate that Cu metals (ppm) were recorded in liver of most species where it was (1.0304±0.0524) in Spangled emperor liver and was (0.2789±0.120) in White spotted rabbit fish liver. The Cu (ppm) was (2.0190±0.0782), (1.1322±01566) and (1.3284±0.0208) in liver, gills and tissue of sea bream, (4.0764±1.0786, 2.0894 ± 0.0537, 1.3401 ± 0.0718) in liver, gills and tissue of Orange-spotted grouper,(1.4467 ± 0.3157, 2.3329 ± 0.1926,1.3990 ± 0.2684) in liver, gills and tissue of Silver sillago, and was (0.2571 ± 0.0437, 1.9870 ± 0.0864, 1.0873 ± 0.1859) in the liver, gills and tissue of Two bar seabream.

The Cd (ppm) metals was not detectable in the gills and tissue in all species, but recorded in the liver of some species, where was (0.1280± 0.0150) in Spangled emperor liver, (0.2064± 0.0386) in the liver of Haffara sea bream, (0.1245 ± 0.0110) in liver of Orange-spotted grouper and (0.3570 ± 0.0577) in the liver of Silver sillago.

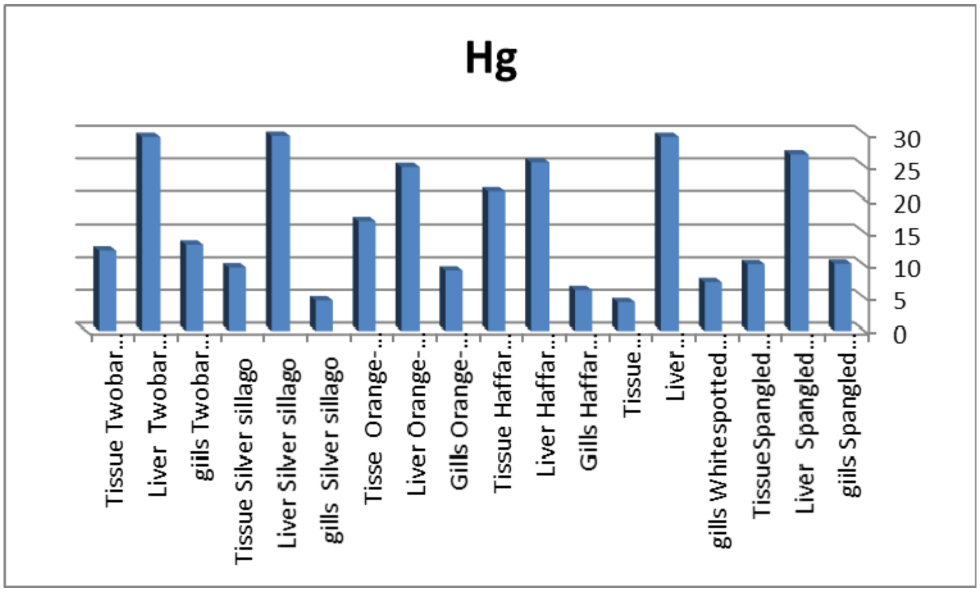
The Pb (ppm) metals were not detectable values in livers, gills and tissues of all fish species were studied except the gills of Orange-spotted grouper.

Table 1: Average means ± standard Error (SE) of Physico-chemical parameters and heavy metals for three water and sediment sites inside the Arabian Gulf

Parameters	Average water sites		Average sediment sites	
	Means	**±SE**	**Means**	**±SE**
Chlorophyll a (µg/l)	25.84	6.36		
NH_3 mg/l	0.020	0.004	0.403	0.163
NO_2 mg/l	0.004	0.001	0.087	0.060
NO_3 mg/l	0.49	0.05	2.890	0.680
Sulfide (µg/l)	5.08	1.56	27.000	5.947
Active phosphorus mg/l	0.42	0.17	1.330	0.093
Total phosphorus l mg/l	0.58	0.16		
Total chloride mg/l	0.02	0.00		
Total Alkalinity mg/l	156.00	5.53		
CO_2 mg/l	1.94	0.19		
DO. mg/l	6.53	0.32		
DO %	79.70	4.75		
PH	8.10	0.02	8.180	0.063
Electric conductivity mg/l	64.69	0.41		
Salinity ‰	43.64	0.29		
Total dissolves salts	31.72	0.19		
Organic Carbon			3.4733	0.5133
Hg (ppm)	0.00828	0.00527	0.0453	0.0297
As (ppm)	0.00277	0.00070	3.2757	2.8585
Zn (ppm)	0.01940	0.00722	3.4021	0.6193
Ni (ppm)	0.00085	0.00025	1.6122	0.5051
Fe (ppm)	0.00010	0.00004	1.1125	0.4031
Cr (ppm)	0.00528	0.00184	6.0351	1.3139

Table 2: Average means ± standard Error (SE) of heavy metals in the gills, livers and tissues for six species of fishes monthly collected from three sites inside the Arabian Gulf

Parts	Hg	Zn	Pb	Ni	Fe	Cu	Cr	Cd	As
Giils Spangled emperor	0.2392 ±0.0979	48.7012 ±2.6081	ND	0.3312 ±0.0505	0.1052 ±0.0201	ND	1.2260 ±0.1499	ND	5.0611 ±0.6751
Livers Spangled emperor	0.6284 ±0.0107	81.5388 ±9.0749	ND	1.8630 ±0.3532	0.9643 ±0.1906	1.0304 ±0.0524	2.3645 ±0.3260	0.1280 ±0.0150	14.0943 ±3.7801
Tissues Spangled emperor	0.2376 ±0.0728	27.1407 ±5.0878	ND	0.1396 ±0.0657	0.1127 ±0.0642	ND	1.0330 ±0.1112	ND	10.3058 ±1.6014
Gills Whitespotted rabbitfish	0.1727 ±0.0766	88.7895 ±20.0072	ND	0.4252 ±0.1369	0.0535 ±0.0079	ND	0.8434 ±0.3206	ND	9.7489 ±3.0481
Livers Whitespotted rabbitfish	0.6914 ±0.0686	94.6570 ±13.9740	ND	1.9456 ±0.0210	0.9875 ±0.0053	0.2789 ±0.0075	1.9870 ±0.058	ND	8.0157 ±0.5791
Tissues White spotted rabbit fish	0.1010 ±0.0212	19.1328 ±2.2187	ND	ND	0.0584 ±0.0071	ND	0.6573 ±0.1255	ND	26.5114 ±7.9119
Gills Haffar seabream	0.1440 ±0.0386	55.6100 ±1.9940	ND	0.3534 ±0.0728	0.1881 ±0.0786	ND	1.1322 ±0.1566	ND	4.5396 ±1.5167
Livers Haffar seabream	0.5996 ±0.0136	80.9525 ±6.3843	ND	3.0781 ±0.2024	1.1086 ±0.1023	1.0105 ±0.0381	2.0190 ±0.0782	0.2064 ±0.0386	10.7287 ±2.3232
Tissues Haffar seabream	0.4957 ±0.1324	50.6205 ±1.4037	ND	0.3578 ±0.0112	0.0347 ±0.0261	0.0070	1.3284 ±0.0208	ND	15.1964 ±7.5035
Gills orange-spotted grouper	0.1954 ±0.0274	51.8271 ±5.7725	0.0036 ±0.0011	0.7745 ±0.2577	0.2617 ±0.0568	0.0147 ±0.0048	4.0764 ±1.0786	ND	9.2153 ±2.4192
Livers orange-spotted grouper	0.5830 ±0.0150	81.1316 ±1.0062	ND	1.7348 ±0.2788	0.9086 ±0.3397	0.9423 ±0.0453	2.0894 ±0.0537	0.1245	12.2510 ±1.8726
Tissues orange-spotted grouper	0.3920 ±0.1466	46.9409 ±1.0155	ND	0.4221 ±0.0214	0.0042 ±0.0022	0.0138 ±0.0034	1.3401 ±0.0718	ND	15.1700 ±5.4191
Gills Silver sillago	0.1070 ±0.0220	79.0885 ±26.6517	ND	0.5290 ±0.2427	0.0764 ±0.0059	ND	1.4467 ±0.3157	ND	4.5982 ±0.2805
Liver Silver sillago	0.6952 ±0.0338	108.3845 ±5.5819	ND	2.6632 ±0.1915	1.8000 ±0.1768	0.8578 ±0.0345	2.3329 ±0.1926	0.3570 ±0.0577	20.8760 ±5.1631
Tissues Silver sillago	0.2260 ±0.0590	42.8942 ±6.3642	ND	0.2848 ±0.0989	0.0340 ±0.0104	0.0045 ±0.0030	1.3990 ±0.2684	ND	16.7580 ±4.8192
Giils Twobar Seabream	0.3080 ±0.0820	51.1946 ±0.0018	ND	0.1463 ±0.0331	0.1299 ±0.1076	0.0018 ±0.0004	0.2571 ±0.0437	ND	2.9800 ±0.6088
Livers Twobar Seabream	0.6914 ±0.0421	94.6570 ±9.3604	ND	1.9456 ±0.0018	0.9875 ±0.0032	0.2789 ±0.0310	1.9870 ±0.0864	ND	8.0157 ±0.8354
Tissues Twobar Seabream	0.2863 ±0.0184	39.0261 ±7.0124	ND	0.1538 ±0.0200	0.0359 ±0.0114	0.0088 ±0.0014	1.0873 ±0.1004	ND	17.6397 ±2.0106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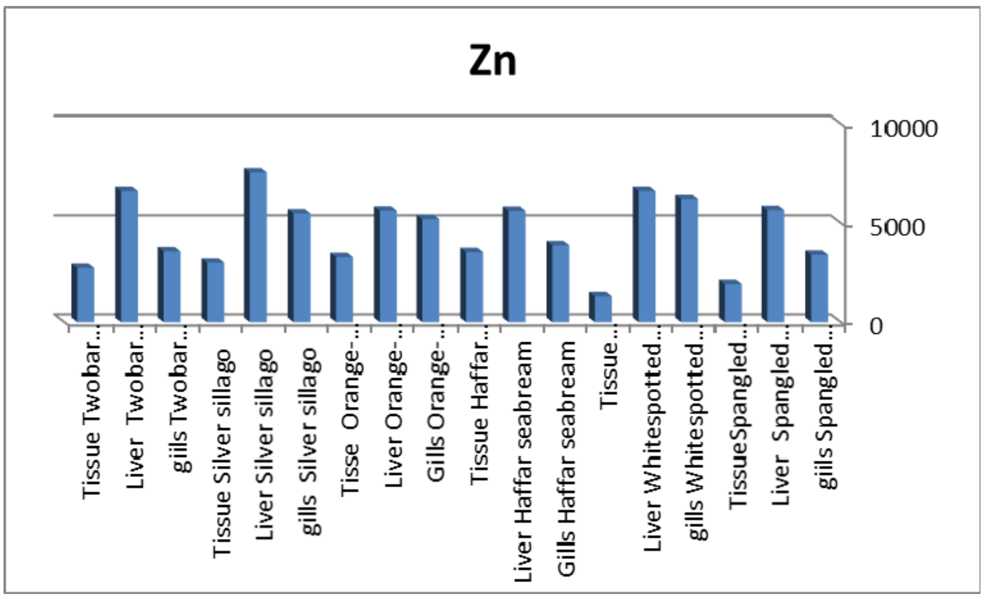
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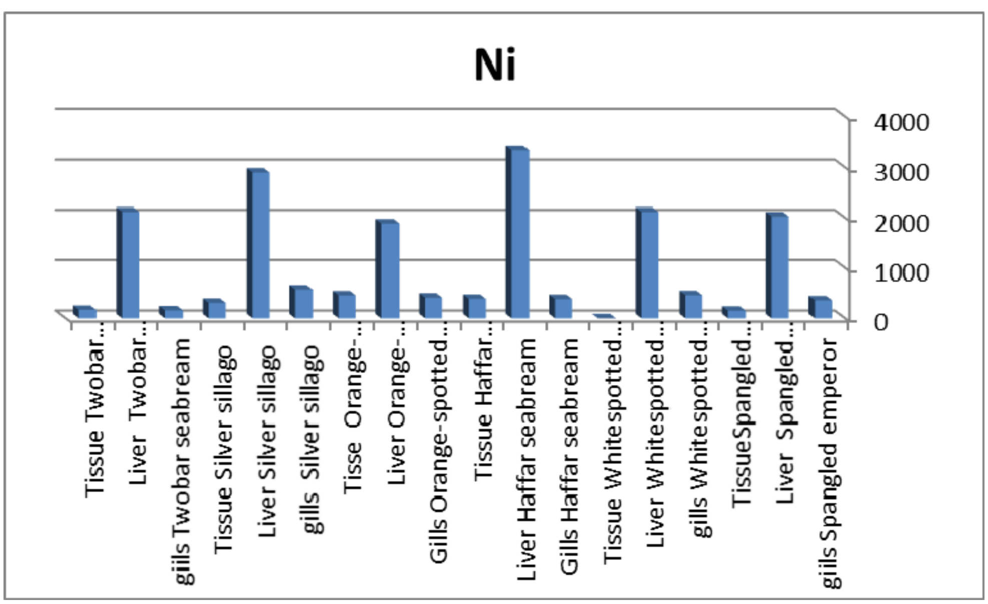
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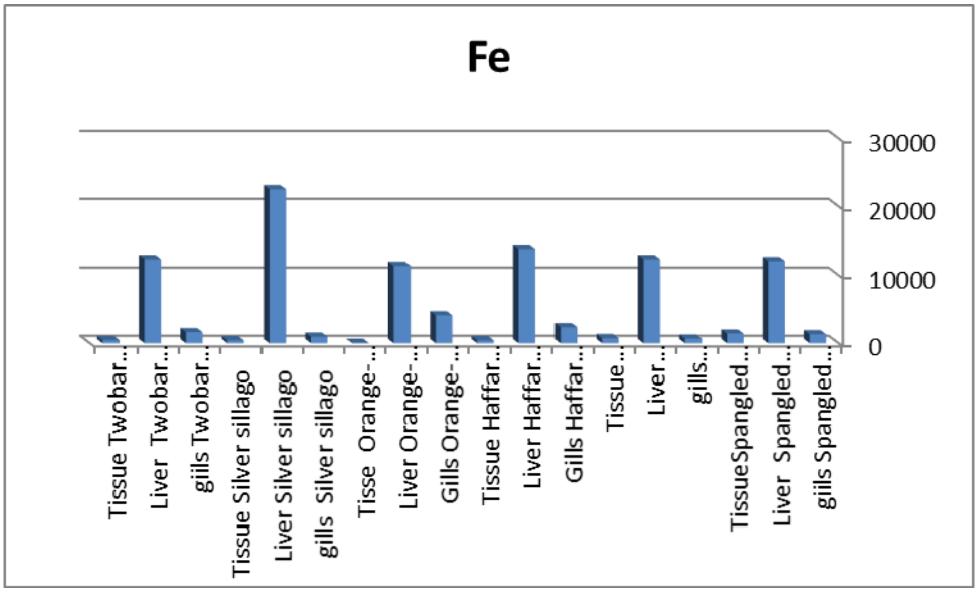

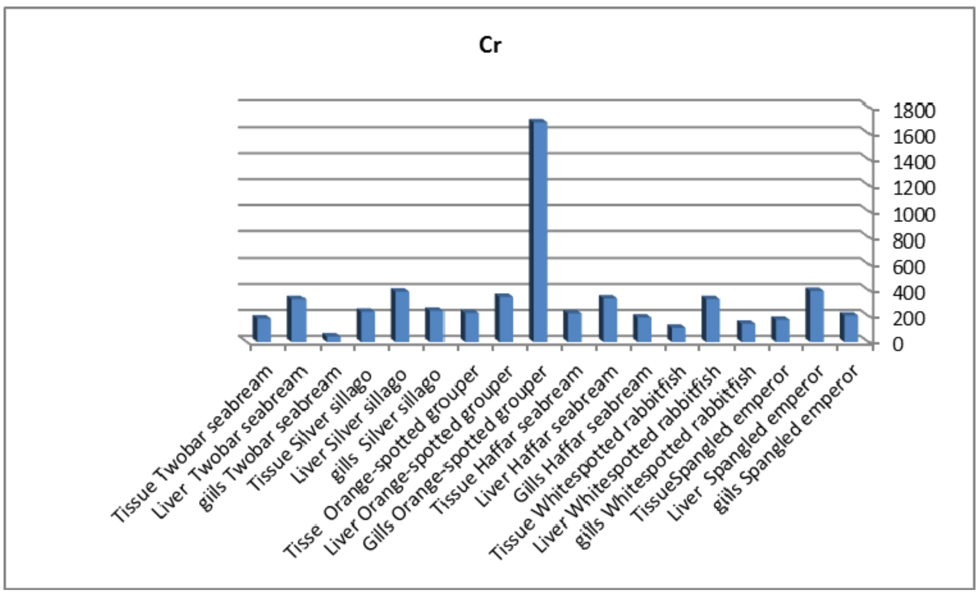

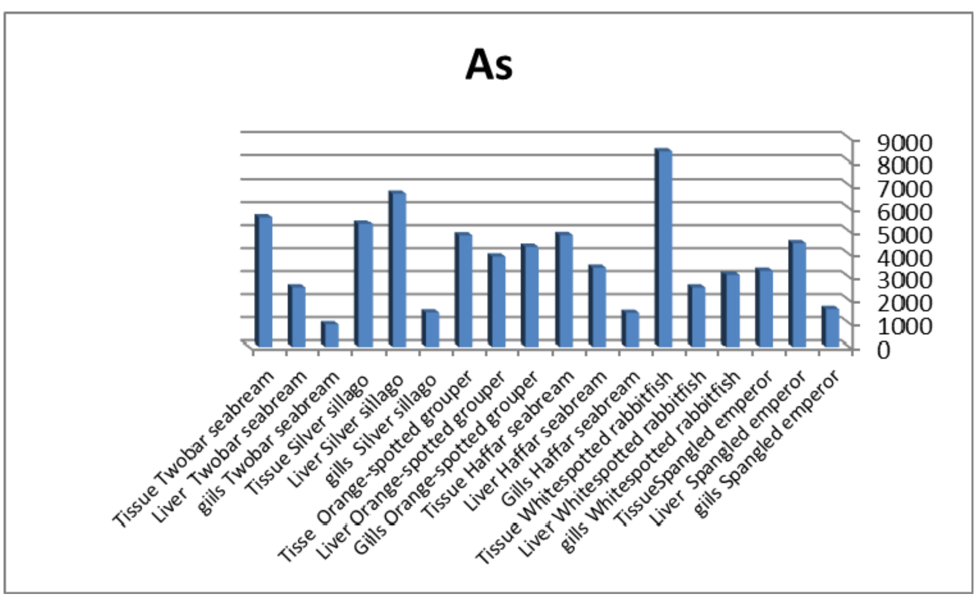

Fig 1: The heavy metal concentrations (Hg, Zn, N i, Fe, Cr and as) in gills, Livers and muscles of six fish species

4. Discussion

Chlorophyll-a is the most widely used parameter among the marine pigments for this purpose as the abundance of phytoplankton is highly associated with nutrient availability, monitoring of chlorophyll-a (chl-a), together with the dissolved nutrients, provides a basis for detecting the possible occurrence of eutrophication, chl-a concentration in a water body is used as an indicator of water quality.

In the present study, pH exhibited within acceptable ranges falls in the desired range of 6.5–9.5. The growth of fish will be good in the range of 7-8 [16]. This study indicates that the study area have a good DO regime (>5 mg/l) The general distribution of dissolved oxygen (DO) indicated high values and the presence of well oxygenated waters, it is a tolerable range for most fish as a guideline value prescribed by [15-18].

Nitrogen naturally exists in marine waters both as inorganic and organic forms, and in dissolved and particulate forms. Inorganic nitrogen is found both as oxidized forms, e.g. nitrate (NO_3) and nitrite (NO_2), and as reduced forms, e.g. ammonium (NH_4), ammonia (NH_3), and nitrogen gas (N_2). In the marine environment, it is widely accepted that nitrogen is the primary nutrient limiting plant growth, but recent evidence has also demonstrated the dominant role of phosphorus on certain seasons [19, 20].

On the other hand, phosphate (PO_4) inputs into the coastal waters usually come from drainage discharges (point sources), although PO_4 loading from agricultural runoff and atmospheric deposition (diffuse sources) can be significant [21].

Similar results of pH and nutrient concentrations were recorded in the Arabian Gulf at Qatar coast [22]; in Red Sea at Rabigh and Jeddah, Saudi Arabia [2]. The increased of nutrient concentration in water may be attributed to the mixed drainage water (sewage, agricultural and industrial waste-water) at these sites, where similar results were obtained in the Arabian Gulf and Red Sea Coast, SA [2].

Generally, the water quality and chemical characters of sediment in these sites showed a less significant deviation from the International standards and Saudi standard values in most parameters values of marine water quality and are well below the accepted levels and no harmful level in micro flora and the fauna of the Arabian Gulf in the study area.

The heavy metal levels in water sample were as follows Zn > Hg > Cr > As > Ni >Fe. The results of metal concentrations in water were used to calculate BCFs. The bioaccumulation factors (BCFs) of the tissues of the investigated fish species were presented in Table 3. BCF is a number that describes the bioaccumulation as the ratio between the accumulated concentration of a given pollutant in any species to the concentration in the surrounding environment according to [14].

Heavy metal concentration in water and sediment recorded in this result may be related to the nutrients, sewage and agricultural discharges at this area, which might be due to anthropogenic inputs from the coastal area. It is found that there is a relationship between heavy metals concentration in water and sediment, where the concentration of heavy metals in sediment were higher than that recorded in water agreement with [23] in the Arabian Gulf. Some heavy metals concentrations in sediment are duplicated (10-100 times) than that recorded in water, while the bioaccumulation in fishes are duplicated hundreds folds them in water and sediment Table 3. Comparison of the mean concentration of heavy metals in the sediments and sediment quality guidelines (SQG) of the USEPA [24] indicated that the sediment samples in the study area were in the range non-polluted to heavy metals Cd, Cr, Cu, Ni, Pb and Zn. These values are considered a part of a risk for fishes consumers and human health, especially for Arsenic.

The heavy metals As, Fe, Cr, Ni, Hg and Zn concentrations in water in the three sampling sites were compared with international standards. The obtained results showed that, the heavy metal concentrations in water did not exceed than international standard and Saudi standard guidelines [24-29].

The metal concentrations obtained from the sediment samples were compared with Sediment Quality Guideline (SQG) which showed that these concentrations did not exceed the probable effect concentration (PEC) levels. Generally, the Cd, Cu and Pb concentrations in water and sediment were not detectable.

The results confirmed the differences of accumulation of heavy metals in different tissues of fishes. The highest concentrations of heavy metals were found in the livers and the lowest values in the muscles. This result agrees with [30]. Similar results were reported from a number of fish species which shows that muscle is not an active tissue in accumulating heavy metals [31, 32]. They reported that the metal concentrations were lowest in muscle, and did not exceed the established quality standards for fish.

Zn toxicity is rare, yet it can be toxic above the limit of 50 μg·g-1 wet weight in muscle. It appears to have a protective effect against the toxicities of both cadmium and lead [33, 34]. Hence, the relatively high levels of these metals compared to other metals studied can be attributed to them essentiality [35].

Cu concentration in all examined fish species was found in livers only and Concentrations of Cd recorded in the current study in the livers of some fish species, but Pb has not found except the gills of orange spotted grouper. A lot of researchers have reported that metals accumulate in high concentrations in the gills and the livers, because these organs have relatively higher potential for metal accumulation than muscle [36, 37].

The standard safety level of Cd (0.04 μg·g−1 wet weight) in fish tissue [26, 38]. This could be attributed to the industrial activities situated close to the coast and directly discharges its effluents into it as well as shipping discharge from fishing boats.

Furthermore, the adsorption of metals onto the gill surface as the first target for pollutants in water could also be a significant influence in the total metal levels of the gill. This is in agreement with the literature [39, 40]. Marine organisms, including fish, crustaceans and squids, may accumulate heavy metals through direct absorption or via their food chain and pass them to human beings, by consumption, causing chronic or acute disorder [41, 42].

The fish which have a close relationship with sediment, shows relatively high body concentration of metals as well as grouper, and can be related to several factors such as habitat, dietary uptake, pollutant bioavailability. The fact that many demersal fish species are long-lived and tend to feed at higher trophic levels than their pelagic fish counterparts may lead to a potentially higher level of accumulation of persistent pollutants, particularly those are not easily metabolized or degraded [43].

The extent of occurrence or accumulation of trace metals by organisms in different tissues is dependent on the route of entry. The accumulation process involves the biological sequestering of metals that enter the organism through respiration and epidermal. It has been indicated that BCFs from environment to fish tissue changes according to the species of the chemical, the metabolite properties of the tissues and the pollution degree of the environmental [44, 45].

Table 3: BCF of heavy metals in the gills, livers and tissues for six species of fish monthly collected from three sites inside the Arabian Gulf

Parts	Hg	Zn	Ni	Fe	Cr	As
Giils Spangled emperor	10	3382	360	1315	204	1607
Liver Spangled emperor	27	5662	2025	12054	394	4474
Tissue Spangled emperor	10	1885	152	1409	172	3272
Gills Whitespotted rabbitfish	7	6166	462	668	141	3095
Liver Whitespotted rabbitfish	29	6573	2115	12344	331	2545
Tissue Whitespotted rabbitfish	4	1329	0	729	110	8416
Gills Haffar seabream	6	3862	384	2351	189	1441
Liver Haffar seabream	26	5622	3346	13857	336	3406
Tissue Haffar seabream	21	3515	389	434	221	4824
Gills Orange-spotted grouper	9	5199	414	4106	1685	4315
Liver Orange-spotted grouper	25	5634	1886	11358	348	3889
Tisse Orange-spotted grouper	17	3260	459	52	223	4816
Gills Silver sillago	5	5492	575	955	241	1460
Liver Silver sillago	30	7527	2895	22500	389	6627
Tissue Silver sillago	10	2979	310	425	233	5320
Gills Twobar seabream	13	3555	159	1624	43	946
Liver Twobar seabream	29	6573	2115	12344	331	2545
Tissue Twobar seabream	12	2710	169	449	181	5600

5. Conclusion

Consequently, it can be concluded that the concentrations of heavy metals in the water, sediment and fishes from the Arabian Gulf are lower than the TSE-266, [24-28]. There were high levels of heavy metals in fish than water and sediment. It was determined that the pollution may be reached to hazardous levels for the health of human. Also, a potential danger may exist in the future, depending on the anthropogenic activity in this region. The accumulation of heavy metals in the edible tissues of the investigated species and other organs may be considered as an important warning signal for fish health and human consumption. The present study shows that precautionary measures need to be taken in order to prevent future heavy metal pollution. Therefore, further monitoring programs should be conducted.

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