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Accumulation of Heavy Metals (Fe, Mn, Cu, Zn, Ni, Pb, Cd and Cr) in Tissues of Narrow-barred Spanish mackerel (Family-Scombridae) Fish Marketed by Karachi Fish Harbor

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Abstract: In the present study, *Scomberomorus commerson* was collected during the period 2006-2011 in order to provide information on the concentrations of eight heavy metals present in this marine species commonly consumed by the population and to have knowledge whether these levels may constitute a hazard to consumers. Liver showed high concentrations of metals in the tissues and organs. Fe is the most accumulated in all tissues and organs. The highest mean concentration of Fe (608.93±113.22 µg/g), Mn (9.79±4.22 µg/g), Cu (38.57±16.62 µg/g), Zn (53.25±26.50 µg/g), Ni (3.22±1.13 µg/g), Pb (1.20±0.64 µg/g), Cd (2.03±0.91 µg/g) and Cr (1.93 + 0.87 µg/g) was determined in liver of fish. The order of abundance of the metals in the fish samples based on concentrations in the muscle tissues analysed were as follows: Fe>Zn>Cu>Mn>Ni>Cd>Pb≥Cr. For the toxic metal, Pb was found to have lower concentration of the mean values than the permissible limits set by FAO/WHO. However, Cd level was higher than the permissible limit and it can be concluded that the contamination of Cd in *S. commerson* may pose threat for the importers due to high per capita consumption of the fish.

Keywords: Accumulation, Heavy Metals, Karachi Fish Harbor, *Scomberomorus commerson*.

1. INTRODUCTION

The seas and oceans, which cover 70% of the world's surface, are one of the man's great hopes for future food supplies. As human populations multiply and industrialization increases, the problems of environmental pollution become more critical [1]. Beijer and Jernelov [2] reported that heavy metals are critical in this regard because of their easy uptake into the food chain and bioaccumulation processes they are enter the marine environment naturally through weathering of the earth's crust. In addition to geological weathering, human activities have also introduced large quantities of metals to local water bodies, thereby disturbing the natural balance in the ecosystem [3]. Industrial effluents, agricultural runoffs, transport, burning of fossil fuels, animal and human excretions and geologic weathering and domestic waste contribute to the heavy metals in the water bodies [4]. In harbors there is usually an increase in the pollution level including heavy metal, which will restrict the range of species including fish, but those organisms that are able to withstand the pollutants can abound.

Karachi is capital of Sindh as well as the largest and most populous metropolitan city of Pakistan and the main seaport and financial center of the country. The population of Karachi is 23.5 million people as of 2013 and area of approximately 3527 km^2, resulting in a density of more than 6,000 people per square kilometer. Karachi is the location of the Port of Karachi and Port Bin Qasim, two of the region's largest and busiest ports. The city is with maximal population density and is located on the Arabian Sea coastline. The main drivers along the coast impacting on the marine environment are: industry, shipping, tourism, port activity, urbanization and agriculture. The main types of industry contributing to the amount of industrial waste waters are chemical industry, energy power stations, automotive industry, textiles, food industry, shipbuilding and ship repair. The waste waters from industrial complexes mainly discharge into the sea. The tourism is another major driver responsible for human impact on the marine water quality.

Fish constitute an important source of protein for many people throughout the world and fish consumption has increased in importance among health-conscious people because it provides healthy and low cholesterol sources of protein and other nutrients [5, 6]. Heavy metals such as copper (Cu) and zinc (Zn) are essential for fish metabolism while some others such as lead (Pb) have no known role in biological systems. For the normal metabolism of fish, the essential metals must be taken up from water, food or sediment. However, similar to the route of essential metals, nonessential ones are also taken up by fish and accumulate in

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Table 1. Lengths (cm) and weights (g) of *S. commerson* samples obtained for analyses and years.

Years	Length (cm) (Min-Max)	Weight (g) (Min-Max)
2006-2007	46-49	618-768
2007-2008	46-51	616-742
2008-2009	46-51	612-768
2009-2010	46-50	608-742
2010-2011	46-50	612-731

their tissues [7-13]. Mansour and Sidky [14] reported fish are often at the top of the food chain and have the tendency to concentrate heavy metals from water. Therefore, bioaccumulation of metals in fish can be considered as an index of metal pollution in aquatic ecosystems [3, 15, 16]. That could be a useful tool to study the biological role of metals present at higher concentration in fish [17, 18].

The objective of this study was to determine the heavy metal concentration in muscles, liver, kidney, gills and gonads of *Scomberomorus commerson* collected from Karachi Fish Harbor. Moreover, the present study sought to establish the potential risk to humans due to consumption of contaminated fish with the heavy metals.

2. MATERIAL AND METHODS

Total of 294 fish samples were purchased from Karachi fish harbor during September 2006-December 2011. The lengths (cm) and weights (g) of fish samples obtained for analyses and years are summarized in Table **1**. The fish samples were collected and transported to the laboratory in ice boxes and stored at -10^{o}C until subjected for further analysis sample cleaned with sterile distilled water and then dissected. Fishes were dissected using steel scissors and scalpels to remove approximately 5 g dorsal muscles, entire liver, 2 rakers of gills, entire kidney and entire gonads were dissected, washed with deionized water and weighed fresh wt. (g). After taken fresh weight of samples were ground and calcinated at 500^{o}C for 3 hours until it turned to white or grey ash. The ashes were dissolved with 10 millilitres of 0.1 M HCl according to the method of Gutierrez *et al.* [19] and after which the dissolved ash residue was filtered with Whatman filter paper. 1 ml filtered solution diluted with 25 ml distilled water and make standards for elemental analysis. The equipment (Analyst 700) was started and prepared programme win lab 32 software. Three standards from 1000 ppm stock solution to 2 ppm, 4 ppm and 6 ppm were prepared. The equipment with the above mentioned standards were calibrated. The samples one by one were aspirated to detect the required metals. Finally the report using the software was prepared. Descriptive statistics (mean, standard deviation, range) and three-way analysis of variance (ANOVA) were conducted using excel software. The data transformations allowed for adjusting of all the zero values in the analytical results prior to the ANOVA test. A three-way ANOVA statistical procedure was employed in the assessment of variation in metal concentrations among fish species. Three-way Analysis of variance (ANOVA) in which the within sample variances and between the sample variances were considered, was utilized to test for the significance of differences between organs and between metals [20].

3. RESULTS AND DISCUSSION

The commercially important fish *S. commerson* was marketed by local fishermen in the most important fishing municipalities in Karachi Fish Harbor, Pakistan to determine eight heavy metals in the tissues and organs of the fish. Concentrations of Fe, Mn, Cu, Zn, Ni, Pb, Cd and Cr in muscles, liver, kidney, gills and gonads of analyzed fish species, expressed by mean value ± standard deviation and range are presented in Table **2**.

Liver showed high concentrations of metals in the tissues and organs of narrow-barred Spanish mackerel during all years of the present study. The highest mean concentration of Fe (608.93±113.22 µg/g), Mn (9.79±4.22 µg/g), Cu (38.57±16.62 µg/g), Zn (53.25±26.50 µg/g), Ni (3.22±1.13 µg/g), Pb (1.20±0.64 µg/g), Cd (2.03±0.91 µg/g) and Cr (1.93 + 0.87 µg/g) was determined in liver of fish. The distribution of these metals in the organs and tissues of *S. commerson* samples is highly non-homogeneous (Table **3**). These results suggest that the mechanism of entry in to the body is mainly through digestive tract, which seems justified by the ingestion of food. There is general concern about the build-up of relatively high concentrations of metals in predators at the top of food chains. It is important to note that this fish species diet is based on small fishes like anchovies, clupeids, carangids, also squids and shrimps. The concentrations of the non-essential metals Cd and Pb were generally lower than those in other metals in the fish analysed. Both metals were found to largely accumulate in the liver. In the meantime, kidney like liver could also be the major reserve organ of any of the metals. Landis and Yu [21] pointed out that liver plays a central role in metabolizing chemicals foreign to the body and is one of the most important metal storage organs by the digestive tract. It should be emphasised that exposure time is important determinant of toxic effects. The lowest mean concentrations of Cd (0.19±0.06) and Pb (0.14±0.12 µg/g) were recorded in the muscle tissues.

It is clear that the gills are also one of the major organ of accumulation of heavy metals, since they are in direct contact

Table 2. Mean concentration of heavy metals (µg/g) in *Scomberomorus commerson* fishes from the coast of Karachi (2006-2011).

Metals	Years	N	Muscles (Mean± SD)	Liver (Mean± SD)	Kidney (Mean± SD)	Gills (Mean± SD)	Gonads (Mean± SD)
Fe	2006-2007	61	27.09±14.67	379.88±134.10	43.03±18.92	13.73±7.13	25.21±14.26
	2007-2008	58	35.77±19.09	301.06±91.80	48.36±23.03	17.09±9.36	23.38±9.18
	2008-2009	60	24.82±18.58	469.24±126.54	47.09±28.54	27.14±18.82	24.13±15.24
	2009-2010	60	44.40±21.22	518.89±128.80	38.91±22.56	37.78±21.85	25.68±13.91
	2010-2011	55	23.71±19.59	608.93±113.22	45.13±19.88	48.31±21.12	32.21±19.14
Mn	2006-2007	61	1.41±0.72	4.35±2.03	2.14±1.02	1.50±1.10	1.71±0.82
	2007-2008	58	1.70±1.59	8.62±2.62	2.93±1.77	2.19±1.17	2.16±1.03
	2008-2009	60	1.30±1.27	8.82±2.71	2.26±2.03	3.91±1.95	2.67±1.29
	2009-2010	60	2.20±1.59	9.79±4.22	3.32±2.08	2.71±1.88	2.41±1.35
	2010-2011	55	2.01±1.66	5.35±2.55	3.66±2.48	2.92±2.17	2.63±1.90
Cu	2006-2007	61	2.78±2.33	31.20±12.89	2.69±2.50	1.59±0.76	2.46±1.78
	2007-2008	58	6.83±3.98	31.18±14.59	2.78±1.62	1.85±0.90	2.39±1.94
	2008-2009	60	6.12±2.87	32.67±17.03	3.17±2.87	2.77±1.45	2.04±1.68
	2009-2010	60	6.65±2.53	37.83±14.02	4.41±3.38	6.54±3.95	3.17±2.43
	2010-2011	55	5.52±2.39	38.57±16.62	3.93±2.80	6.98±3.78	3.64±2.31
Zn	2006-2007	61	3.17±1.96	16.46±9.90	3.02±2.73	3.74±2.45	2.76±2.11
	2007-2008	58	4.99±2.66	30.85±17.64	2.76±1.98	3.41±1.05	2.56±1.72
	2008-2009	60	8.94±4.22	43.93±20.31	3.83±2.35	2.95±1.71	3.35±2.34
	2009-2010	60	7.16±5.68	53.25±26.50	4.50±3.66	2.90±1.93	3.13±1.98
	2010-2011	55	9.43±5.76	31.33±19.23	3.75±1.63	3.14±2.88	4.14±1.88
Ni	2006-2007	61	0.16±0.12	1.61±0.66	0.26±0.18	0.33±0.21	0.58±0.16
	2007-2008	58	0.34±0.18	1.44±0.86	0.35±0.20	0.45±0.24	0.55±0.17
	2008-2009	60	1.48±0.28	2.17±0.96	0.36±0.29	0.33±0.25	0.54±0.21
	2009-2010	60	0.57±0.31	2.45±0.32	1.40±0.42	0.37±0.29	0.42±0.30
	2010-2011	55	0.62±0.41	3.22±1.13	0.46±0.32	0.32±0.20	0.36±0.28
Pb	2006-2007	61	0.14±0.12	0.41±0.22	0.16±0.10	0.21±0.16	0.28±0.12
	2007-2008	58	0.22±0.14	0.58±0.26	0.23±0.17	0.32±0.22	0.46±0.22
	2008-2009	60	0.34±0.23	1.20±0.64	0.28±0.16	0.33±0.24	0.42±0.22
	2009-2010	60	0.40±0.30	0.85±0.65	0.41±0.22	0.56±0.22	0.42±0.26
	2010-2011	55	0.54±0.37	1.01±0.94	0.33±0.20	0.49±0.23	0.63±0.37
Cd	2006-2007	61	0.19±0.06	0.54±0.16	0.36±0.24	0.46±0.28	0.32±0.21
	2007-2008	58	0.36±0.25	1.11±0.52	0.51±0.27	0.48±0.29	0.43±0.26
	2008-2009	60	0.22±0.19	1.94±0.67	0.55±0.26	0.58±0.31	0.43±0.39
	2009-2010	60	0.57±0.31	1.14±0.75	0.40±0.31	1.51±0.50	0.43±0.32
	2010-2011	55	0.68±0.42	2.03±0.91	1.22±0.68	0.56±0.21	1.18±0.46

Table 2. contd…

Metals	Years	N	Muscles (Mean± SD)	Liver (Mean± SD)	Kidney (Mean± SD)	Gills (Mean± SD)	Gonads (Mean± SD)
Cr	2006-2007	61	0.17±0.08	0.78±0.39	0.42±0.26	0.26±0.10	0.24±0.15
	2007-2008	58	0.14±0.06	0.62±0.18	0.19±0.10	0.25±0.14	0.17±0.12
	2008-2009	60	0.38±0.24	1.45±0.56	0.25±0.16	0.17±0.10	0.28±0.20
	2009-2010	60	0.44±0.20	1.66±0.63	0.29±0.26	0.74±0.31	0.46±0.33
	2010-2011	55	0.51±0.22	1.93±0.87	0.42±0.21	1.34±0.64	0.43±0.24

Table 3. Analysis of variance (ANOVA) in *Scomberomorus commerson* fish from the coast of Karachi.

Metals	Effect	Sum of square	df	Mean square	F	*P*
Fe	Organs	4.2417	4	1.0607	4436.901	.000
	Year	599955.781	4	149988.945	62.769	.000
	Organs * Year	1707501.243	16	106718.828	44.661	.000
	Error	7.1737	1368	2389.542		
	Total	4.9907	1468			
Mn	Organs	7428.249	4	1857.062	394.294	.000
	Year	760.316	4	190.079	40.358	.000
	Organs * Year	913.597	16	57.100	5.284	.000
	Error	6452.485	1370	4.710		
	Total	39410.926	1470			
Cu	Organs	260511.938	4	65127.985	1373.996	.000
	Year	3937.988	4	984.497	20.770	.000
	Organs * Year	2895.251	16	196.418	4.144	.000
	Error	64891.163	1369	91.647		
	Total	531383.189	1469			
Zn	Organs	271966.105	4	67991.526	906.882	.000
	Year	13947.159	4	3486.790	46.507	.000
	Organs * Year	34366.097	16	2147.881	28.649	.000
	Error	102712.825	1370	74.973		
	Total	663680.190	1470			
Ni	Organs	768.244	4	192.061	523.130	.000
	Year	33.319	4	8.330	22.688	.000
	Organs * Year	95.034	16	5.943	.685	.951
	Error	502.980	1370	.367		
	Total	2482.335	1470			
Pb	Organs	82.783	4	20.696	127.128	.000
	Year	28.402	4	7.101	43.617	.000
	Organs * Year	10.783	16	.674	4.140	.000
	Error	277.099	1395	.163		
	Total	748.763	1495			
Cd	Organs	95.645	4	23.911	155.291	.000
	Year	20.134	4	5.033	32.689	.000
	Organs * Year	11.456	16	.325	2.109	.000
	Error	210.949	1370	.154		
	Total	932.161	1470			

Table 3. contd…

Metals	Effect	Sum of square	df	Mean square	F	*P*
Cr	Organs	383.021	4	95.755	618.573	.000
	Year	71.644	4	17.911	115.703	.000
	Organs * Year	35.873	16	2.242	14.484	.000
	Error	212.076	1370	.155		
	Total	1468.717	1470			

with the water and therefore are the first barrier of defence and because gills are the first organs to be exposed to suspended particles in the water column.

Fe is the most accumulated in all tissues and organs. The essential metals, particularly Mn, Zn and Cu were evenly distributed across the different fish tissues and organs (Table **2**). Zn, Cu and Ni levels were higher in the muscles than those in other tissues except liver. The order of abundance of the metals in the fish samples based on concentrations in the muscle tissues analysed were as follows: Fe>Zn>Cu> Mn>Ni>Cd>Pb≥Cr. Recently, Bat [22] reviewed the results of the selected heavy metals (Fe, Zn, Ni, Cu, Mn, Pb, Cd and Co) in fish captured from the Black Sea coasts of Turkey and the metal concentrations decrease in the order Zn>Fe>Cu> Mn>Pb>Ni>Co>Cd. Below is a summary of the general distribution of the eight heavy metals in the tissues and organs:

Fe: Liver> Kidney>Muscles> Gills> Gonads

Mn: Liver> Kidney> Gills> Gonads>Muscles

Cu: Liver>Muscles>Gills >Kidney>Gonads

Zn: Liver>Muscles> Kidney> Gills> Gonads

Ni: Liver>Muscles>Kidney>Gonads>Gills

Pb: Liver>Gonads>Gills>Muscles>Kidney

Cd: Liver>Gills>Kidney>Gonads>Muscles

Cr: Liver>Gills>Muscles>Kidney>Gonads

In general, results of the present study show a trend similar to what many authors have pointed out before around the world, about the accumulation of these metals in fish tissues [7, 10, 23-29]. Also, authors agree that liver is the highest concentrations of heavy metals analysed are shown.

Legal thresholds are not available for essential elements in Europe. Commission Regulation (EC) [30] indicates that maximum level is 0.30 mg kg^{-1} wet weights for Pb. However, in the muscle tissues of fish the mean metal concentrations exceed the permissible limits of Pb established by Commission Regulation (EC) [30], but was still acceptable according to Georgian and Russian Federation limits for Pb (mg/kg wet weight) which is 1.0 [31, 32]. Pb contents in the fish samples were within the permissible limits.

The Joint FAO/WHO Expert Committee on Food Additives established a PTWI for Pb of 0.025 mg/kg body weight/week which was equivalent to 1.725 mg/week for a 70 kg adult [33]. In Pakistan, where the average weekly consumption of fish is 35 g per person and the maximum Pb levels in muscle tissues of *S. commerson*, weekly intake calculated ranged from 0.005±0.004 mg (35 g x 0.14±0.12 mg/1000 g) in 2006-2007 to 0.019±0.013 mg (35 g x 0.54±0.37 mg/1000 g) in 2010-2011, which is less than the maximum permissible intake of Pb established by FAO/WHO [33]. Similarly, Tabinda *et al.* [34] showed that Pb levels in *Pampus argenteus* and *Tenualosa ilisha* from Keti Bunder, south-east of Karachi in Thatta district of Sindh province, Pakistan exceeded the recommended limits.

In case of Cd, Commission Regulation (EC) [30] indicates that maximum level is 0.05 mg kg^{-1} wet weight. The permissible limits for Cd Moreover, the Joint FAO/WHO Expert Committee on Food Additives established a PTWI for Cd of 0.007 mg/kg body weight/week which was equivalent to 0.49 mg/week for a 70 kg adult [33]. By using the means of weekly fish consumption in Pakistan of 35 g per person and the maximum Cd levels in muscle tissues of *S. commerson*, weekly intake calculated ranged from 0.007±0.002 mg (35 g x 0.19±0.06 mg/1000 g) in 2006-2007 to 0.024±0.012 mg (35 g x 0.68±0.42 mg/1000 g) in 2010-2011. As regards Cd concentrations in the muscle tissues of *S. commerson*, the estimated PTWI in the present study is higher than permissible limits and seems not to be appropriate for human health. Here, the results are similar to those reported by Raza *et al.* [35]. They found that Cd was found maximum in *Parastromateus niger* from northwest coastal area of Karachi. It is possible that short-term exposure to this metal doesn't cause immediate health threats to humans. It is also worth pointing out that metal contents were expressed as $\mu g.g^{-1}$ dry weight in the present study.

Results of these previous studies and the present study confirm that heavy metal concentrations in marine organisms have increased in coast of Karachi (see Figs. **1-8**). In the present study an approx. 1.7-1.8 fold increase of Ni and Zn levels in the fish samples in 2010-2011 compare with 2006-2007 (Figs. **4-5**). The results showed that Fe and Mn have increased 1.5 fold and Pb and Cr have increased 2.5 fold in 2010-2011 compare with 2006-2007 (Figs. **1**, **2**, **6** and **8**). It was also observed that Cu and Cd have increased 1.44 fold and 3 fold, respectively during past five years (Figs. **3** and **7**). Heavy metal pollution of the Karachi coasts has become an important problem because of industrialization processes and pollutant loads from two main rivers discharging into the sea [35].

CONCLUSION

The heavy metal concentrations except Cd in the commercially important fish do not present any danger to human

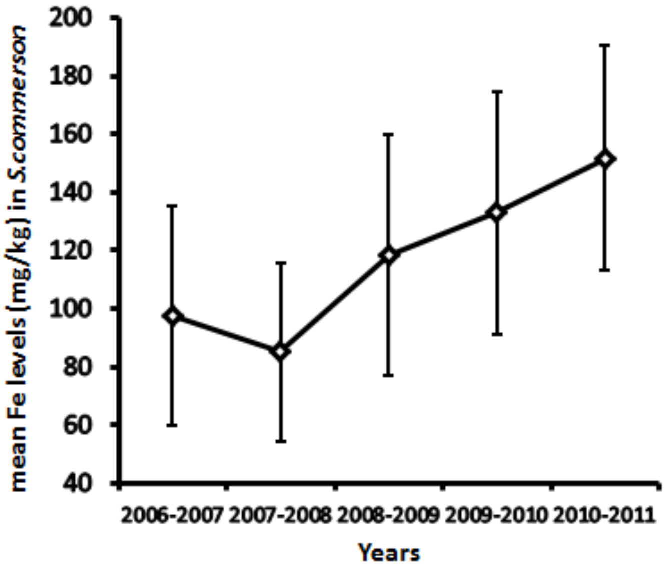

Fig. (1). Fe levels (mean± SD) in *S. commerson* from Karachi coast between 2006 and 2011.

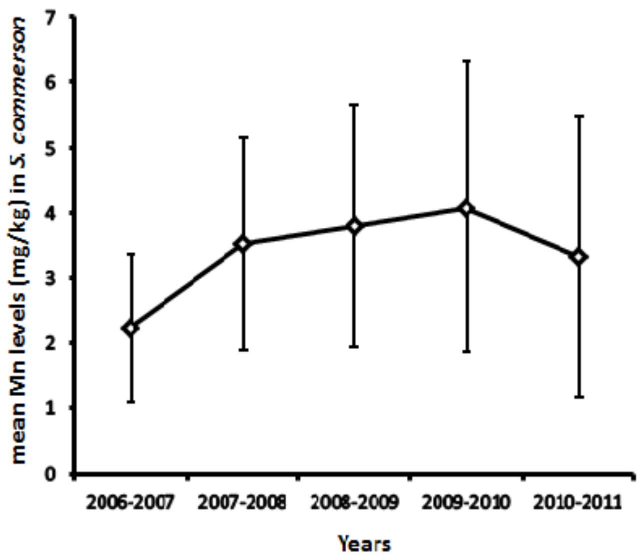

Fig. (2). Mn levels (mean± SD) in *S. commerson* from Karachi coast between 2006 and 2011.

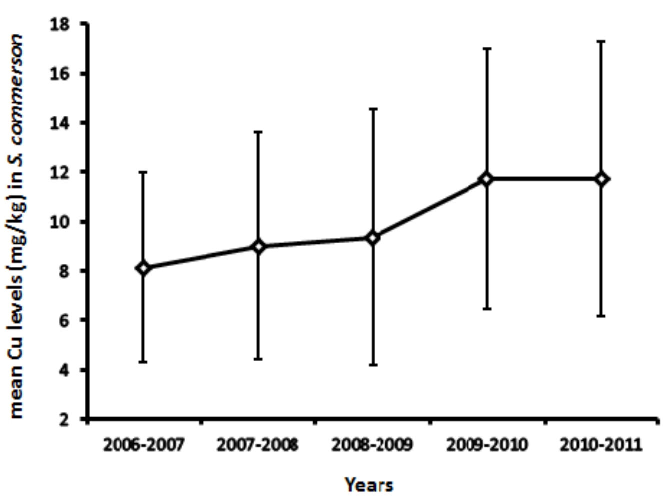

Fig. (3). Cu levels (mean± SD) in *S. commerson* from Karachi coast between 2006 and 2011.

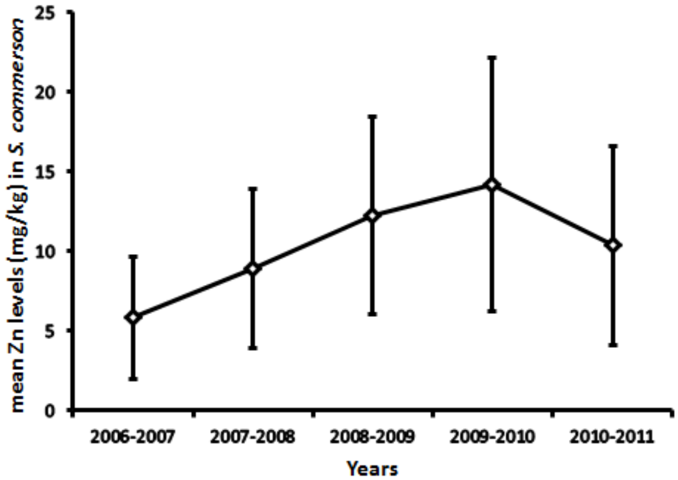

Fig. (4). Zn levels (mean± SD) in *S. commerson* from Karachi coast between 2006 and 2011.

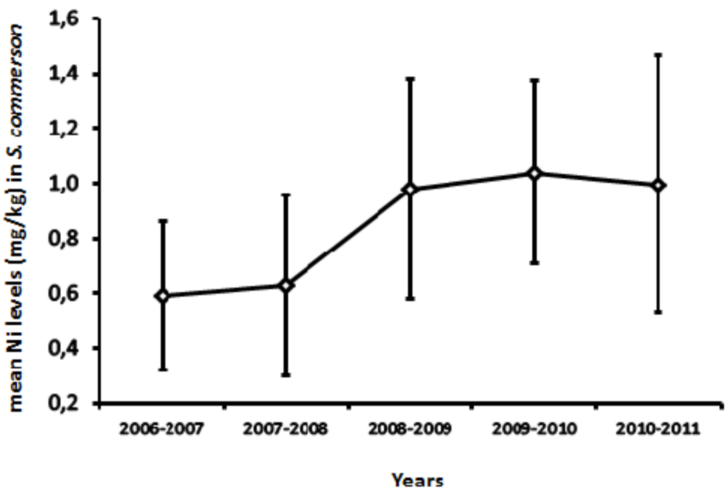

Fig. (5). Ni levels (mean± SD) in *S. commerson* from Karachi coast between 2006 and 2011.

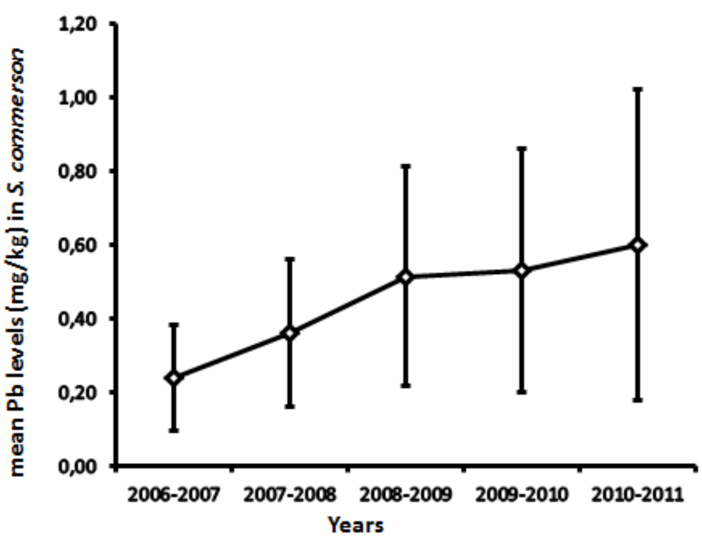

Fig. (6). Pb levels (mean± SD) in *S. commerson* from Karachi coast between 2006 and 2011.

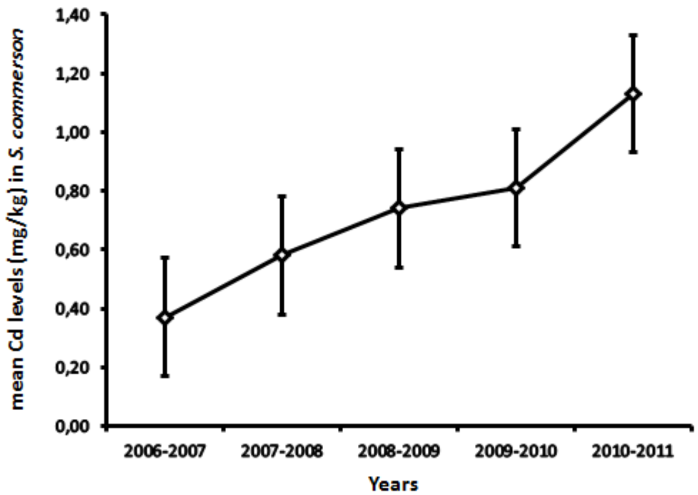

Fig. (7). Cd levels (mean± SD) in *S. commerson* from Karachi coast between 2006 and 2011.

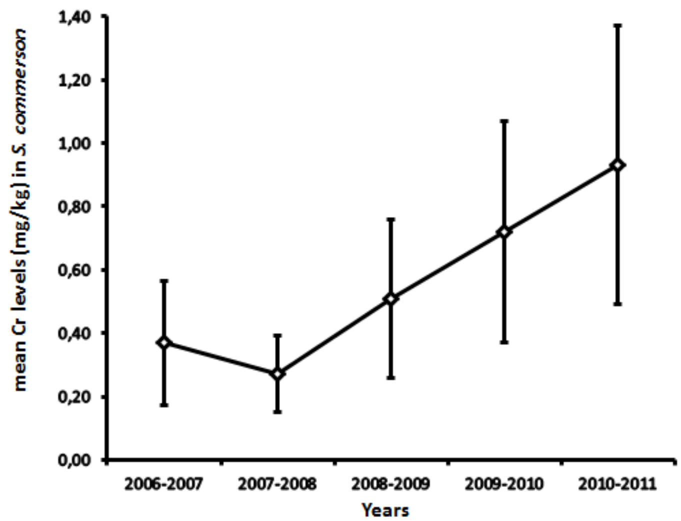

Fig. (8). Cr levels (mean± SD) in *S. commerson* from Karachi coast between 2006 and 2011.

health. However, in the presence of intense local pollution in the sea, the pollution of the commercial fish may well attain the limiting concentrations. Another relevant consideration is the proportion of fish in diets in different countries and regions. In the light of the available data and the anthropogenic activities in the natural levels and relative proportions of the metals in the marine environment, investigations of marine organisms, especially those of commercial importance, must be continued.

CONFLICT OF INTEREST

The authors confirm that this article content has no conflict of interest.

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Declared none.

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