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Health risk assessment of mercury and arsenic associated with consumption of fish from the Persian Gulf

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Abstract Concentrations of mercury and arsenic in fish from the Persian Gulf were determined by graphite furnace atomic absorption spectrometry. Concentrations of the metals in muscle samples were 0.049–0.402 μg g^{-1} for mercury and 0.168–0.479 μg g^{-1} for arsenic, with means of 0.133 and 0.312 μg g^{-1}, respectively. The maximum daily consumption rate (grams per day) and meal consumption limit (meals per month) was calculated to estimate health risks associated with fish consumption. According to the results, the maximum allowable consumption rate varies between 8–56 and 15–96 g/day base on mercury and arsenic content, respectively. The results of this study indicate that the concentration of mercury and arsenic is well below the maximum permissible levels for mercury (0.5 μg g^{-1}) and arsenic (6 μg g^{-1}) according to international standards.

Keywords Mercury · Arsenic · Persian Gulf

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Introduction

Mercury and arsenic are considered the most important form of pollution of the aquatic environments because of their toxicity and accumulation by marine organisms (Emami Khansari et al. 2005). Mercury and arsenic discharged into the marine environment can damage aquatic species, ecosystems, and consumers due to their toxicity and accumulative behavior (Tuzen 2009). The accumulation of the metals vary widely in fish and other aquatic animals, depending on age, size, environmental conditions, the position of the species in the food chain, time of exposure, and pollution level (Raissy et al. 2011). Mercury and arsenic accumulate in organisms at the bottom of the food chain and experience biomagnifications up the food chain reaching its highest concentrations in top predator fish. Consequently, in addition to the ecological aspects, special attention must be paid to the public health.

It is generally accepted that consumption of fish and seafoods is one of the major sources of mercury and arsenic exposure for humans (Agah et al. 2010; Alina et al. 2012). Hence, it is important to investigate the levels of mercury and arsenic in these organisms to assess whether the concentration of mercury and arsenic is within the permissible amount and will not pose any hazard for human consumption. There is limited information on health risk assessment of mercury and arsenic via consumption of fish from the Persian Gulf. Therefore, the aim of this study was to determine the levels of mercury and arsenic in fish from the Persian Gulf and health risk assessment of mercury and arsenic associated with consumption of fish.

Materials and methods

Samples

A total of 80 fish including *Psettodes erumei* (n=40), *Otolithes ruber* (n=10), *Scomberomorus commerson* (n=10), *Lutjanus lutjanus* (n=10), and *Pamous argenteus* (n=10) were obtained from local fish market in Hendijan, South Iran during summer 2012. The samples were immediately transported to the laboratory in clean plastic containers filled with crushed ice. After measuring fish weight and size, dorsal muscle samples (10–20 g) were dissected from the fish (next to the dorsal fin) and were stored at −18 °C for analysis of mercury and arsenic concentrations.

Apparatus and reagents

A PerkinElmer model 4100 atomic absorption spectrometer equipped with a GTA graphite furnace and deuterium background corrector was used. Samples were injected into the graphite furnace using PerkinElmer AS-800 autosampler. The atomic absorption signal was measured as a peak height mode against an analytical curve. PerkinElmer Analyst 4100 model AAS equipped with CVAAS system was used for mercury and arsenic determination. The recoveries of the metals were determined by adding increasing amounts of mercury and arsenic to the samples and taking them through the digestion procedure.

All reagents and solvents were of analytical reagent grade (Merck, Germany). ASTM® type I water (from an ELGA® filtration system-ELGA LLC, USA) acidified to 1 % nitric acid was used to make the calibration blank and standards. The stock solutions of mercury and arsenic (1,000 mg/L) were obtained by dissolving appropriate metal salts (Merck, Germany). The working solution were freshly prepared by diluting an appropriate aliquot of the stock solutions using 1 M HCl and 5 % H2SO4 for diluting mercury solution and 7 M HCl for diluting arsenic solution. Stannous chloride, for mercury analysis, was freshly prepared by dissolving 10 g in 100 ml of 6 M HCl. The solution was boiled for about 5 min, cooled, and nitrogen bubbled through it to expel any mercury impurities (Voegborlo et al. 1999).

Digestion and determination of heavy metals

In the laboratory, the moisture content of the tissue samples was determined according to AOAC method in triplicate (WHO, 1993). Samples were digested using the wet digestion techniques (Oze et al. 2006), then the metals were determined against aqueous standards. Mercury and arsenic were determined using cold vapor atomic absorption spectrophotometer flow injection mercury/hydride analyzer (FIAS 4100, PerkinElmer) equipped with hollow cathode mercury lamp at a wavelength of 253.7 and 248.3 nm for mercury and arsenic, respectively (Table 1). The standard reference material (SRM) used in this study was dogfish (*Squalus* sp.) muscle, certified by the National Research Council of Canada as DORM-2. For each run, a duplicate sample, spiked samples, and two blanks were carried through the whole procedure. SRM was analyzed once for every three fish samples.

Statistical analysis

Data were transferred to Microsoft Excel (Microsoft Corp., Redmond, Washington, USA) for analysis. SPSS 18.0 statistical software (SPSS Inc., Chicago, Illinois, USA), was used for ANOVA test; differences were considered significant at values of $p<0.05$.

Calculation of daily consumption limits

Daily consumption limits were calculated according to the following equation. It shows allowable daily consumption of mercury and arsenic contaminated fish

Table 1 Instrument settings for analysis of arsenic

Working conditions	As
Wavelength (nm)	248.3
Slit width (nm)	0.2
Lamp current (mA)	10
Argon Flow (ml/min)	250
Injection volume (μl)	20
Heating program temperature °C (ramp time (s), hold time (s))	
Drying1	110 (1,20)
Drying2	154 (5,30)
Pyrolysis	1,150 (15,10)
Atomization	2,100 (0,5)
Cleaning	2,600 (1,2)

based on a contaminant's carcinogenicity, expressed in kilograms of fish consumed per day:

$$CR_{lim} = \frac{RfD \times BW}{C_m}$$

CRlim Maximum allowable fish consumption rate (kilograms per day)
RfD Reference dose (0.1 μg/kg/day for mercury and 0.3 μg/kg/day for arsenic)
BW Consumer body weight (kilograms)
Cm Measured concentration of chemical contaminant *m* in a given species of fish (milligrams per kilogram).

Calculation of meal consumption limits

The consumption limit is determined in part by the size of the meal consumed. A 0.227 kg meal size was assumed. The following equation can be used to convert daily consumption limits to the number of allowable meals per month:

$$CR_{mm} = \frac{CR_{lim} \times T_{ap}}{MS}$$

CRmm Maximum allowable fish consumption rate (meals per month)
CRlim Maximum allowable fish consumption rate (kilograms per day)
MS Meal size (0.227 kg fish/meal)
Tap Time averaging period (365.25 days/12 months=30.44 days/month).

Results

Detection limit is defined as the concentration corresponding to three times the standard deviation of ten blanks. The method detection limit for Hg and As was determined to be 0.0006 and 0.0017 μg/g for mercury and arsenic, respectively.

The relative standard deviations were less than 10 % for all investigated elements. The accuracy of the method was evaluated by means of trace element determination in SRM. The achieved results were in agreement with certified values. The mean recovery values of mercury and arsenic were 96.5 and 96.6 %, respectively (Table 2).

Table 2 Recovery of mercury and arsenic from shrimp samples

Metal	Concentration of the metal added ($\mu g\ g^{-1}$)	Concentration of the metal recovered ($\mu g\ g^{-1}$)	% Recovery
Mercury	0.010	0.0098	98
	0.020	0.019	95
	0.030	0.029	96.6
Arsenic	0.005	0.0049	98
	0.010	0.0097	97
	0.020	0.019	95

Data are mean of three samples of three replicates

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A total of 80 fish caught from the Persian Gulf were studied for mercury and arsenic content. The total length and weight of the studied fish ranged from 12–36 cm (20.8±0.77) and 88.6–822.7 g (279.3±69.3), respectively.

The concentration of mercury and arsenic in studied samples and the permissible limits are presented in Table 3. The results of this study indicated that the concentration varied from 0.049 to 0.402 with a mean of 0.133 $\mu g\ g^{-1}$ for mercury and from 0.168 to 0.479 with a mean of 0.312 $\mu g\ g^{-1}$ for arsenic. Mean level of both metals are lower than the maximum allowable levels (0.5 $\mu g\ g^{-1}$ for mercury and 1 $\mu g\ g^{-1}$ for arsenic), according to international standards (Commission of the European Communities 2006; ANZFA 1998; FAO/WHO 2004). The lowest mean concentration of mercury and arsenic was found in *L. lutjanus* and *S. commerson*, respectively, and the highest concentration of both heavy metals was found in *P. erumei* (Table 3). Statistical analysis of results by ANOVA showed no significant difference in heavy metal content of fish with different age, weight, and length (p>0.05).

Estimates of the health risks associated with consumption of Hg and As contaminated fish are presented according to daily (kilograms per day) and monthly (meals per month) limits for the 3 to 75-year-old population demographic (Table 4). According to the results, the maximum allowable fish consumption rate for an adult person with mean 71.5 kg body weight was 55 and 93 g/day based on mercury and arsenic concentration, respectively.

Discussion

Mercury and arsenic are global environmental pollutants which potentially accumulate in aquatic food web.

Table 3 Heavy metals concentration in studied samples (μg g^{-1})

Fish species	No. of fish	Heavy metal	Mean ± SD	Range	Permissible amount
Psettodes erumei	40	Mercury	0.151±0.056	0.058–0.402	Hg: 0.5 [a,b,c] As: 6 [a], 1[c,d]
		Arsenic	0.335±0.072	0.212–0.418	
Otolithes ruber	10	Mercury	0.115±0.048	0.098–0.242	
		Arsenic	0.319±0.052	0.202–0.387	
Scomberomorus commerson	10	Mercury	0.107±0.059	0.061–0.365	
		Arsenic	0.280±0.061	0.195–0.479	
Lutjanus lutjanus	10	Mercury	0.103±0.059	0.071–0.302	
		Arsenic	0.320±0.091	0.168–0.402	
Pamous argenteus	10	Mercury	0.136±0.039	0.049–0.278	
		Arsenic	0.309±0.062	0.170–0.413	
Total	80	Mercury	0.133±0.039	0.049–0.402	
		Arsenic	0.312±0.062	0.168–0.479	

[a]Commission of the European Communities 2006

[b]MAFF 1995

[c]FAO/WHO 2004

[d]Australian standard [ANZFA (1998)]

It is generally common that seafood is one of the major sources of heavy metals in the human food chain as about 95 % of the methyl mercury in humans is originated from the seafood (Houseuova et al. 2007). However, the efficiency of metal uptake from contaminated water and food may differ in relation to ecological needs, metabolism, and the contamination level of metal in water, food, and sediment as well as physicochemical factors of water such as salinity, temperature, and interacting agents (Canli and Furness 1993).

In this study, the concentration of mercury and arsenic was studied in some fish species from the Persian Gulf. According to the results, the highest concentrations of mercury and arsenic were found in *P. erumei*. Benthic fish species such as *P. erumei* may take up heavy metals from both water and sediments. These species have numerous pathways for accumulation of heavy metals including absorption at the gill surface, ingestion of water and sediment, and consumption of contaminated prey (Raissy et al. 2011). Thus, bioaccumulation of metals in benthic fish can be utilized as an indicator of environmental metals pollution.

Bioaccumulation and toxic effects of heavy metals in cultured and wild aquatic species have been studied in many countries (Raissy et al. 2010, 2011; Saei-Dehkordi et al. 2010; Zhiyou et al. 2010; Alina et al. 2012). The arsenic

Table 4 Maximum allowable fish consumption rate according to the mercury and arsenic content

Age (year)	Average body weight for males and females (kg)	Maximum allowable fish consumption rate (kg/day)		Maximum allowable fish consumption rate (meals/month)	
		Mercury	Arsenic	Mercury	Arsenic
3–6	11.6	0.008	0.011	1.072	1.475
6–9	25	0.018	0.024	2.413	3.218
9–12	36	0.027	0.034	3.620	6.168
12–15	50.6	0.038	0.048	5.095	6.436
15–18	61.2	0.046	0.058	6.168	7.777
18–25	67.2	0.050	0.064	6.704	8.582
25–35	71.5	0.053	0.068	7.107	8.716
35–45	74.0	0.055	0.071	7.375	9.520
45–55	74.5	0.056	0.071	7.509	9.520
55–65	73.4	0.055	0.070	7.375	9.386
65–75	70.7	0.053	0.067	7.107	8.984

concentrations in sardine, mackerel, and anchovy from Adriatic Sea were reported equal to 2.82–8.08, 0.56–1.06, and 2.59 μg g^{-1} of fresh weight, respectively (Jureša and Blanuša 2003). Results of another study showed that arsenic contents in the muscle of ten fish species collected from Manchar Lake and same species from Indus River in Pakistan were quantified from 2.11 to 14.1 μg g^{-1} on dried basis (Shah et al. 2009). Saei-Dehkordi et al. (2010) studied arsenic and mercury concentration in different fish species from the Persian Gulf. According to their results, the concentrations ranged between 0.156 and0.834 μg $g^{-1}$for arsenic and between 0.120 and 0.527 μg g^{-1} for mercury. The mean arsenic and mercury for *P. erumei* was 0.388 and 0.454 μg g^{-1}, respectively, which is similar to the results of our study (arsenic 0.230, mercury 0.129 μg g^{-1}). These results are also comparable to Agah et al. (2010). According to their study, the arsenic concentrations ranged between 0.2 and 2 μg g^{-1}in four fish species from the Persian Gulf including *Pomadasys* sp., *Platycephalus* sp., *Epinephelus tauvina*, and *P. argenteus* (mean 0.6 μg $g^{-1}$of wet weight). Khansari et al. (2005) reported that the canned tuna fish marketed in Iran had metal levels ranging from 0.0369 to 0.269 μg g^{-1} (mean 0.128 μg g^{-1}) for arsenic and from 0.043 to 0.253 μg g^{-1} (mean 0.117 μg g^{-1}) for mercury.

In this study, the mercury and arsenic concentrations ranged between 0.049 and 0.0402 and between 0.168 and 0.479, respectively. The results of this study showed that mercury and arsenic levels in our study were lower than mean mercury and arsenic levels in different fish species in other researches (Table 5), while comparing mean arsenic level demonstrates that our results were at least four times higher than those in the Hunan, China (Zhiyou et al. 2010) but was lower than those reported in many other researches including Persian Gulf. The variability of mercury and arsenic concentrations in different studies could be attributed to several factors including physiology of the examined fish, age and body weight, habitat, trophic level, and time of the study (Canli and Furness 1993; Raissy et al. 2012).

The maximum allowable fish consumption rate by person (3–75 year) was calculated on basis of an average 227 g (EPA 1999) of fresh fish muscle consumption per day. According to the results, depending on the consumer's age, the maximum allowable consumption rate varies between 8–56 and 15–96 g/day base on mercury and arsenic content, respectively. The maximum allowable consumption rate has been reported equal to 10–70 g/day for cultured fish from Taiwan base on the arsenic content (Liao and Ling 2003). Kannan et al. (1998) also found that consuming fish from South Florida Estuaries at rates greater than 70 g/day was estimated to be hazardous to human health.

The results obtained for mercury and arsenic in studied samples were acceptable for human consumption according to international standards (Commission of the European Communities 2006; ANZFA 1998; FAO/WHO 2004). Based on the international standards, there is no health risk with respect to the concentrations of mercury and arsenic in fish caught off the Persian Gulf. Considering the importance of the Persian Gulf, aquatic species from this area should be analyzed more often with respect to toxic metals from the human consumption and environmental points of view.

Table 5 Comparison of mercury and arsenic concentrations in fish in this study with other researches (μg g^{-1})

Location	Fish species	Metals levels	References
Persian Gulf (Iran)	Different species	Hg: 0.12–0.52 As:0.15–0.83	Saei Dehkordi et al., 2010
South Florida Estuaries (US)	Different species	Hg: 0.03–2.22	Kannan et al. 1998
Manchar Lake and Indus River (Pakistan)	Different species	As: 2.11–14.1	Shah et al. 2009
The Straits of Malacca (Malaysia)	*Psettodes erumei*	Hg: 1.7 and 3.7 As: 0.59 and 1.06	Alina et al. 2012
Persian Gulf (Iran)	*Psettodes erumei*	Hg: 0.07	Rezayi et al. 2011
River Bravona (France)	*Salmo trutta*	As: 0.62–0.13	Foata et al. (2009)
Hunan (China)	Different species	Hg: 0.0027–0.243 As: 0.009–0.152	Zhiyou et al. 2010
Persian Gulf (Iran)	Different species	Hg: 0.049–0.402 As: 0.168–0.479	This study

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