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Mercury Content in Shrimp and Fish Species from the Gulf Coast of Saudi Arabia

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Mercury is one of the most common and persistent heavy metals in aquatic environments. It can enter the aquatic ecosystem either as a result of human activity or from natural sources such as degassing of the earth's crust, emissions from volcanoes and evaporation from natural bodies of water. The worldwide mining of mercury is estimated to yield about 10,000 tons/year, whereas natural emissions of mercury has been estimated to be of the order of 2700-6000 tons per year (WHO 1990). The change in mercury speciation from inorganic to methylated forms is the first step in the aquatic bioaccumulation process. Once methyl mercury is released, it enters the food chain by rapid diffusion and tight binding to proteins. Fish can become a depository for methyl mercury directly through the water or by bioconcentration. Because methyl mercury has a biological half-life of 2 years, it can be a persistent contaminant in fish (Hazards of mercury 1971). Severe neurological illnesses were reported following the consumption of methylmercury- contaminated fish and shellfish (Matsumoto et al 1965; WHO 1990; Harada 1995; Mergler et al 1998; Clarkson 1998). After the tragic incident of mercury poisoning at Minamata, Japan, a number of investigations have been carried out to study mercury pollution in aquatic ecosystems throughout the world (Raldua and Pedrocchi 1996; Ali et al 1997; Joiris et al 1995, 1997a,b; Pinkney et al 1997; Henry et al 1998; Olivero et al 1998; Han et al 1998; Svobodova et al 1999). Comparatively, little information is available for mercury levels in marine species from the Arabian Gulf (Khordagui and Al-Ajmi 1991; Ahmad and Al-Ghais 1996). There is a major concern about the contamination of Arabian Gulf fishes with heavy metals and polycyclic aromatic hydrocarbons after the Gulf war in 1991. Furthermore, the presence of major petroleum industries in the Eastern Province might contribute significantly to contamination of aquatic environment.

Total fishery production of the Kingdom of Saudi Arabia in 1997 was 53,170 metric tons and the production in the Arabian Gulf was 22,875 metric tons (Fisheries statistics of Saudi Arabia 1997). The ten major species fish groups caught in 1997 for the Arabian Gulf were shrimps, emperors, king fishes, seabreams, groupers, scads-jacks-trevallies, rabbitfish, snappers-jobfishes, marine crabs and tunas.

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This study is undertaken in an attempt to provide some information on the level of mercury in four different fish species and one crustacean commonly consumed by the population of the Arabian Gulf at three different sites where there are agricultural, municipal, and petroleum industrial activities.

MATERIALS AND METHODS

A total of 55 fish individuals and 15 shrimps were caught during the period 16/11/1998 until 05/12/1998 at three locations along the coastal Arabian Gulf of the Eastern Province of Saudi Arabia. Mainly agricultural and municipal wastes influenced Dammam and Sharq-dareen sites (an approximate distance of 26km between the two sites). Industrial production was the source of pollution of Mounifah site. The latter was located 160 km from Dammam and Sharq Dareen sites. The fish and shrimp species were presented in Table 1. They are commonly consumed by the local population in Saudi Arabia. A composite sample for each species was prepared and homongenized from the whole tissue of 15 specimens in a food processor and stored at –20C, except for Mounifah site where no doublebar-bream fish was caught and the total number were 10. Reference mercury standard was purchased from Plasma Standard (Philips Scientific York Street, Cambridge CB1 2PX England).

Table 1. Studied fish species.

Common Name	Scientific Name
Shrimps	Penaeus semisulcatus
Emperors	Lethrinus miniatus
Greasy-grouper	Epinephelus tauvina
Rabbitfish	Siganus canaliculatus
Doublebar-bream	Acanthoparagus bifasciatus

Mercury measurements were performed using the ICP-701 Inductively Coupled Plasma emission spectrometer equipped with the hydride generation system (ATI UNICAM, Cambridge, UK) and 221 XL Gilson autosampler (Gilson, 95400 Villiers le Bel, France). A weighed sample of approximately 1.0 g whole fish tissue were reacted with 5 ml of concentrated nitric acid into a 100 ml reflux Pyrex digestion tubes. An automated Digestion system 12,0019 with a 1012 Autostep Controller (Tecator AB, Hoganas, Sweden) was programmed as follows: 10 minutes ramp to 150°C and hold for 5 minutes. When the digestion was complete, and the tubes cooled at room temperature for two hours, 3 ml of 30% hydrogen peroxide (Fisher Scientific Co., A.C.S.) was added to each sample. The mixture was heated to 100°C for 5 minutes and hold for 20 minutes. After cooling for two hours at room temperature and then kept overnight at 4°C, the digestate was diluted to 10 ml with deionized water and filtered through filter paper (Whatman No. 541). The sample is then ready for analysis. Five replicates of each species were prepared for the analysis of mercury from each of the three sampling sites. Triplicate determinations by the ICP were made on all samples.

Working standard solutions were made up each day in the range 2.0 to 16.0 μg/L using 50% nitric acid solution. A calibration curve of emission intensity versus concentration of mercury was drawn and the concentrations of the unknown samples were read from the calibration graph. There was a good linear relation between emission intensity and standard concentration of mercury. Linear calibration curves were generated with linear correlation coefficients between 0.999 to 1.0. Mercury was analyzed in five fish sample of each species. Concentrations were expressed per weight as μg/g wet weight of fresh fish.

The accuracy of the method was determined by measuring the recovery of mercury added to fish samples. These spiked fish samples were run with the test samples using the same analytical procedure. The analytical recovery for mercury at the various concentrations tested (0.05, 0.1, and 0.2 μg/g) was 97-98%.

The detection limit for mercury in fish samples of 1g according to the method used was 0.004μg/g. The calculated detection limit, defined as 2 standard deviation above the average blank signal.

RESULTS AND DISCUSSION

The mercury concentration found in each fish species from three sampling sites are summarized in Table 2. No significant variations in mercury levels were noted among the three sites ($p>0.05$), possibly reflecting similar pattern of pollution received by waters of Arabian Gulf. Kruskal-Wallis one way analyses of variance were carried out to test whether there is a variation in the concentration of mercury among the different species within each sampling site (Table 2). Significant differences were noted. In Dammam and Mounifah sites, the highest mercury concentration was found in Greasy Grouper, whereas Rabbitfish had the lowest mercury concentrations in the three studied sites. On the other hand, Doublebar-bream sampled from Sharq-dareen site had the highest mercury concentrations.

Table 2. Arithmetic means ± SD (μg/g wet wt.) and ranges of mercury in the whole tissues of four species of fish and one crustacean from the Arabian Gulf classified by area.

Sampling Site	Shrimps (n=5)	Emperors (n=5)	Rabbitfish (n=5)	Doublebar-bream (n=5)	Greasy Grouper (n=5)	Test-Statistic (p)
Dammam	0.022 ± 0.008 (0.015-0.036)	0.055 ± 0.009 (0.044-0.067)	0.002 ± 0.004 (<DL-0.008)	0.038 ± 0.004 (0.033-0.042)	0.105 ± 0.009 (0.096-0.12)	22.922 (0.0001)
Mounifah	0.012 ± 0.001 (0.011-0.014)	0.059 ± 0.006 (0.054-0.066)	0.006 ± 0.001 (0.004-0.007)	ND	0.111 ± 0.013 (0.099-0.131)	17.884 (0.0005)
Sharq-dareen	0.039 ± 0.012 (0.027-0.056)	0.052 ± 0.002 (0.049-0.053)	0.001 ± 0.001 (<DL-0.003)	0.198 ± 0.011 (0.182-0.212)	0.028 ± 0.003 (0.025-0.032)	21.582 (0.0002)

Number between parenthesis represent the replicate of sample analysis that was taken from the composite of each species.

Table 3. Mercury in the whole tissue of four selected fish species and one shrimp (μg/g wet wt.).

Species	Dammam	Mounifah	Sharq-dareen	Test-statistics (p-value)
Double-bream (n=10)	0.038 ± 0.004* (0.033-0.042)**	N/A	0.198 ± 0.011 (0.182-0.212)	6.859 (0.009)
Emperors (n=15)	0.055 ± 0.009 (0.044-0.067)	0.059 ± 0.006 (0.054-0.066)	0.052 ± 0.002 (0.049-0.053)	5.333 (0.07)
Greasy Grouper (n=15)	0.105 ± 0.009 (0.096-0.12)	0.111 ± 0.013 (0.099-0.131)	0.029 ± 0.003 (0.025-0.032)	3.654 (0.008)
Rabbitfish (n=15)	0.002 ± 0.004 (<DL-0.008)	0.006 ± 0.001 (0.004-0.007)	0.001 ± 0.001 (<DL-0.003)	6.469 (0.039)
Shrimps (n=15)	0.022 ± 0.008 (0.015-0.036)	0.012 ± 0.001 (0.011-0.014)	0.039 ± 0.012 (0.027-0.056)	11.501 (0.003)

* Arithmetic means ± SD
** Ranges

Therefore, it was decided to pool the results of each species within each sampling site. Apart from Emperors, Kruskal-Wallis one way-analysis of variance showed statistical significant differences in mercury concentrations among the different species within each sampling sites (Table 3). In Dammam and Mounifah, the highest mercury concentrations was found in Greasy Gouper, whereas Rabbitfish had the lowest mercury concentration. The pattern of mercury concentrations among the four fish species and shrimp in Sharq-dareen site were in the following order: (Doublebar-bream 0.198 μg/g wet wt.) > Emperors (0.052 μg/g wet wt.) > Shrimps (0.039 μg/g wet wt.) > Greasy-grouper (0.029 μg/g wet wt) > Rabbitfish (0.001 μg/g wet wt.).

Graphical presentation of the average mercury concentrations, according to the species of the studied fish and one shrimp from each sampling site is shown in Figure 1. Variations in the contamination level of fish might be related to a number of factors such as age, size of fish, species, feeding habits, bioavalibility of chemicals in food and water and physio-chemical parameters of the aquatic environment (Pieters et al 1994; Ali et al 1997; Joiris et al 1997b; Castilhos et al 1998; Marins et al 1998; Bodaly and Fudge 1999; Svobodova et al 1999).

The levels of mercury reported here are lower than the corresponding proposed Saudi legislation and the U.S.FDA limits in fish and shellfish of 1 μg/g (FDA 1979; SASO 1997) and below the maximum permissible limit established for food by the Japanese and Brazilian legislation of 0.4 and 0.5 μg/g wet wt respectively (Nakagawa et al 1997; Kehrig et al 1998). However, when we calculated the daily intake of mercury, expressed as μg/day, obtained by multiplying the average quantity of fish consumed per Saudi family per day (9.967g) (Al-Nozha et al 1991) by the concentration of mercury in studied fish and shrimp and divided by 6 (the average Saudi family members), it was found

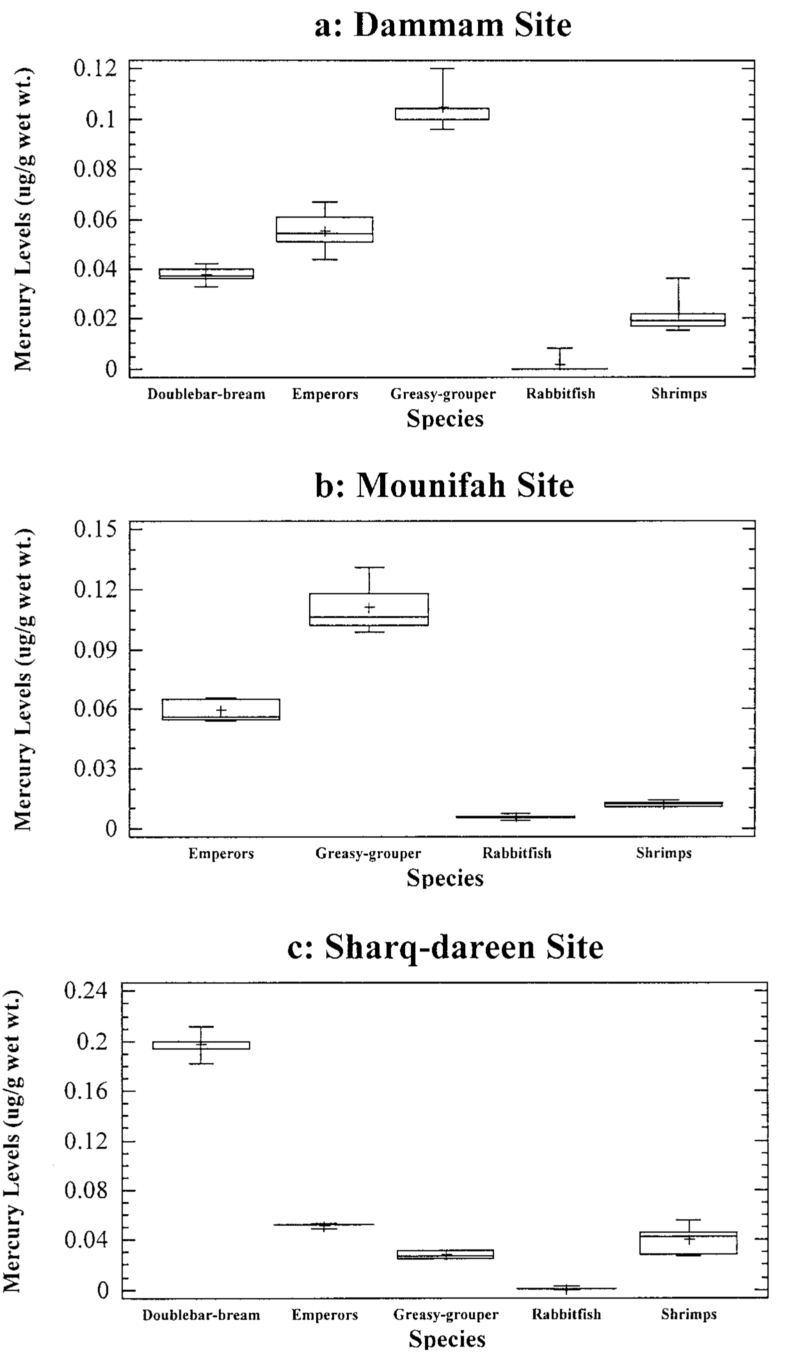

Figure 1. Box-plots of mercury concentrations (µg/g wet wt.) in the whole tissue of four fish species and one shrimp classified by sampling site: (a) Dammam Site; (b) Mounifah Site; and (c) Sharq-dareen Site. The mean lines represent the median, the "boxes" the central 50% region, the "whiskers" the total range.

that higher daily intake comes from the consumption of Doublebar-bream caught from Sharq-dareen site (Table 4). Consumption of Greasy-grouper caught from Dammam and Mounifah contributes also to higher daily intake of mercury. These figures were higher than the maximum acceptable intake of mercury in Saudi Arabia and the JECFA PTWI of 0.005 mg/kg body weight/week for a 60 kg adult (SASO 1997; MAFF 1998).

Table 4. Calculated daily intake of mercury (μg/day) from the four fish species and shrimp sampled.

Species	Dammam Mean	Dammam Max.	Mounifah Mean	Mounifah Max.	Sharq-dareen Mean	Sharq-dareen Max.
Double-bream (n=10)	0.063	0.069	-		0.329	0.352
Emperors (n=15)	0.091	0.111	0.098	0.11	0.086	0.088
Greasy-grouper (n=15)	0.175	0.199	0.184	0.218	0.048	0.053
Rabbitfish (n=15)	0.003	0.013	0.01	0.012	0.002	0.005
Shrimps (n=15)	0.037	0.059	0.02	0.023	0.065	0.093

It is reassuring that the reported mercury concentrations in the studied fish species from the Arabian Gulf is sufficient to be of concern regarding their consumption by the local population and its effect on health. Given the fact that frequent consumption of these four fish species and shrimp which are the most popular and economically important seafood in Saudi Arabia may increase the overall mercury intake to dangerous levels especially for those who depend only on seafood as their protein source. There is also a growing concern mercury exposure from high fish diet adversely affects the fetus or children's neurological development (Myers et al 1998; Murta et al 1999; Grandjean et al 1999). Further investigations are needed to assess the magnitude of the problem along other coastal regions of Saudi Arabia and to evaluate human health risks from mercury among high fish consumption populations in Saudi Arabia.

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