Published in final edited form as:

Mar Pollut Bull. 2017 August 15; 121(1-2): 143–153. doi:10.1016/j.marpolbul.2017.04.024.

Mercury accumulation in *Lethrinus nebulosus* from the marine waters of the Qatar EEZ

Ebrahim M.A.S. Al-Ansari^a, Mohamed A.R. Abdel-Moati^b, Oguz Yigiterhan^{a,*}, Ibrahim Al-Maslamani^a, Yousria Soliman^c, Gilbert T. Rowe^d, Terry L. Wade^e, Ismail M. Al-Shaikh^{a,1}, Ahmed Helmi^a, Ligita Kuklyte^d, Mark Chatting^a, and Mehsin A. Al-Ansi Al-Yafei^c

^aQatar University, Environmental Science Center, P.O. Box 2713, Doha, Qatar

^bMinistry of Municipality & Environment, Environmental Assessment Department, P.O. Box 39320, Doha, Qatar

^cQatar University, Department of Biology and Environmental Science, P.O. Box 2713, Doha, Qatar

^dTexas A & M University at Galveston Campus, Marine Biology Department, TX, USA

^eTexas A & M University at College Station, Geochemical and Environmental Research Group, TX, USA

Abstract

Total mercury (THg) and methylmercury (MeHg) were recorded in the commercial demersal fish *Lethrinus nebulosus*, caught from six locations in Qatar EEZ (Exclusive Economic Zone). Concentrations of THg decreased in the order: liver > muscle > gonad. THg concentrations in fish tissue ranged from 0.016 ppm in gonad to 0.855 ppm (mg kg⁻¹ w/w) in liver tissues, while concentrations in muscle tissue ranged from 0.24 to 0.49 ppm (mg kg⁻¹ w/w) among sampling sites. MeHg concentrations were used to validate food web transfer rate calculations. Intake rates were calculated to assess the potential health impact of the fish consumption. There is no major threat to human health from the presence of Hg in *L. nebulosus*, based upon reasonable consumption patterns, limited to no more than three meals of *L. nebulosus* per week.

Keywords

Mercury; Methylmercury; Bioaccumulation; *Lethrinus nebulosus*; Qatar EEZ; Arabian Gulf

1. Introduction

Mercury (Hg) is found in the environment due to natural and human activities, circulating naturally between earth's different spheres and reservoirs through physical and chemical processes (Fitzgerald et al., 2007). Industrial activities have increased the levels of Hg in the atmosphere and the oceans, and have contaminated some local environments, which cause decline of fish stocks (Nithyanandan et al., 2016). In the marine environment, Hg has long

*Corresponding author: oguz@qu.edu.qa (O. Yigiterhan).

¹ExxonMobil Research Qatar Inc., P.O. Box 22500, Doha, Qatar.

been recognized as a threat to human health (Withrow and Vail, 2007; Lubick and Malakoff, 2013) due to bioaccumulation and biologic transfer (biotransfer) of methylated toxic organic form with the formula CH_3HgX . It is a dangerous family of compounds that are often found in polluted water and arise by a process known as biomethylation (NRC, 2000).

Considerable data on total-mercury (THg) and methyl-mercury (MeHg) concentrations in the global marine environment has been previously reported, while limited studies has been conducted in the Persian/Arabian Gulf (hereafter the Gulf), which has hot, dusty, humid but arid climate (Subba Rao and Al-Yamani, 2000). This includes measurement in the sediments (Kureishy and Al-Muftah, 1999; de Mora et al., 2004; Kureishy and Ahmed, 1994; Al-Naimi et al., 2015), as well as body-burden in marine organisms, such as prawn, lobster, benthic, and pelagic fish species (Kureishy, 1991, 1993; Madany et al., 1996; Sadiq and McCain, 1993; de Mora et al., 2004) and marine plants (Kureishy and Al-Muftah, 1999). Pre-war studies surveys on heavy metals and organic contaminants, conducted by Fowler et al. (1993), have proved invaluable for the identification of pollution hot spots and assessing the consequences of the 1991 Gulf War in near shore sediments and biota. There has been the environmental impact of three wars in the past two decades. Several studies have compared Hg contamination prior to and after the 1991 Gulf War (listed in de Mora et al., 2004) on marine biota and coastal sediment. To assess the environmental consequence of the massive oil spill at the Northern shelf of the Gulf during the Iraqi invasion to Kuwait (Bu-Olayan et al., 1998), the concentrations of trace metals in seawater and particulate matter, as well as origins of potential contaminants and the quality of local seafood with respect to metal contamination, were investigated by de Mora et al. (2004) in the following years. The influence of fish sex and body length on the metal accumulation of those metals in the tested fish organs (liver, skin, and muscle) was critically investigated by Al-Yousuf et al. (2000) as an environmental indicator in water areas affected by human activities and as a monitoring technique for assessing the efficiency of control measures. To date, the only known area of high Hg contamination is the northern extremity of the Gulf, adjacent to industrial effluent discharge points (Haghighat et al., 2011). Sadiq (2002) reported similar finding which reflects very high Hg in the sediment samples from the immediate vicinity of the outfall, and decreased progressively away from it. At this location, the popular food-fish de-mersal Flounder is known to concentrate Hg in liver and muscle tissue adjacent to a chlor-alkali processing plant on the Iranian coast. High concentrations of MeHg have been also found in many apex predators fish caught for human consumption in the region (Agah et al., 2007). However, no other studies in the Gulf have reported concentrations above the widely-accepted WHO maximum consumption limit of 0.5 ppm of Hg wet weight (2.5 ppm dry weight) for sea food products (Agah et al., 2007).

Once entered the marine environment, Hg go through chemical and biochemical transformations into MeHg, a particularly hazardous form, that bioaccumulates and biomagnify in the food chain, with the highest concentrations reported at the top of the food chain, such as tuna fish and other top predators (Chen et al., 2008; Hammerschmidt and Fitzgerald, 2006a, 2006b; USGS, 2000; Taylor et al., 2014). The quantification of Hg biomagnification in marine organisms is poorly parameterized, although it is widely assumed that this is prevalent in the marine food chain, with increased concentrations at higher trophic levels (Booth and Zeller, 2005; Lamborg et al., 2002; Lamborg et al., 2003),

where > 95% is as the toxic form of MeHg at some cases (Koeman et al., 1973; Wagemann et al., 1998). Through a process of biologic transfer in the marine food chain, Hg can reach humans in the form of MeHg via the consumption of sea food (e.g. fish). Therefore, bioaccumulation followed by biomagnification of MeHg in the marine food webs serves as fast transport vectors to human population. Because of this reason, Hg and its forms are classified by the International Agency for Research on Cancer as possible carcinogenic to humans (IARC, 1993). Accordingly, there is international and national interests in monitoring Hg in the environment to ensure that Hg levels in marine biota do not exceed the levels set by WHO and environmental protection agencies for safe human consumption (WHO/UNEP DTIE Chemicals Branch, 2008).

The noticeable study by de Mora et al. (2004) in the Gulf region, focused on two fish species of commercial importance, namely the orange spotted grouper (*Epinephelus coioides*, known locally as “Hamoor”) and the spangled emperor (*Lethrinus nebulosus*, known locally as Sha’ri) together with selected biota. Another study by Al-Yousuf et al. (2000) focused on trace metals in liver, skin, and muscle of *Lethrinus lentjan* fish species in relation to body length and sex.

In this study, we aimed to measure and assess total- and methyl-Hg concentrations of the *L. nebulosus* in order to better understand the mercury accumulation at the Qatar’s Exclusive Economic Zone (EEZ). The fish species were sampled from both near-shore and off-shore locations which represent different pollution sources as well as distinct biogeochemical features in their marine waters. This is part of a large-scale study to address gaps in Hg data sets by gathering novel data from the Gulf. It extends to show the correlation between body size parameters and rate of Hg bioaccumulation; as well as other key factors enhancing bioaccumulation on the target fish tissue and organs. The most widely consumed fish species was investigated as an environmental indicator of aquatic ecosystem quality to evaluate potential risks to human health. This is quite important for detecting safe consumption limits of the most widely caught emperor or scavenger fish “*L. nebulosus*” (Forskål, 1775).

2. Methodology

2.1. Sampling

Qatar is located between 24° 28’ 12” N (min.) – 26° 9’ 36” N (max.) latitudes and 50° 45’ 0” E (min.) – 51° 37’ 48” E (max.) longitudes on the mid-western coastline of the Gulf (Fig. 1). It has long, shallow, productive shoreline, which supports rich fishing grounds together with its large EEZ.

L. nebulosus (spangled emperor, Fig. 2) fish samples were collected during the various cruises of the R/V *Mukhtabar Al Bihar* and the R/V *Janan* conducted in the Qatar’s EEZ in 2011 and 2012. Six sites were selected for fish sampling. The geographic names, numbers, and coordinates of the sampling stations are given in Fig. 1. The sampling areas were selected to represent quite different environments. The waters near Doha are influenced by the discharges from urbanized capital city, desalination plants, and heavy traffic by contrast to the other locations, where they are subjected to intense activity related to oil exploration, heavy industries, oil-gas exploitation-refinery-storage etc. Discharges of formation water

occur at Halul Island 100 km from the East cost of the peninsula. A total number of 69 individual *L. nebulosus* samples (24 for THg and 45 for MeHg) were collected at different months from the East Coast using new, clean fishing lines, and nets. Sampling was seasonal and reflecting both hot summer and winter periods. The sampling for Hg research was done in 2011 for THg species only. The sampling strategy was extended to cover MeHg analysis in 2012. This led us to collect fresh tissue samples in the following year, which was essential to prevent Hg loss in fish tissue, mostly observed by degradation of MeHg in samples > 6 months old. Due to limited sources, only MeHg species was analyzed on this 2nd set of samples; THg analysis has not been conducted.

Trace metal 'clean techniques' were applied at all steps in the sampling, preparation, preservation, and analysis procedure (e.g. U.S. EPA, 1996; Cullen et al., 2001). Fish samples were immediately chilled at dark in a double Ziploc bags, then transferred to a closed laboratory onboard the vessel for careful processing without sample contamination. The body size parameters; such as total body weight, organ weight (muscle, liver, gonad), body length, and age of fish were carefully measured, then samples were frozen immediately to prevent Hg loss, then stored in a deep freezer at -20 °C until analysis.

2.2. Analytical methods

Fish samples were analyzed for methyl- and total-mercury (MeHg, THg). Two different set of fish wet tissue samples, collected in 2011 and 2012, were analyzed from the same sites. The frozen samples were brought back to room temperature in a double plastic Ziploc bag. Fish were dissected in a laminar flow fume hood using clean stainless steel scalpels and Teflon dissecting forceps. The organs were collected as a whole from liver and gonads. These samples were carefully weighed on a digital balance. The skeletal muscle was extracted as much as possible after discharging skin, bones, organs, and fish head. The subsamples from dorsal skeletal muscle, liver, and gonad were collected. These wet tissue samples were later ground and homogenized prior to analyses using acid clean tools for Hg analysis.

MeHg was extracted from fish muscle tissues into water using nitric acid (HNO₃) digestion (Hammerschmidt, 2005; US EPA 821-R-01-013, 2001). Fish muscle tissue was analyzed according to the United States Environmental Protection Agency (U.S. EPA) Method 1630 (U.S. EPA, 1991). The non-desiccated (wet) muscle tissue samples were acid digested as it is, using 30% v/v HNO₃ before heating at 60 °C for 12 h. Sample pH was carefully adjusted using KOH, and the final volume was brought up to 20 ml prior to ethylation, approximately 1 ml of sample digest solution was taken for analysis and the vial was completed up to throat with no air bubbles and subsequent purging steps (Tseng et al., 2004; Hammerschmidt and Fitzgerald, 2005) before analysis. A Brooks Rand Merx System with Hg Speciation GC & Pyrolysis was utilized for MeHg analysis. The system was operated in conjunction Brooks Rand-Model III Automatic Fluorescence Spectrophotometer, Autosampler, and MeHg distillation system.

For THg analysis, a normal acid digestion with concentrated HNO₃ in accordance with EPA Appendix to Method 1631 (U.S. EPA 821-R-01-013, 2001) was used. The samples were immediately analyzed after digestion via Automated Hg analyzer gold trap technique

(CVAAS). Analyses were conducted using an AULA 254 Automatic Hg Analyzer (Gold Trap System) with Automatic Sample Digester for THg.

2.3. Precision and accuracy

Duplicate samples, spiked samples, and certified reference materials were digested and analyzed multiple times with each batch of samples for quality assurance. For certification of the MeHg content, SRM 2977 (Mussel tissue certified reference material for organic contaminants and trace elements), originated by National Institute of Standards and Technology (NIST) was used. For certification of the THg content of the fish tissue samples, DORM-4 (Fish protein certified reference material for trace metals) was used, provided by National Research Council Canada (NRC). Samples analyzed in duplicate shared a mean relative standard deviation of 9.6% for THg. The mean relative standard deviation for duplicate samples was 7.5% for MeHg. Recovery of known additions averaged 94.1% for MeHg and 97.8% for THg (Table 1). Mean measured concentrations of standard reference materials (SRM) were 6% below the SRM-NIST 2977 ($0.0366 \pm 0.001 \mu\text{g/g}$) and 2.4% below the SRM-DORM 4 ($0.412 \pm 0.036 \mu\text{g/g}$) certified reference values. Analytical precision was approximately 10% of the mean. In almost all cases, the average measured values were within the 95% confidence limit of certified values, and thus the accuracy of analysis was comparable to, or better than analytical precision.

2.4. Statistical analysis

Statistical analysis was done using R 3.1.2 software. A one-way analysis of variance (ANOVA) was used to investigate whether there were differences in THg and MeHg between sites, as well as THg and tissues sampled. Post-hoc tests were performed on data sets that showed significant differences using Tukey Honest Significant Difference (Table 2). Linear regression was used to examine any relationship between fish length and age with THg. Correlation of *L. nebulosus* fish biometric body size parameters with each other at 99% confidence levels, was given in Table 3.

3. Results

Dry and wet weight mean concentrations for two independent studies were given as $\mu\text{g/g}$ (ppm) for total-Hg (Table 4). The analysis showed that relatively similar THg concentrations were obtained when fish muscle was analyzed, using either wet or dry tissue analysis techniques. On the contrary, higher variations were observed for gonad and liver tissues between the application of dry and wet tissue analysis techniques. These results reflect that wet tissue analysis give about 4 times THg than the sample group dissected before analysis. Mercury accumulation in tissue depends on various spatial, temporal, and body size parameters. The variance can be due to difference in age and Hg uptake of the *L. nebulosus* fish samples selected at different locations for the comparison of dry and wet techniques. A summarized data was presented in Table 5 for wet weight mean THg concentrations in muscle, gonad and liver samples for male and female *L. nebulosus* from selected sampling location. Unfortunately, THg was not measured for Mesaieed and Doha sites at this stage of the research due to difficulties of sampling at stormy weather conditions. The highest mean THg concentrations were observed at liver tissue from Al-Ruwais site ($0.771 \pm 0.120 \text{ mg kg}$

$^{-1}$ w/w), followed by Ras Laffan site (0.554 ± 0.186 mg kg $^{-1}$ w/w), while lowest concentrations were observed for gonad tissue samples from the same sites (0.081 ± 0.003 and 0.065 ± 0.032 mg kg $^{-1}$ w/w, respectively). The gonad THg values were about 10-fold less than the liver values. The highest values of THg were measured in fish liver tissue (0.934 mg kg $^{-1}$ w/w, 3.85 years) of female fish from Ras Laffan, followed by another female fish from Al Ruwais (0.855 mg kg $^{-1}$ w/w, 5.37 years). These numbers are in good agreement with finding of similar studies (de Mora et al., 2004) for contaminated sites. When the average of all THg values was compared between the organs, it was observed that fish gonad (0.071 ± 0.031 mg kg $^{-1}$ w/w) has the lowest values, followed by muscle (0.349 ± 0.139 mg kg $^{-1}$) and liver (0.602 ± 0.192 mg kg $^{-1}$ w/w) tissues. These numbers are higher than a previous study conducted in the Gulf for various species where THg concentrations recorded in fish ranged from 0.0123 to 0.0867 mg kg $^{-1}$ w/w (0.0614 to 0.433 mg kg $^{-1}$ d.w.) (Kardovani, 1995). On the other hand, higher numbers, similar to this study, were reported for the studies from the Gulf region (Agah et al., 2007) and other seas (Zhu et al., 2013). A post-Gulf sea water pollution assessment program was carried out by Al-Yousuf et al. (2000) for the liver, skin, and muscle tissues of the localized *L. lentjan* fish species, showed that average metal concentrations in liver, skin, and muscle of female fish were found to be higher than those found in the male fish.

Theoretically, the wet weight tissue values (mg kg $^{-1}$ w/w or ppm) can be multiplied by five for an estimate of the concentration as a function of dry weight (Stephan et al., 1985). Concentration calculated assuming a moisture content of 80% (wet weight = $0.2 \times$ dry weight.), based on the average of moisture content reported for benthic infauna and fish species. Similar study conducted by Burger and Gochfeld (2006) also estimated comparable dry-weight concentrations for tissue samples by multiplying an average conversion factor of 5.4 (dry/wet). The results of wet weight versus dry weight-based mean Hg concentration variance comparisons are shown to determine if variance estimates for dry weight-based concentrations were less than those for wet weight-based values. The results from *L. nebulosus* THg analysis shows that removal of tissue moisture to produce dry weight-based tissue Hg concentrations does not produce any significant reductions in sample variance estimates (Table 4). Other data set shows similar findings for Largemouth bass and Yellow perch (MassDEP, Office of Research and Standards, 2006). The range of THg mean concentrations observed for muscle (0.025 ppm), gonad (0.076 ppm), liver (0.600 ppm) in Table 5 is similar to findings of Kureishy (1993) before and after the Gulf war measurements which reported *L. nebulosus* THg concentrations are 0.008, 0.007 ppm for muscle and 0.004, 0.004 ppm for liver.

Wet weight mean concentrations of *L. nebulosus* muscle tissue samples for MeHg with limits were given in the Table 6 for all sampling sites for male, female, and cumulative values. The table also reflects average biometric body size parameters with limits at each sampling site. The MeHg data vs selected biometric body size parameters - (a) weight, (b) length, (c) age was plotted for muscle tissue in Fig. 3. The cumulative and individual R-squared (R^2) values were shown above the linear trend lines to show coefficient of determination for each sampling set. The highest concentrations were detected at Ras Laffan station for the average of both male and female individuals, with 0.773 ± 0.216 ppm mean concentration. This number was followed by Halul Island sampling site with 0.450 ± 0.187

ppm mean concentration. The heaviest body size averages were recorded at Al-Ruwais and Doha sampling sites; which are also recorded as highest values 762 ± 299 and 747 ± 250 g for body weight and 38.3 ± 4.7 and 38.2 ± 4.8 cm for body length. It was also observed that *L. nebulosus* has mean minimum MeHg concentration of $0.071 \text{ mg kg}^{-1} \text{ w/w}$ and maximum concentration $0.320 \text{ mg kg}^{-1} \text{ w/w}$, which are used as selected values in Table 7 for MeHg exposure calculations.

The bioaccumulation of THg vs biometric body size parameters (weight, length, age) at three selected sampling sites for muscle tissue of *L. nebulosus* was plotted in Fig. 4. While the near-shore of Ras Laffan industrial area was giving the highest value of coefficient of determination ($R^2 = 0.832$), the highest THg concentrations were observed for the samples, collected from Al Khor sampling site, with slightly lower values of determination ($R^2 = 0.805$). It was clear that the heaviest ($\sim 1 \text{ kg}$) and oldest (~ 5.5 years) Sha'ri was accumulating as much as 5 times more THg than the smaller individuals from the Halul island off-shore station. Similar high accumulation rates (over 0.6 ppm MeHg) were detected in Fig. 5 for tissue samples collected from Mesaieed, which was the southernmost coastal sampling location.

A good representative of how THg accumulation varies between tissues was given in Fig. 6. It was clear stated that with about 1 ppm maximum Hg concentration, liver tissue has the highest Hg loading within the selected tissues. This was about $2\times$ more than muscle and $10\times$ more than gonad tissue accumulation rates with same body size parameters.

4. Discussion

Sources of Hg in the Gulf likely include waste (as a catalyst) from chloralkali plants and wastewater discharge from desalination and power plants where Hg cells are used in the electrolysis of the brine. Moreover, Hg is also introduced into the environment via regional and/or transcontinental Aeolian transport and deposition, which is a principal source of Hg to the ocean (Fitzgerald and Clarkson, 1991). Hg in the atmosphere is derived mainly from combustion processes e.g. burning biomass, petroleum, especially coal. Long-range atmospheric transport from these sources renders the Gulf as a critical ecosystem under extreme anthropogenic pressure that requires continuous monitoring for Hg contamination (Kureishy, 1993). Hg deposited or delivered to surface waters of the Gulf may be re-emitted to air, remain suspended or dissolved in the water column, be deposited either in sediments or absorbed-ingested by living organisms (Lusk et al., 2005).

L. nebulosus is quite well known and commercially important for many other Gulf nations and commonly spelled as Sha'ri (Carpenter et al., 1997) all around the Gulf. It is commonly caught at approximately 17 cm in tail length (TL), where adults commonly reach to 70.0 cm TL (Bouhlef, 1988), but specimens can grow up to recorded 87.0 cm TL male/unsexed (Assadi and Dehghani P., 1997). It's maximum published weight was recorded as 8.4 kg (IGFA, 2001) with the detected age of 28 years (Andrews et al., 2011). *L. nebulosus* inhabits in tropical marine waters, at preferably 28°C (Cheng et al., 2013); and at depths of between 10 and 75 m (Carpenter and Allen, 1989). It is a nonmigratory species, and is found on coral and rocky reefs, seagrass beds, mangrove swamps, as well as over sandy substrates. It feeds

mainly on mollusks, echinoderms, and crustaceans. It also eats polychaeta and other fish, but less commonly (Bailly, 2015a, 2015b).

L. nebulosus is a principal fish species, widely consumed in the southern part of the Gulf. It is a species of emperor fish also called as spangled emperor (Bailly, 2015a, 2015b; WoRMS, 2014) (Fig. 2). *L. nebulosus* has important commercial as well as nutritional value for the residents of the Gulf States. It is ranked as the top of 11 species contributing to Qatar fish landings with a percentage of 16.2% of total landings, according to the same reports released in 2014 (Al-Abdullrazzak et al., 2015; Stamatopoulos and Abdallah, 2015). Its lengths, weights, and growth rate are well documented, and they are exploited by local fishers (Grandcourt et al., 2004). Basic life-cycle history information for the *L. nebulosus*, an intermediate food web element, was studied as a function of body size parameters and expressed in the following equations for age (years), length (cm), and weight (g) of samples from the Qatar EEZ.

$$\text{(Age vs. Length)} \quad y = 0.0002x^3 - 0.0186x^2 + 0.6618x - 7.3344; \quad R^2 = 0.9969$$

$$\text{(Weight vs. Length)} \quad y = 0.0053x^3 + 0.4019x^2 - 1657x - 83.547; \quad R^2 = 0.9704$$

$$\text{(Age vs. Weight)} \quad y = 0.0044x + 0.2832 \quad R^2 = 0.9483$$

The highest regression coefficient (R^2) values were calculated as 0.9969 for Age vs. length correlation within the given equations. Mehanna et al. (2012) showed similar findings, where weight versus length plots yielded $R^2 = 0.9898$ (male) and 0.9901 (females) for total of 2731 individuals.

The rate at which an individual of any species accumulates MeHg (per year or any unit of time) should equal to the following = MeHg concentration (wet weight biomass) [mg kg^{-1}] $\times$ total mass (muscle only or muscle plus liver) [kg]/age of the animal [years] (Agah et al., 2007, 2009, 2010). This value provides the MeHg in the individual per unit of time over the life of the individual. This assumes that MeHg is concentrated uniformly (at the same rate) in young and old individuals (Fig. 3). Actually, this rate may increase as the individual increases in body size and consumes larger prey items which themselves can be expected to have higher Hg levels. Based on this relationship, the total prey consumption can be determined over the life of the organism, leading to a probable MeHg concentration estimate in the tertiary consumer in the food chain.

There is usually a predictable increase in MeHg concentration as a function of length (Hammerschmidt and Fitzgerald, 2006a, 2006b; Agah et al., 2007). Thus, as individuals grow, they not only experience an increase in MeHg concentration (Fig. 3) but also in total Hg loading (Fig. 4), where the total loading is equal to individual fish biomass times concentration. One of the largest fish from the samples collected in our wet/dry tissue

comparison study has a loading of $596 \text{ g} \times 0.618 \text{ mg kg}^{-1} \text{ w/w MeHg g}^{-1}$, where the fish weight is “wet weight” and MeHg concentration is ppm of “dry weight”. The dry weight is approximately equal to 20% of the wet weight, and thus a concentration of $596 \text{ g} \times 0.618 \times 0.2$, equals a total $73.7 \text{ mg kg}^{-1} \text{ w/w MeHg}$ for this individual fish. This has accumulated over the life of the fish or, in this case, 2.86 years, where the length was recorded as 29 cm. This equates to $25.76 \text{ mg kg}^{-1} \text{ w/w MeHg}$ per year or $0.07 \text{ mg kg}^{-1} \text{ w/w MeHg}$ per day. This loading of MeHg per fish can be expressed by the equation (Fig. 5):

$$\text{MeHg(ppm)} = -2.3 + 0.081(\text{length})$$

Thus, a fish sample of 28.9 cm fish weighs 342 g (wet weight) at an age of 1.77 years, or: $(0.061 \text{ mg kg}^{-1} \text{ w/w MeHg} \times 0.2 \text{ dry to wet weight conversion} \times 342 \text{ g wet weight per individual}) / (1.77 \text{ years} \times 365.25 \text{ d/year}) = 0.0065 \text{ mg kg}^{-1} \text{ w/w MeHg}$ accumulation per day among the smaller sized individuals. Thus, the rate of accumulation increases by an order of magnitude with age and size over the size range sampled (Table 7).

A wide spectrum of different marine taxa has assessed prior to, during, and after the ‘Gulf War’ of 1991 (Kureishy, 1993) to determine the war’s impact on sea food contamination. While little evidence indicate that the war had deleterious effects on marine life, the data are valuable because of the wide spectrum of organisms sampled and analyzed. Other studies (Abdul-Wahab et al., 2013; Sheppard et al., 2010) showed that the grouper (Hamoor) *Epinephelus coioides* (= *E. tauvina*) had Hg loading that followed the relationship:

$$[\text{Hg}] = 0.00316(\text{length}) + 0.0149,$$

where [Hg] is in ppm of wet weight.

This relationship differs somewhat from the findings of de Mora et al. (2004), where liver Hg concentrations were higher than for muscle tissue. However, liver tissue in fish amounts to only 1 to 3% of the total weight of the fish, meaning that liver loadings of Hg have been omitted in the calculation.

Based on the Hg loading equation given above for Hamoor grouper fish, similar calculations can be applied as those for the *L. nebulosus* where the largest individuals were assumed to live 7 years in the Gulf waters. The Hg uptake rate can be expressed by the following equation:

$$([\text{Hg concentration}] \times \text{wet to dry weight conversion} \times \text{fish biomass}) / \text{age in days}$$

Based on equation above; the uptake rate of Hg increases markedly as the *L. nebulosus* ages. This increase with age is common among many species, but the rates differ depending on the species. An emerging realization is that the amount of Hg in fish is directly related to the age and size of fish. Thus, while eating fish of an average size, and thus with an average Hg loading, may be safe, consuming larger fish specimens of a species could present a health hazard (Figs. 3 and 4).

The U.S. EPA (1997) derived a “Reference Dose” (RfD) as a tool to estimate daily intake levels of MeHg that are expected to be without an appreciable risk of deleterious health effects, even if exposure persists over a person’s lifetime. U.S. EPA (2001) developed an RfD for MeHg and recommends the use of the RfD to protect adults and children in the general population. The U.S. EPA (2001) also established a water quality criterion for methylmercury of 0.3 milligrams of methylmercury per kilogram of fish tissue on a wet weight basis (mg kg^{-1} w/w) based on the RfD. U.S. EPA (2001 RfD). U.S. EPA (2002) indicated that to protect consumers of fish and shellfish among the general population, the concentration of MeHg in tissue should not be exceeded based on an average consumption of 17.5 g of fish and shellfish per person per day (Lusk et al., 2005). The provincial tolerable intake of heavy metal cadmium was calculated as $1.2 \mu\text{g kg}^{-1}$ body weight/day by Al-Yousuf et al. (2000). To reach this limit the person will be required to consume 800 g of *L. lentjan* fish per day, which is far more than the average consumption of fish by a normal person per day.

It is possible to calculate the amount of Hg, an individual person will be exposed to, by eating the spangled emperor fish from the Qatar EEZ using the data generated from these studies. The United States Environmental Protection Agency (EPA’s 2001) Reference Dose (RfD) for methylHg is $0.1 \mu\text{g Hg per kg body weight per day}$ or $0.0001 \text{ mg Hg/kg/day}$ meaning that for an individual who weighs 70 kg, and consumes 227 g meal of fish per week (0.23 kg/week), assuming a mean MeHg concentration of *L. nebulosus* from Al-Khor site, which has the lowest MeHg values, i.e. 0.071 mg kg^{-1} w/w in muscle tissue (Table 5, min. of average values), the body burden for the numbers of meals that can be safely consumed can be calculated (Table 7). This hypothetical consumer could eat up to $7 \mu\text{g MeHg per day}$ to be at, or below, the U.S. EPA’s RfD which is equivalent to three *L. nebulosus* meals per week. FAO (2013 and 2014b) reports a fish supply of 24.5 kg per capita for the State of Qatar with a total production, including capture and aquaculture of 13 thousand tons in 2011. The apparent consumption is approximately 469.7 g per capita/week (Costenla, 2014). Larger individuals (weighing > 70 kg) could eat more fish; whereas as those with a lower body weight should eat proportionately less. It should be noted however that 0.32 mg kg^{-1} is a mean concentration, where larger fish and the fish from impacted sites would have higher MeHg values. It was observed that some of the larger size *L. nebulosus* has over 4–5 times of MeHg accumulation than smaller size of the same species (Fig. 3). This may conclude that eating small fish would be safer than eating larger ones (Table 7; Fig. 3a, b). Slightly better correlations were obtained as the MeHg data was compared with age of the same fish samples. (Fig. 3c). It can be concluded that MeHg bioaccumulates as fish mature in life span similar to other organisms.

Statistical analysis revealed a significant difference in MeHg between sites ($p < 0.05$, Fig. 3). Post-hoc analysis showed that Ras Laffan site located opposite to the industrial city was different from other sites (Ras Laffan – Al Khor $p < 0.01$, Ras Laffan – Al Ruwais $p < 0.01$, Ras Laffan – Halul $p = 0.02$, Ras Laffan – Mesaieed $p < 0.01$; Table 2), it had the highest mean MeHg of 0.77 ppm (Table 6). In addition, Al-Khor, which has a mean MeHg of 0.07 ppm had significantly lower levels of MeHg than Al Ruwais ($p < 0.01$) and Halul sites ($p < 0.01$). Analysis also revealed no significant difference in THg or MeHg between male and female individuals sampled ($p = 0.20$ and $p = 0.68$ respectively). Also, no significant

seasonal difference was detected in either THg or MeHg ($p = 0.06$ and $p = 0.80$ respectively). All tissues sampled displayed significantly different levels of THg ($p < 0.05$). Post-hoc analysis showed differences in all tissues with liver (mean: 0.60 ppm) containing the most Hg, followed by muscle (mean: 0.33 ppm) then gonad (mean: 0.07 ppm) in Fig. 6 (liver – gonad $p < 0.01$, liver – muscle $p < 0.01$, and gonad – muscle $p < 0.01$). Regression analysis also showed a significant positive relationship between fish age and THg ($p < 0.05$, $R^2 = 0.24$, Fig. 4). This is in good agreement with further analysis showing a significant relationship between fish length and THg ($p < 0.05$, $R^2 = 0.22$). Correlation of *L. nebulosus* fish biometric body size parameters with each other at 99% confidence levels were presented in Table 3. The dark shading shows the strongest correlation while lighter shading shows correlations still at acceptable levels.

While global estimates of Hg supply suggest important open ocean reservoirs of MeHg, only coastal processes and food webs are known sources of MeHg production, bioaccumulation, and biotransfer (Chen et al., 2008). In the Gulf, extensive oil and gas related activities are concentrated on the southeastern, northeastern, and northern coast of Qatar, together with desalination and energy production plants. High accumulations of MeHg in *L. nebulosus* specimens were detected at all sites except Al-Khor site, when MeHg concentrations are compared in *L. nebulosus*'s body size parameters (Fig. 3). It is obvious from Fig. 3a that as the weight of the fish increases, the amount of MeHg increases such that the toxicity level in the fish muscle tissue increases regardless of the fish location, length or age (Fig. 3b, c). As we compare MeHg and THg data in Figs. 3 and 4, data indicates that most of the Hg bioaccumulation in the *L. nebulosus* specimens analyzed is in the form of total-Hg deposition, compared to other more toxic-organic Hg species (mono-and di-methyl mercury). However, both figures direct us that Hg accumulates as body size parameters increase. Overall, consumers should avoid eating large adult fishes if smaller sized individuals are available (Table 7).

The highest accumulation rate for THg was observed at nearshore sites of Ras Laffan than Al-Khor and Halul Island sampling sites (Fig. 4) based on the sampling location for the *L. nebulosus*. This result for THg also supports elevated MeHg concentrations, observed in Fig. 3 for the same sites. This might be due to higher Hg(II) salts loading rates by industrial effluents, which are under the influence of coastal currents, formed by water masses from the north of the Peninsula. It is reported that higher Hg(II) loading rates may cause elevation of methylated form of Hg concentrations in zooplankton, benthic invertebrates, and consequently yield higher bioaccumulation rates in fish tissue at sites close to industrial cities (Orihel et al., 2007).

The amount of Hg in fish has been found to vary with different species, size, and age (Wiener et al., 2003). The distribution of THg concentrations versus body size parameters (age, length, weight) for the *L. nebulosus* specimens are shown for liver, muscle, and gonad tissues. The highest concentrations were found in liver tissue, while gonad tissues had the lowest concentration (Fig. 6a, b and, c) with almost no seasonal variations.

5. Conclusion

The present study concludes that Hg in fish tissues were within allowable concentrations and pose no threat to public health. *L. nebulosus* sampled from Qatar's marine EEZ showed no evidence of MeHg concentrations exceeding the U.S. EPA Reference Dose. However, consumers should be encouraged to limit themselves to no more than three meals of *L. nebulosus* (Sha'ri) per week. This may conclude that eating small fish would be safer than larger and older ones. It is recommended that, extensive monitoring for Hg should be extended to near-shore locations, close to industrial activities, as well as offshore locations around Qatar where a wider variety of fish seafood species, are present.

It is imperative that environmental contaminants, including marine Hg concentrations continue to be monitored to better understand their biogeochemical cycling and bioaccumulation. This is necessary to prevent health risks and to ensure nutritional safety conditions. Potential health effect of chronic low-level metal exposure may result from modest to several servings per week of fish consumption. Given that EEZ of Qatar is undergoing rapid industrial and urban development, a precautionary approach should be adopted not only in terms of conserving marine fish stocks in the marine EEZ, but also in relation to the potential bioaccumulation of contaminants, including Hg.

Acknowledgments

The authors would like to thank Qatar National Research Fund (QNRF) for funding and supporting this project under the National Priorities Research Program (NPRP) award number NPRP 09-505-1-081. The manuscript content is solely the responsibility of the authors and do not necessarily represent the official views of Qatar National Research Fund. We also would like to thank the Captain and crew of both Qatar University Research Vessels Mukhtabar Al-Bihar and Janan for their effort during sampling. The technical support of Mr. Hamood Al-Saadi, Mrs. Marwa Moustafa Al-Azhary and Ms. Maryam Hussien from Environmental Science Center (ESC), Qatar University during Hg samples preparations, pretreatment, and analysis is highly acknowledged. Thanks are extended to Dr. Jeffrey P. Obbard for reading this manuscript.

References

- Abdul-Wahab SA, Al-Husaini IS, Rahmalan A. Using grouper fish as bio-indicator of Cd, Cu, Pb and V in the vicinity of a single buoy mooring (SBM3) at Mina Al Fahal in the Sultanate of Oman. *Bull Environ Contam Toxicol*. 2013; 91:684. (2013). <http://dx.doi.org/10.1007/s00128-013-1126-0>. [PubMed: 24145924]
- Agah H, Leermakers M, Elskens M, Fatemi SMR, Baeyens W. Total mercury and methyl mercury concentrations in fish from the Persian Gulf and the Caspian Sea. *Water Air Soil Pollut*. 2007; 181:95. (2007). <http://dx.doi.org/10.1007/s11270-006-9281-0>.
- Agah H, Leermakers M, Elskens M, Fatemi SMR, Baeyens W. Accumulation of trace metals in the muscle and liver tissues of five fish species from Persian Gulf. *Environ Monit Assess*. 2009; 157:499. (2009). <http://dx.doi.org/10.1007/s10661-008-0551-8>. [PubMed: 18850288]
- Agah H, Egtesadi P, Owfi F, Fatemi S. Concentrations of some trace metals in the tissues of two commercial fishes from Tonekabon. *J Persian Gulf (Mar Sci)*. 2010; 1(2):55–64. December (2010) Vol.9.
- Al-Abdullrazzak D, Zeller D, Belhabib D, Tesfamichael D, Pauly D. Total marine fisheries catches in the Persian/Arabian Gulf from 1950 to 2010. *Reg Stud Mar Sci*. 2015; 2:28–34. 2015.
- Al-Naimi HA, Al-Ghouti MA, Al-Shaikh I, Al-Yafe M, Al-Meer S. Metal distribution in marine sediment along the Doha Bay, Qatar. *Environ Monit Assess*. 2015; 187(3):130. [PubMed: 25701472]

- Al-Yousf MH, El-Shahawi MS, AL-Ghais SM. Trace metals in liver, skin and muscle of *Lethrinus lentjan* fish species in relation to body length and sex. *Science of the Total Environment*. 2000; 256:87–94. [http://dx.doi.org/10.1016/S0048-9697\(99\)00363-0](http://dx.doi.org/10.1016/S0048-9697(99)00363-0). [PubMed: 10902836]
- Andrews AH, Kalish JM, Newman SJ, Johnston JM. Bomb radiocarbon dating of three important reef-fish species using Indo-Pacific D14C chronologies. *Mar Freshw Res*. 2011; 62:1259–1269. 2011.
- Assadi, H., Dehghani, PR. Atlas of the Persian Gulf and the Sea of Oman fishes. Iranian Fisheries Research and Training Organization; Iran: 1997.
- Bailly, N. Froese, R., Pauly, D., editors. *Lethrinus nebulosus* (Forsskal, 1775). 2015a. FishBase. 2015a. Accessed through: World Register of Marine Species at <http://www.marinespecies.org/aphia.php?p=taxdetails&id=212081> on 2015-12-29
- Bailly, N. Froese, R., Pauly, D., editors. *Lethrinus lentjan* (Lacepède, 1802). (2015b). FishBase. 2015b. Accessed through: World Register of Marine Species at <http://www.marinespecies.org/aphia.php?p=taxdetails&id=212061> on 2016-10-01
- Booth S, Zeller D. Mercury, food webs, and marine mammals: implications of diet and climate change for human health. *Environ Health Perspect*. 2005; 113
- Bouhlef, M. Poissons de Djibouti. RDA International, Inc.; Placerville (California, USA): 1988. p. 416
- Bu-Olayan AH, Subrahmanyam MNV, Al-Sarawi M, Thomas BV. Effects of the Gulf War oil spill in relation to trace metals in water, particulate matter, and PAHs from the Kuwait coast. *Environ Int*. 1998; 24:789–797.
- Burger J, Gochfeld M. Locational differences in heavy metals and metalloids in Pacific Blue Mussels *Mytilus [edulis] trossulus* from Adak Island in the Aleutian Chain, Alaska. *Sci Total Environ*. 2006; 368:937–950. [PubMed: 16828845]
- Carpenter, KE., Krupp, F., Jones, DA., Zajonz, U. Living Marine Resources of Kuwait, Eastern Saudi Arabia, Bahrain, Qatar, and the United Arab Emirates. FAO; Rome: 1997. FAO species identification field guide for fishery purposes; p. 293
- Carpenter, KE., Allen, GR. “Emperor Fishes and Large-eye Brems of the World (Family Lethrinidae)”, An annotated and Illustrated Catalogue of Lethrinid Species Known to Date. Vol. 9. FAO; Rome: 1989. FAO species catalogue; p. 118 FAO Fish. Synop. 125(9) <http://www.fishbase.org/summary/1846>
- Chen C, Amirbahman A, Fisher N, Harding G, Lamborg C, Nacci D, Taylor D. MethylHg in marine ecosystems: spatial patterns and processes of production, bioaccumulation, and biomagnification. *EcoHealth*. 2008; 5(4):399–408. (2008). <http://dx.doi.org/10.1007/s10393-008-0201-1>. [PubMed: 19015919]
- Cheng Z, Hong-Sheng W, Duc J, Sthiannopkao S, Xing GH, Kim KW, Salleh M, Yasin M, Hisham Hashim J, Wong MH. Dietary exposure and risk assessment of mercury via total diet study in Cambodia. *Chemosphere*. 2013; 92(1):143–149. 2013, June 2013. [PubMed: 23499220]
- Costenla, M. Heavy Metal Contamination in Seafood and Consumer Exposure in the Gulf Cooperation Council Faculdade de Ciências e Tecnologia da Universidade Nova de Lisboa. 2014. July 2014, MSc. Thesis paper
- Cullen JT, Field MP, Sherrell RM. Determination of trace elements in filtered suspended marine particulate material by sector field HR-ICP-MS. *J Anal At Spectrom*. 2001; 16:1307–1312. 2001.
- de Mora S, Fowler S, Wyse E, Azemad S. Distribution of heavy metals in marine bivalves, fish and coastal sediments in the Gulf and Gulf of Oman. *Mar Pollut Bull*. 2004; 49:410–424. 2004. [PubMed: 15325209]
- Fitzgerald W, Clarkson T. Mercury and monomethyl mercury: present and future concerns. *Environ Health Perspect*. 1991; 96:159–166. 1991. [PubMed: 1820259]
- Fitzgerald WF, Lamborg CH, Hammerschmidt CR. Marine biogeochemical cycling of mercury. *Chem Rev*. 2007; 107(2):641–662. (2007). <http://dx.doi.org/10.1021/cr050353m>. [PubMed: 17300143]
- Forskål P. Descriptiones animalium avium, amphibiorum, piscium, insectorum, vermium; quæ in itinere orientali observavit Petrus Forskål. Post mortem auctoris edidit Carsten Niebuhr. Adjuncta est materia medica Kahirina atque tabula maris rubri geographica. 1775:1–20. I–XXXIV [=1–34], 1–164, 1 map. Hauniæ. (Möller).

- Fowler SW, Readman JW, Oregioni B, Villeneuve JP, McKay K. Petroleum hydrocarbons and trace metals in near shore Gulf sediments and biota before and after the 1991 war: an assessment of temporal and spatial trends. *Mar Pollut Bull.* 1993; 27:171–182.
- Grandcourt EM, Al Abdessalaam TZ, Francis F, Al Shamsi T. Biology and stock assessment of the Sparids, *Acanthopagrus bifasciatus* and *Argyrops spinifer* (Forsskal, 1775), in the Southern Arabian Gulf. *Fish Res.* 2004; 69(1):7–20. 2004.
- Haghighat M, Savabieasfahani M, Nikpour Y, Pashazanoosi H. “Hg in the Oriental Sole (*Brachirus orientalis*) near a Chlor-Alkali plant in the Persian Gulf”, Iran. *Bull Environ Contam Toxicol.* 2011; 86:515–520. 2011. [PubMed: 21442211]
- Hammerschmidt CR, Fitzgerald WF. Methylmercury in mosquitoes related to atmospheric mercury deposition and contamination. *Environ Sci Technol.* 2005; 39:3034–3039. 2005. [PubMed: 15926549]
- Hammerschmidt CR, Fitzgerald WF. Bioaccumulation and trophic transfer of methylmercury in Long Island Sound. *Arch Environ Contam Toxicol.* 2006a; 51(3):416–424. <http://dx.doi.org/10.1007/s00244-005-0265-7>. [PubMed: 16823518]
- Hammerschmidt C, Fitzgerald WF. Methylmercury in freshwater fish linked to atmospheric mercury deposition. *Sci Technol.* 2006b; 40(24):7764–7770. (2006). <http://dx.doi.org/10.1021/es061480i>.
- Hammerschmidt, CR. PhD dissertation. 2005. The Biogeochemical Cycling of Methylmercury in Coastal Marine Sediments. Abstract (2 leaves) bound with copy Thesis (Ph D.)- University of Connecticut 2005 Includes bibliographical references. Adviser: William F. Fitzgerald
- IARC. Monographs on the Evaluation of Carcinogenic Risks to Humans Volume 58. Lyon, France: 1993. p. 444
- Kardovani, P. Iranian Marine Ecosystem (The Persian Gulf and the Caspian Sea). Ghomes; Tehran Iran: 1995.
- Koeman JH, Peeters WHM, Koudstaal-Hol CHM, Tjioe PS, De Goeij JJM. Hg-selenium correlations in marine mammals. *Nature.* 1973; 245:385–386. 1973. [PubMed: 4593498]
- Kureishy TW. Heavy metals in algae around the coast of Qatar. *Mar Pollut Bull.* 1991; 22:414–416. 1991.
- Kureishy TW. Concentration of heavy metals in marine organisms around Qatar before and after the Gulf War oil spill. *Mar Pollut Bull.* 1993; 27:183–186. 1993.
- Kureishy TW, Ahmed MH. Total mercury distribution in surface sediments from the Arabian Gulf. *Qatar Univ Sci J.* 1994; 14:390–394. 1994.
- Kureishy TW, Al-Muftah AR. Total mercury levels in the coastal environment of Qatar (Arabian Gulf). *Qatar Univ Sci J.* 1999; 19:274–284. 1994.
- Lamborg CH, Fitzgerald WF, Damman AWH, Benoit JM, Balcom PH, Engstrom DR. Modern and historic atmospheric mercury fluxes in both hemispheres: global and regional mercury cycling implications. *Glob Biogeochem Cycles.* 2002; 16 (ISSN: 0886-6236). <http://dx.doi.org/10.1029/2001GB001847>.
- Lamborg CH, Tseng CM, Fitzgerald WF, Balcom PH, Hammerschmidt CR. Determination of the mercury complexation characteristics of dissolved organic matter in natural waters with “reducible Hg” titrations. *Environ Sci Technol.* 2003; 37(15):3316–3322. (2003) Aug 1. [PubMed: 12966976]
- Lubick N, Malakoff D. With pact’s completion, the real work begins. *Science.* 2013; 341:1443–1445. (2013). http://scim.ag/pod_6153. [PubMed: 24072901]
- Lusk, JD., Rich, E., Bristol, RS. Methylmercury and Other Environmental Contaminants in Water and Fish Collected From Four Recreational Fishing Lakes on the Navajo Nation, 2004. Prepared for the Navajo Nation Environmental Protection Agency, Completion report for intergovernmental agreement; 2005. <http://ifw2es.fws.gov/NewMexico/>. Accessed 17 October 2005
- Madany IM, Wahab AAA, Al-Alawi Z. Trace metals concentrations in marine organisms from the coastal areas of Bahrain, Arabian Gulf. *Water Air Soil Pollut.* 1996; 91(1996):233–248.
- Massachusetts Department of Environmental Protection (MassDEP), Office of Research and Standards. Final Report: Massachusetts Fish Tissue Mercury Studies: Investigations of Seasonal and Other Sources of Variation. Boston, MA 02043 USA: 2006.

- Mehanna, S., Zaki, S., Al-Kiuyumi, F., Al-Kharusi, L., Al-Bimani, S. Biology and fisheries management of spangled emperor; Jounieh – Lebanon. 06–08 November-2012; 2012. International Conference on Land-Sea Interactions in the Coastal ZoneINOC-CNRS
- National Research Council (U.S.) – Board on Environmental Studies and Toxicology. National Academies Press. 2000. Toxicological Effects of Methylmercury. 978-0-309-07140-6
- Nithyanandan M, Jones DA, Esseen M. Fishery resources of Sabah Al-Ahmad Sea City waterways: a potential contributor for Kuwait's fisheries. *Aquat Ecosyst Health Manag*. 2016; 19(4):452–460.
- Orihel DM, Paterson MJ, Blanchfield PJ, (Drew) Bodaly RA, Hintelmann H. Experimental evidence of a linear relationship between inorganic mercury loading and methylmercury accumulation by aquatic biota. *Environ Sci Technol*. 2007; 41(14):4952–4958. (2007). <http://dx.doi.org/10.1021/es063061r>. [PubMed: 17711208]
- Subba Rao DV, Al-Yamani F. The Arabian Gulf Seas at the Millennium. 2000:2.
- Sadiq, M., McCain, JC., editors. The Gulf War Aftermath: An Environmental Tragedy Environment and Assessment. Kluwer; 1993. p. 320
- Sadiq M. Metal contamination in sediments from a desalination plant effluent outfall area original research article. *Sci Total Environ*. 2002; 287(1–2):37–44. 15 March 2002. [PubMed: 11883759]
- Sheppard C, Al-Husiani M, Al-Jamali F, Al-Yamani F, Baldwin R, Bishop J, et al. The Gulf: A young sea in decline. *Mar Pollut Bull*. 2010; 60(1):13–38. (January 2010). <http://dx.doi.org/10.1016/j.marpolbul.2009.10.017>. [PubMed: 20005533]
- Stamatopoulos C, Abdallah M. Standardization of fishing effort in Qatar fisheries: methodology and case studies. *J Mar Sci Res Dev*. 2015; 5:170. (2015). <http://dx.doi.org/10.4172/2155-9910.1000170>.
- Stephan, CE., Mount, DI., Hansen, DJ., Gentile, JH., Chapman, GA., Brungs, WA. Guidelines for Deriving Numerical National Water Criteria for the Protection of Aquatic Organisms and their Uses. National Technical Information Service; Springfield, VA: 1985. p. 98
- Taylor DL, Kutil NJ, Malek AJ, Collie JS. Mercury bioaccumulation in cartilaginous fishes from Southern New England coastal waters: contamination from a trophic ecology and human health perspective. *Mar Environ Res*. 2014; 99:20–33. (2014 Aug). <http://dx.doi.org/10.1016/j.marenvres.2014.05.009>. [PubMed: 25081850]
- Tseng CM, Hammerschmidt CR, Fitzgerald WF. Determination of methyl-mercury in environmental matrices by on-line flow injection and atomic fluorescence spectrometry. *Anal Chem*. 2004; 76:7131–7136. [PubMed: 15571370]
- U.S. Environmental Protection Agency. Method 2001: Methods for the Determination of Metals in Environmental Samples. 1991 EPA-600/4-91-010 June 1991.
- U.S. Environmental Protection Agency. Method 1669: Sampling Ambient Water for Trace Metals at EPA Water Quality Criteria Levels. 1996
- U.S. Environmental Protection Agency. A Review of the Reference Dose and Reference Concentration Processes. Prepared for the Risk Assessment Forum U.S. Environmental Protection Agency; Washington, DC: 1997. EPA/630/P-02/002F December 2002 Final Report
- U.S. Environmental Protection Agency. Appendix to Method 1631: Total Mercury in Tissue, Sludge, Sediment, and Soil by Acid Digestion and BrCl Oxidation. 2001a US EPA-821-R-01-013 January 2001.
- U.S. Environmental Protection Agency. Method 1630: Environmental Protection Agency, Methyl Mercury in Water by Distillation, Aqueous Ethylation, Purge and Trap, and CVAFS. 2001b EPA 821-R-01-020.
- U.S. Environmental Protection Agency. US Environmental Protection Agency Mercury Update: Impact on Fish Advisories. EPA Office of Water; Washington, DC: 2002. EPA-823-F-01-011
- U.S. Geological Survey. Mercury in the Environment. Fact Sheet 146-00. 2000 Oct. 2000 URL. <http://www.usgs.gov/themes/factsheet/146-00/index.html>
- Wagemann RTE, Boila G, Lockhart WL. Methyl-mercury and total mercury in tissues of Arctic marine mammals. *Science*. 1998; 125:45–49.
- Wiener, JG., Krabbenhoft, DP., Heinz, GH., Scheuhammer, AM. Ecotoxicology of mercury. In: Hoffman, DJ.Rattner, BA.Burton, GA., Jr, Cairns, J., Jr, editors. Handbook of Ecotoxicology. second. CRC Press; Boca Raton, Florida: 2003. p. 409-463.Chapter 16

- Withrow, S., Vail, D. Withrow and MacEwan's Small Animal Clinical Oncology. Elsevier 0721605583; 2007. p. 73-74.
- WHO/UNEP DTIE Chemicals Branch. Guidance for Identifying Populations at Risk From Mercury Exposure. UNEP DTIE Chemicals Branch and WHO Department of Food Safety, Zoonoses and Foodborne Diseases; Geneva, Switzerland: 2008. p. 167 Publication details August, 2008 English (<http://www.chem.unep.ch/>). (www.who.int/foodsafety/publications/chem/mercuryexposure.pdf, page 57)
- WoRMS. World Register of Marine Species - *Lethrinus nebulosus* (Forsskål, 1775). 2014 Marinespecies.org. Retrieved 2014-05-26.
- Zhu A, Zhang W, Xu Z, Huang L, Wang W. Methylmercury in fish from the South China Sea geographical distribution and biomagnification. *Mar Pollut Bull.* 2013; 77(2013):437–444. [PubMed: 24095204]

Fig. 1.
Locations of sampling sites and coordinates for *L. nebulosus* in the EEZ of Qatar.

Fig. 2.
Sampled fish species *L. nebulosus* (Sha'ri) from Qatar EEZ. (Credit: Illustration by Dr. Ibrahim A. Al-Maslamani, QU-ESC).

Fig. 3.

The *L. nebulosus* MeHg data (muscle) vs selected biometric body size parameters - (a) weight, (b) length, (c) age - from six sampling sites. R-squared (R^2) values were given next to linear trend lines to show coefficient of determination for each sampling set. The thicker, solid black line shows the cumulative data trend line for all sites, except Al Khor, which was not included to this trend line, due to unpaired bioaccumulation behavior.

Fig. 4.

The bioaccumulation of THg vs biometric body size parameters (weight, length, age) at three selected sampling sites for *L. nebulosus*. R-squared (R^2) values were given next to linear trend lines to show coefficient of determination for each sampling set. The thicker, dashed red line shows the cumulative data trend line for all sampling sites. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

Fig. 5. Methylmercury (MeHg in dry weight) concentration as a function of size (length in cm) in *L. nebulosus* collected off Masaied industrial area (coastal waters of Qatar).

Fig. 6. The bioaccumulation of Hg vs biometric body size parameters (weight, length, age) in 3 different tissues of *L. nebulosus*. The thicker, dashed lines show the cumulative data trend line for all each type of tissue.

Table 1

Quality assurance (QA) of the fish tissue samples analyzed in the study. (LBS: Laboratory Blank Spiked, N: Number of samples analyzed).

QA samples	N (#)	Measured value (µg/g)	Reference value (µg/g)	Recovery (%)
MeHg SRM (NIST-2977)	4	0.0344	0.0366 ± 0.001	94.0
0.010 µg MeHg LBS	4	0.009	0.010	88.9
0.050 µg MeHg LBS	1	0.052	0.050	104.9
0.100 µg MeHg LBS	4	0.942	0.100	94.2
0.500 µg MeHg LBS	1	0.520	0.500	103.9
THg SRM (DORM-4)	3	0.402	0.412 ± 0.036	97.6
1.600 µg THg LBS	3	1.619	1.600	101.2
C78164 + 0.3 µg Hg	1	0.607	0.307 (C78164)	97.7
C78187 + 0.3 µg Hg	1	0.408	0.116 (C78187)	98.0

Table 2

The tukey post hoc analysis results were given for 6 sampling locations, using MeHg data (on the top), together with Sha'ri organ tissues, using THg data (on the bottom).

	Al Khor	Al Ruwais	Doha	Halul	Mesaieed	Ras Laffan
Al Khor		0.01	0.46	0.00	0.10	0.00
Al Ruwais	0.01		0.76	0.98	0.58	0.01
Doha	0.46	0.76		0.38	0.99	0.00
Halul	0.00	0.98	0.38		0.14	0.02
Mesaieed	0.10	0.58	0.99	0.14		0.00
Ras Laffan	0.00	0.01	0.00	0.02	0.00	

	Gonad	Liver	Muscle
Gonad		0.0001	0.0001
Liver	0.0001		0.0079
Muscle	0.0001	0.0079	

Correlation of *L. nebulosus* fish biometric body size parameters with each other at 99% confidence levels. The color code (highlighted) dark gray shows the strongest correlation while light gray shows correlations still at acceptable levels.

Table 3

<i>Correlation</i>	Total weight (g)	Total length (cm)	Age (yr)	Flesh weight (g)	Gonad weight (g)	Liver weight (g)
Total weight (g)	1.00					
Total length (cm)	0.96	1.00				
Age (yr)	0.95	0.98	1.00			
Flesh weight (g)	0.74	0.75	0.58	1.00		
Gonad weight (g)	0.30	0.32	0.23	0.32	1.00	
Liver weight (g)	0.80	0.78	0.79	0.69	0.26	1.00

Table 4

Dry and wet weight mean concentrations for total Hg were given as mg kg^{-1} w/w (ppm) in selected *L. nebulosus* muscle, gonad, and liver tissue samples from the Qatar EEZ. N = Number of Sample.

Type (organ)	THg (dry weight)	THg (wet weight)	N
Muscle	0.015 ± 0.004	0.025 ± 0.023	25
Gonad	0.016 ± 0.004	0.076 ± 0.03	11
Liver	0.018 ± 0.002	0.600 ± 0.180	9

Wet weight mean concentrations for THg were given as $\text{mg kg}^{-1} \text{w/w}$ (ppm) in muscle, gonad, and liver tissue samples for male and female *L. nebulosus* from the 4-selected sampling location. N = number of sample analyzed.

Table 5

Location ↓ Gender →	Muscle			Gonad			Liver		
	Male	Female	Male + Female	Male	Female	Male + Female	Male	Female	Male + Female
Al-Ruwais	0.494 N = 1	0.323 N = 1	0.409 ± 0.121 N = 2	0.079 N = 1	0.083 N = 1	0.081 ± 0.003 N = 2	0.686 N = 1	0.855 N = 1	0.771 ± 0.120 N = 2
Ras Laffan	0.349 ± 0.048 N = 5	0.508 ± 0.004 N = 2	0.394 ± 0.087 N = 7	0.056 ± 0.028 N = 5	0.088 ± 0.040 N = 2	0.065 ± 0.032 N = 7	0.457 ± 0.037 N = 5	0.797 ± 1.194 N = 2	0.554 ± 0.186 N = 7
Al-Khor	0.508 ± 0.123 N = 3	0.476 ± 0.104 N = 3	0.492 ± 0.104 N = 6	0.124 N = 1	0.102 N = 1	0.113 ± 0.016 N = 2	NA	NA	NA
Hahul Island	0.181 ± 0.071 N = 5	0.294 ± 0.097 N = 5	0.237 ± 0.100 N = 10	NA	NA	NA	NA	NA	NA

Wet weight mean concentrations for Methyl-Hg ($\pm$ standard deviations) were given in the table as mg kg⁻¹ w/w. (ppm) from all six-sampling location for male, female and cumulative *L. nebulosus* muscle samples (at the center). N = number of sample analyzed (depending sample catch at each station). The table also reflects the results of the *L. nebulosus* biometric body size parameters and its standard deviation for the cumulative number of catch at each site (on the right).

Table 6

Site #↓	Site name ↓	MeHg (ppm)		Weight (g)		Length (cm)		Age (yr)	
		Male	Female	Male + female	Male + female	Male + female	Male + female	Male + female	Male + female
1	Al-Ruwais	0.450 ± 0.211 N = 4	0.272 ± 0.206 N = 2	0.390 ± 0.209 N = 6	762 ± 299 N = 6	38.3 ± 4.7 N = 6	3.02 ± 0.35 N = 3		
2	Ras Laffan	0.626 ± 0.337 N = 2	0.871 ± 0.013 N = 3	0.773 ± 0.216 N = 5	498 ± 139 N = 5	32.1 ± 3.4 N = 5	2.29 ± 0.57 N = 5		
3	Al-Khor	0.064 ± 0.044 N = 9	0.138 ± N.A N = 1	0.071 ± 0.048 N = 10	604 ± 218 N = 10	36.6 ± 4.4 N = 10	3.30 ± 0.97 N = 10		
4	Doha	0.386 ± 0.187 N = 2	0.110 ± 0.075 N = 2	0.248 ± 0.197 N = 4	747 ± 250 N = 4	38.2 ± 4.8 N = 4	3.14 ± 0.88 N = 3		
5	Mesaieed	0.164 ± 0.051 N = 2	0.295 ± 0.178 N = 9	0.257 ± 0.159 N = 13	444 ± 99 N = 13	31.6 ± 2.8 N = 13	2.21 ± 0.48 N = 13		
6	Halul Island	0.463 ± 0.047 N = 2	0.445 ± 0.228 N = 5	0.450 ± 0.187 N = 7	334 ± 77 N = 7	29.4 ± 2.5 N = 7	1.82 ± 0.39 N = 7		

Table 7

Consumption of *L. nebulosus* in terms of meals of 8 oz. each and amounts of Hg uptake per meal. Calculations are based on assuming that *L. nebulosus* has a minimum MeHg concentration of 0.071 mg kg^{-1} at Al-Khor station and maximum of MeHg concentration of $0.320 \pm 0.257 \text{ mg kg}^{-1}$ (average of maximum concentration recorded at each sampling site).

Meals per week	Consumption rate in oz./week	Consumption rate in kg/week	Hg exposure mg/week (max. MeHg)	Hg exposure mg-day (max. MeHg)	Hg exposure mg/week (max. MeHg)	Hg exposure mg-day week (max. MeHg)
1	8	0.2268	0.0161	0.0023	0.0726	0.0104
2	16	0.4536	0.0322	0.0046	0.1451	0.0207
3	24	0.6804	0.0483	0.0069	0.2177	0.0311
4	32	0.9072	0.0644	0.0092	0.2903	0.0415
10	80	2.2680	0.1610	0.0230	0.7257	0.1037