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

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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.

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 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

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Location	Site #	Stn. #	Latitude	Longitude
Al-Ruwais	1	3	N 26°15.717'	E 051°16.985'
Ras Laffan	2	4	N 25°49.629'	E 051°42.444'
Al-Khor	3	5	N 25°36.953'	E 051°42.932'
Doha	4	6	N 25°19.479'	E 051°42.461'
Mesaieed	5	7	N 24°51.642'	E 051°32.153'
Halul Island	6	11	N 25°40.083'	E 052°25.266'

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

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 demersal 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