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Mercury Contamination in Pelagic Fishes of the Gulf of Mexico

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Abstract

Methyl mercury (MeHg) bio accumulates from small benthic invertebrates to large pelagic fish; therefore high end consumers and terminal predators have elevated mercury concentrations. In this study total mercury concentrations were measured in 10 Gulf of Mexico pelagic fish species using a DMA 80 analyzer. Total mercury concentration ranged from 0.004 to 3.55 ppm (wet wt). The highest mean concentration (1.04 ppm wet wt) was recorded for king mackerel (*Scomberomorus cavalla*), exceeding US Food and Drug Administration (FDA) recommended criteria of 1ppm. Dolphinfish (*Coryphaena hippurus*) and vermilion snapper (*Rhomboplites aurorubens)* had the lowest mean Hg concentrations (<0.3 ppm). The rest of the species were above the US Environmental Protection Agency (EPA) advisory level of 0.3 ppm. Wahoo (*Acanthocybium solandri)*, greater amberjack (*Seriola dumerili*) and gag grouper (*Mysteroperca microlepsis*) had high mercury concentrations of approximately 0.7 ppm wet wt. Blackfin tuna (*Thunnus atlanticus*) and yellowfin tuna (*Thunnus albacores)* had moderate mercury concentrations (0.39 and 0.36 ppm wet wt). Little tunny (*Euthynnus alletteratus*) and blacktip shark (*Carcharhinus limbatus*) had mean concentrations of 0.69 and 0.51 ppm respectively. The relationship between fish length and mercury concentrations was significant for four species. Mercury is a neurotoxin and poses a great risk to humans. It is especially dangerous to pregnant women and developing children. Knowledge of mercury concentrations in fish is essential to ensure protection of the environment and human health. This project is supported by the Qatar National Research Fund (QNRF) through the Environmental Studies Center of Qatar University, Doha.

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Introduction

Mercury is a toxic, naturally occurring element that is hazardous to humans. Anthropogenic activities contribute to natural atmospheric mercury levels mainly by fossil fuel burning, gold mining, paint and wood pulp industry, and cement production (Pacyna et al. 2006). It is estimated that mercury concentration has increased three-fold since pre-industrial levels (Fain et al. 2009). Sedimentary and atmospheric measurements estimate anthropogenic input exceeds natural sources by a factor of 2.4 – 4 (Slemr & Langer 1992). In nature, mercury gas is introduced to the atmosphere by degassing of the earth's crust, emissions from the ocean's surface or created by volcanic activity (Pirrone et al. 2010). Mercury vapor is relatively inactive, but it is of high concern due to its volatility. In the atmosphere mercury can be transported great distances from point sources making it a global pollutant that is returned to the earth's surface by dry or wet deposition (Schroeder & Munthe 1998). Two ionic states, mercury (I) and mercury (II), can form salts that readily dissolve in water. Once in solution mercury can be methylated by iron or sulfur reducing bacteria forming its most toxic form: methyl mercury (MeHg). Microbial mercury methylation is enhanced under acidic, anoxic conditions and elevated temperatures (Merritt & Amirbahman 2009, Kelly et al. 2003). MeHg bioaccumulates in aquatic food chains from algae, small benthic invertebrates to large pelagic fish and it reaches highest concentrations in the top trophic levels (Wang 2002). MeHg makes up about >95% of total mercury in fish muscle (Bloom 1998). Humans and other terminal predators are exposed through fish consumption (Pentreath 1976, Hall et al. 1997). Although nutritious, some large pelagic fish species are not recommended for pregnant women because the fetus is very sensitive to mercury toxicity (Kris-Etherton et al. 2002). Methyl mercury is a neurotoxin and has been linked to cardiovascular problems in humans (Koren & Bend 2010). Fish contaminated with mercury pose a great risk to consumers (Selin 2009).

The main objective of this study was to measure mercury contamination in targeted Gulf of Mexico fish. In addition we determined the relationship between mercury levels and fish size and attempt to explain Hg variations in different fish species based on feeding and life history traits.

Sample collection and preservation

Fish samples were obtained with hook and line at docks and offshore in three Gulf of Mexico regions. The majority were collected in Venice, LA (48) and Freeport, TX (38), with the remaining collected in Port Aransas, TX (28) (Figure 1). All the sampling was carried out from February to November in 2002. For each fish about 20g of muscle tissue was removed from the dorsal region behind the head, individually bagged, labeled and stored on ice for transportation to the laboratory where they were stored permanently at -20°C until analyzed. Ten species were chosen (Table 1) because they were suspected to concentrate Hg, their life histories (age, growth rate and maximum sizes) are

well established in the literature, and they were available among the archived specimens at TAMUG.

Mercury analysis

Fish white muscle tissues were analyzed for total mercury content using the direct mercury analyzer DMA-80, Milestone inc. It employs drying and combustion steps, followed by gold sequestration and mercury quantification using atomic absorption spectrometry (Haynes et al. 2006). Samples were taken from the freezer, cut into 0.5-1 g pieces and placed into 20 ml glass scintillation vials (purchased from Kimble). Vials were pre cleaned by soaking overnight in 10% hydrochloric acid (HCl), with subsequent combustion for 6 hours in the drying oven at 60°C. Vials with frozen fish tissues were placed into a vacuum freeze drier for 72 hours to ensure complete dehydration. Dry samples were homogenized and pulverized using a glass mortar and pestle. All utensils were cleaned with methanol to avoid cross contamination between samples. Pulverized samples were individually weighed and placed into a nickel coated sample boat for analysis in the DMA-80. Drying time and temperature were chosen based on published literature (Cizdziel et al. 2002). The dried samples were then combusted to reduce all mercury species to the elemental form. Gold amalgamator was used for mercury sequestration. Subsequent amalgamator heating releases trapped mercury. Continuous stream of oxygen caries Hg gas through two cuvette cells and Hg was quantified using atomic absorption spectrometry. Mercury concentrations in the sample were calculated based on most recent calibration curve. Daily calibration was performed following the EPA method 7473. Standard reference material (SRM) (dogfish liver) DOLT-4 [2.58+/-0.22 ppm], purchased from the National Research Council of Canada were used to generate quadratic calibration curve. Three replicates of the first three fish were analyzed after each calibration. Once the relevant percent difference was within 10%, then samples were analyzed in singles or repeated. Blanks (empty boats) were analyzed every eight samples to ensure that mercury was not carried over between samples. A separate SRM (fish protein) DORM-3 [Hg: 0.382+/-0.06 ppm] was analyzed every eighth sample to assure accuracy. Best fit calibration curves were obtained using a quadratic function with an R^2= 1.00 (ranged from 0.99- 1.00). Recovery of the SRM DORM-3 ranged from 99% to 115%, mean = 107%. The sample precisions based on the coefficients of variation of the three replicates were 0.01-9.6 %, with a mean of 3.2%. Total mercury was used as a proxy for organic methyl mercury (MeHg) as it is well-established that > 95% of total mercury in edible muscle is MeHg (Bloom 1992). Recovery was calculated based on the mean published certified DORM -3 value and all recovered mercury in fish standard was within certified mercury range of 0.382- 0.44 ppm.

Statistical analysis

Linear regression analysis was used to determine the relationship between the mercury concentration and fish length (Figure 3-6). The regression model was fitted such as that $\log_{10}$ ppm = β_0 j +β_1 j * cm, for j = 1, 2, . . . ,10 . using separate slopes ($\beta_{1)}$ and intercepts (β_0) for each species. We chose to use $\log_{10}$ [Hg] ppm instead of [Hg] ppm as the dependent variable to get a better agreement with regression assumptions (normality of residuals, homos cedasticity). Assumptions of normality were examined using Shapiro Wilk test. To account for multiple comparisons the slopes in the above model were judged significant if a 99% confidence interval did not contain 0.

Results

Mercury concentrations of 10 commonly consumed pelagic Gulf of Mexico fish species were determined (Table 1) and compared to national action levels and guidelines (Figure 2). The highest mercury concentration in dry weight was recorded for a specimen of wahoo: 12.11 ppm, and the lowest in yellowfin tuna: 0.035 ppm. To ease comparisons, all Hg concentrations were converted from dry weight to wet weight using the conversion equation y= 3.80x + 0.04 , where y= mean [Hg] ppm dry weight and x= mean [Hg] ppm wet weigh (Cai et al. 2007). Mean mercury levels per species ranged between 0.05 and 1.04 ppm wet weight. The only two species that had concentrations below the 2002 US Environmental Protection Agency (EPA) reference dose of 0.3 ppm wet wt. were vermillion snapper and dolphinfish 0.05 ppm and 0.21 ppm wet wt, respectively (EPA 2002). The rest of the examined species had higher mercury concentrations than a recommended advisory level set by the US EPA (Table 2): wahoo, greater amberjack and gag grouper had a mean [Hg] 0.7 ppm wet wt. Blackfin tuna mean [Hg] 0.39 ppm wet wt; yellowfin tuna mean [Hg] 0.36 wet wt and little tunny mean [Hg] 0.69 ppm wet wt. King mackerel had the highest mean mercury concentration of 1.04 ppm wet wt., exceeding both the EPA limit and the higher action level of 1.0ppm wet wt. set by the Food and Drug Administration (FDA 2001).

Four species in this study showed a significant positive relationship between mercury concentration and fish length. The regression model was significant ($F_{19,86}$ = 37.31, $p < 10^{-16}$) and the regression assumptions were met. No point was unduly influential-all Cook's distances were below 0.5. For the regression slopes for each species 99% confidence intervals were calculated. For the slopes that were significantly above 0, the regression model for the mercury level in that species was calculated. The model for blackfin tuna showed the highest increase in mercury levels - 6.41% for each additional cm in length. The rate of mercury accumulation was calculated to be lowest for yellowfin tuna, with a 2.87% increase of mercury concentration per cm of length. Models for wahoo and dolphinfish showed a 4.51% and 4.3 % increase in Hg concentration for each cm increase of length, respectively (Figures 3-6).

Discussion

Variation among species was correlated with fish length, and large aquatic organisms tend to accumulate Hg through their diet (Downs et al. 1998, Wang 2002). The non-linear increase as fish grow larger may be because larger fish consume larger prey that contain higher concentrations of mercury.

King mackerel, with the highest mean Hg concentration (1.04 ppm wet wt) exceeding both the EPA and the FDA limits, deserve special attention. Like other predatory fish, king mackerel feeds mainly on other fish: sardines, mullet, drums, jacks, even as a juvenile it has the diverse diet of a pelagic carnivore (Finucance et al., 1990). However, it was not the largest fish in the study. Higher mercury levels in king mackerel can be related to its long lifespan (~ 20 years), yielding longer exposure times for older fish. Thus, high concentrations may be due to age and food source, rather than size alone.

In contrast, vermillion snapper, with the lowest concentrations (0.05 ppm wet wt), was also the smallest from all fish examined, growing only to about 60 cm in length (Grimes 1978). In some instances it has been observed that closely related sympatric snapper species may have very different levels because of different prey consumption. For example (Bank et al., 2007) documented an increased of mean Hg concentration in grey snapper (0.15 ppm) compared to that in red snapper (0.06 ppm). They related this to a slightly higher trophic level in combination with a preference for more pelagic bony fish rather than benthic prey, which is not what might have been expected if the source is the sea floor.

Dolphinfish had low mercury concentrations (0.21 ppm wet wt) and this species has a relatively short lifespan (3 - 4 years) (Schwenke & Buckel 2008). Even though it is an oceanic fish and generally piscivorous, it has lower metabolic demand than other migratory species as it is distributed in warmer seas, show high site fidelity, and tend to have lower mercury concentrations (Oxenford & Hunte 1999). Again, this points to age rather than size as a critical factor in accumulation.

Wahoo (0.73 ppm wet wt) can attain exceptional burst speeds that allow it to catch mackerel, butterfish, herring, scads and jacks; in addition wahoo grow rapidly, especially its first year, but can live up to ten years. However, the reported age averages to 1.8 years (McBride et al. 2008). Feeding habits and rapid growth along with high metabolic requirement for pelagic lifestyle may result in high observed Hg concentrations in muscle.

Positive linear relationships were found between total length and logarithmic Hg concentration in four fish species: yellowfin and blackfin tuna, wahoo and dolphinfish (Figures 3-6). A larger sample size might have resulted in a significant relationship in other species as well, as previously observed (Cai et al. 2007, Adams 2004). The high rate of Hg accumulation in blackfin tuna compared to yellowfin tuna (6.41% and 2.87% per cm) may be due to their different habitats. Blackfin tuna and yellowfin tuna consume a variety of fish, crustaceans and mollusks (Manooch et al. 1985). Both tuna species are highly migratory (swimming thousands of miles), but blackfin tuna are neritic and

forage closer to the shoreline, generally over continental shelves. In contrast, yellowfin tuna are oceanic, swim continuously in the top 100 meters of the water column and feed opportunistically on available prey (Adams et al. 2003). Small fish are dominant prey for blackfin tuna, whereas for yellowfin tuna in addition to fish, cephalopods are also a very important food source. Mercury content in squid is typically less than 0.1 ppm, which is considerably lower than for fish consumed by tuna (Falandysz 1990). Blackfin tuna predominantly feed on prey having a higher Hg concentration and therefore their exposure is increased.

Elemental mercury is converted to highly toxic methyl-mercury (MeHg) in the sediment water interface (Ullrich et al. 2001). MeHg is readily assimilated first by microbes, benthic biota and then passed to higher predators. Fish examined here feed mainly on other fish rather than benthic organisms. Despite the direct link from benthic source of MeHg, these fish accumulate high mercury levels through complex and as yet poorly identified steps.

Mercury concentrations of the pelagic fish measured were compared to national action levels and guidelines (Figure 2). Humans (70 kg body weight) should not consume more than one meal per month of fish that have Hg concentrations of 0.5 ppm wet weight. A meal is described as 226.8 g of uncooked edible fish tissue (EPA 2002). This advisory would apply for all species examined in this study except vermillion snapper and dolphinfish.

Location, environmental parameters such as pH, temperature and concentrations of organic matter all have been linked to mercury accumulation in fish (Merritt & Amirbahman 2009). Significant regional differences in mercury concentration were observed for king mackerel in Atlantic (0.94 ppm) and Gulf locations (1.51 ppm) (Adams & McMichael 2007).

In general, the results and insights gained from this work confirm that species that are higher in the food chain, such as king mackerel, accumulated more Hg compared to vermillion snapper, a smaller species at a lower trophic level. The findings of this study were in agreement with previous work studying mercury accumulation in large fish species (Mason et al. 2000, Baeyens et al. 2003, Bank et al. 2007, Hogan et al. 2007). This study was also consistent with previous investigations that reported positive relationships between fish size and muscle tissue mercury concentration (Sonesten 2003, Trudel & Rasmussen 2006, Cai et al. 2007).

Figure 1. Map of sampling locations at the docks and offshore from Freeport (Texas), Port Aransas (Texas) and Venice (Louisiana) in the NW of Gulf of Mexico.

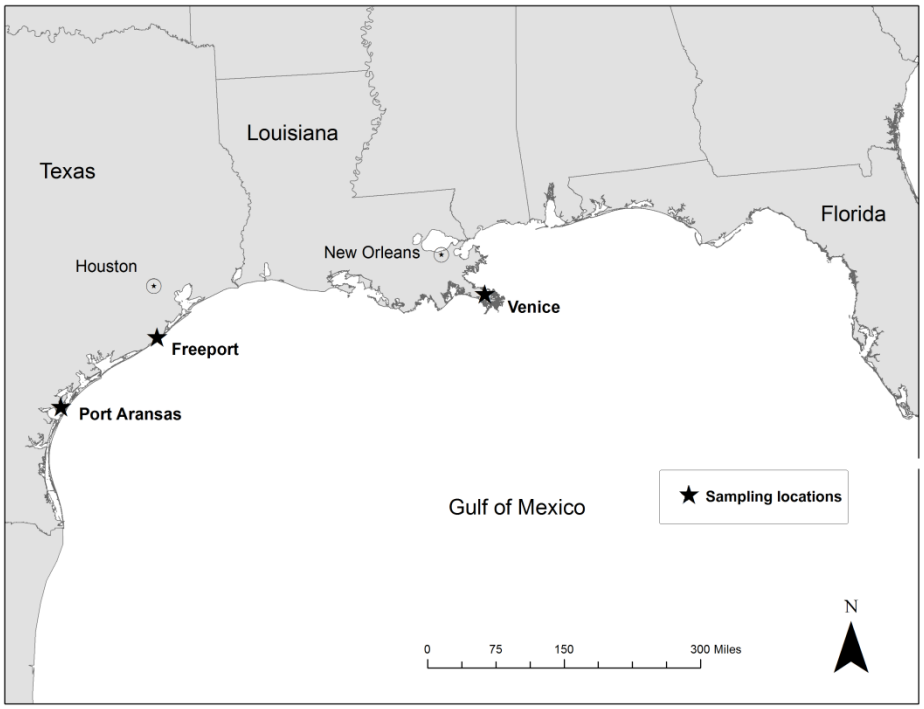

Figure 2. Mean mercury concentrations in ppm wet weight of ten pelagic fish of Gulf of Mexico (±1 standard error) compared to EPA advisory limit 0.3ppm (dashed line) and FDA action level 1.00ppm (solid line),

AJ: Greater amberjack, BS: blacktip shark, BX: blackfin tuna, DF: dolphinfish, GG: gag grouper, KM: king mackerel, LT: little tunny, VS: vermillion snapper, WA: wahoo, YT: yellowfin tuna.

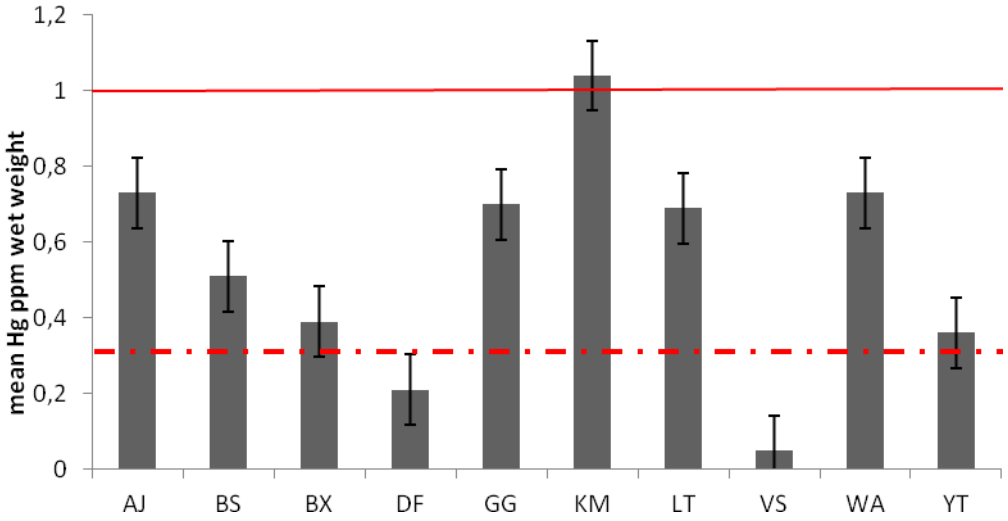

Figure 3. Mean mercury concentration log [Hg] ppm for blackfin tuna (*Thunnus atlanticus*) versus total length of fish (cm)

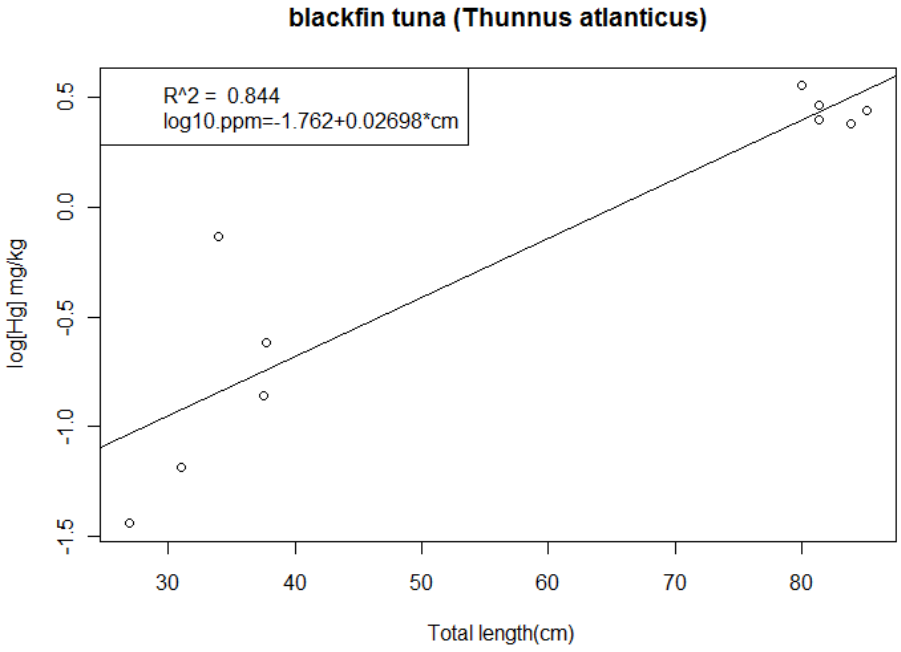

Figure 4. Mean mercury concentration log [Hg] ppm for yellowfin tuna (*Thunnus albacares*) versus total length of fish (cm)

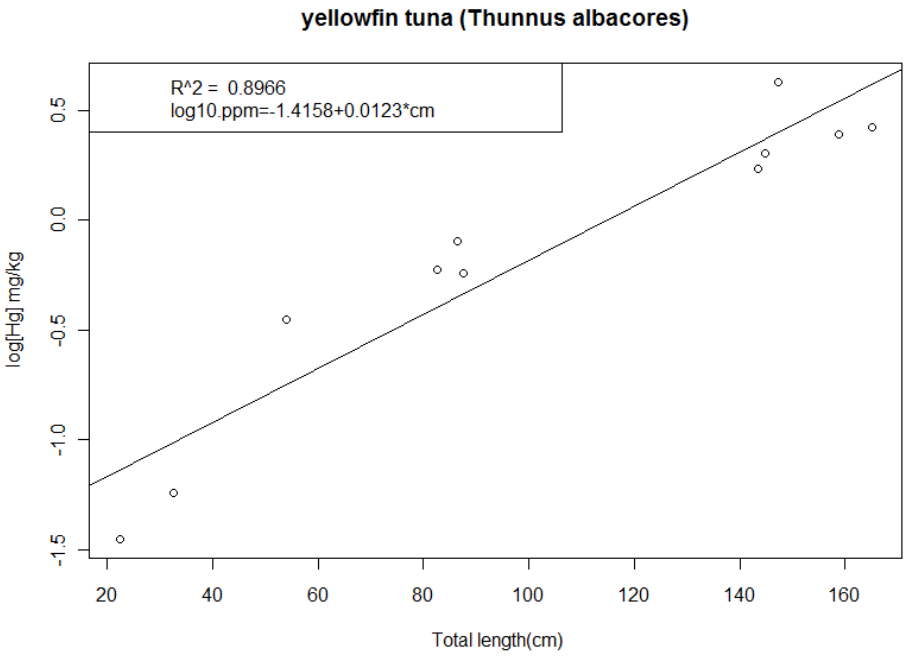

Figure 5. Mean mercury concentration log [Hg] ppm for Wahoo (*Acanthocybium solandri)* versus total length of fish (cm)

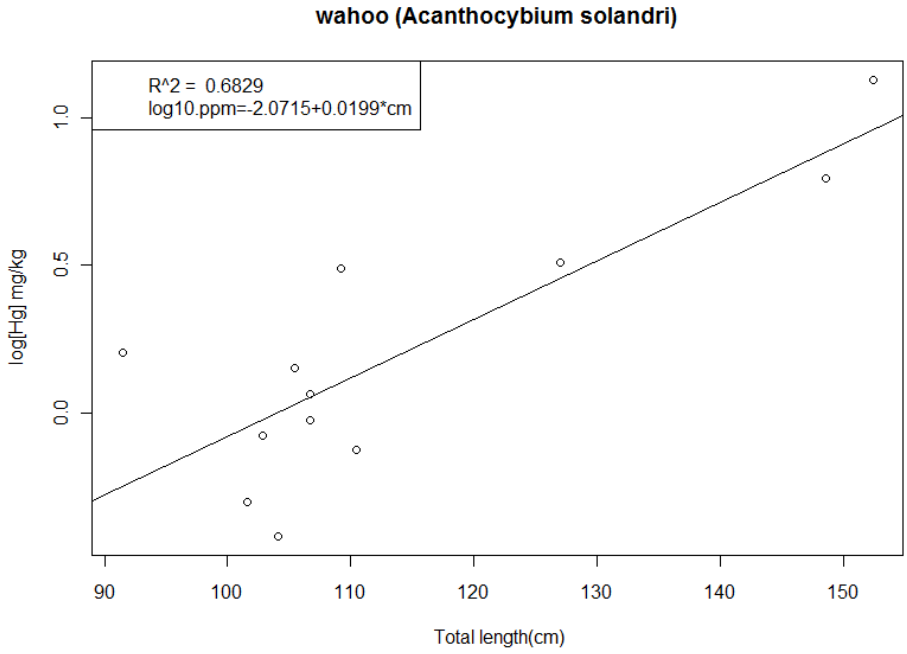

Figure 6. Mean mercury concentration log [Hg] ppm for dolphinfish (*Coryphaena hippurus*) versus total length of fish (cm)

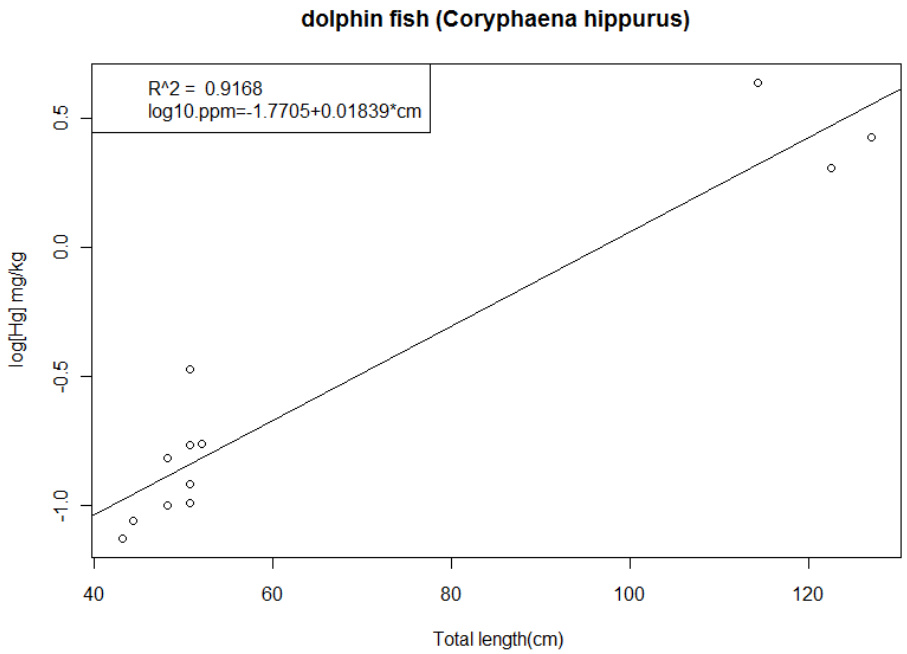

Table 1. Species selected for this study, total mercury levels in [ppm] dry weight and transformed to wet [Hg] concentrations ppm wet wt. *Above U.S. EPA 2002 recommended criteria level 0.3 ppm wet wt. **Above FDA 2001 recommended criteria level 1.0 ppm wet wt. Conversion to wet weight y = 3.80x + 0.04 was based on (Cai et al. 2007)

Common name	Scientific name	N	size range cm	range [Hg] dry wt	Mean [Hg] wet wt
*blackfin tuna (BX)	*Thunnus atlanticus*	11	27-80	0.036- 3.58	***0.39***
*yellowfin tuna (YT)	*Thunnus albacares*	11	22-147	0.035- 4.26	***0.36***
*little tunny (LT)	*Euthynnus alletteratus*	9	53-60	1.57- 3.66	***0.69***
*wahoo (WA)	*Acanthocybium solandri*	12	91-152	0.38- 13.52	***0.73***
**king mackerel (KM)	*Scomberomorus cavalla*	12	70-98	2.04- 5.92	***1.04***
*Greater amberjack (AJ)	*Seriola dumerili*	12	73-119	2.04 – 5.92	***0.73***
*gag grouper(GG)	*Mycteroperca microlepis*	10	74-109	1.34- 5.23	***0.70***
vermilion Snapper(VS)	*Rhomboplites aurorubens*	11	25-48	0.1- 0.47	***0.05***
dolphinfish (DF)	*Coryphaena hippurus*	12	43-123	0.07- 4.33	***0.21***
*blacktip shark (BS)	*Carcharhinus limbatus*	6	56-173	0.48- 5.94	***0.51***

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