INTRA- AND INTERSPECIES VARIABILITY IN MERCURY CONCENTRATIONS IN TEXAS MARINE FISH AND SHELLFISH

by

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ABSTRACT

Mercury (Hg) is a toxic pervasive global pollutant that can bioaccumulate in marine organisms and biomagnify in marine food webs. The Gulf of Mexico has a higher concentration of Hg than the connecting Atlantic Ocean. Humans are mainly exposed to Hg through seafood consumption and the per capita rate of fish consumption is higher in Gulf states, potentially exposing the seafood consuming public to elevated concentrations of Hg. This study investigated the concentration of Hg in 26 species of fish and 4 species of shellfish (*n* = 1,468 individuals) caught along the Texas coast during 2016 and 2017 using a Direct Mercury Analyzer, and investigated the relationship between Hg concentration and body size for 26 of these species. A significant positive relationship was found between Hg concentration and body size in 7 nearshore fish species, 10 offshore fish species, and 1 shellfish species, indicating that Hg was bioaccumulating over time in these species. In comparison, there was a negative relationship between Hg concentration and body length in stripped mullet, possibly as a result of growth dilution. No relationship was observed in 7 of the species investigated which could be a result of low sample size and/or narrow range in body length. Spatial differences in Hg concentration were investigated in 4 residential species; no difference in Hg concentration was observed among sites for southern flounder, but there was a site difference for red drum, spotted seatrout, and black drum. Overall, when combined together, offshore fish had the highest wet wt Hg concentration (1.61 µg/g) followed by nearshore fish (0.175 µg/g) and shellfish (0.035 µg/g). For individual species, the average wet wt Hg concentration was highest in blue marlin (11.6 µg/g) and sailfish (1.21 µg/g) and lowest in brown shrimp (0.0086 µg/g) and American oyster (0.006 µg/g). Three offshore fish species (blue marlin, sailfish, and king mackerel) had an average Hg concentration that exceeded the FDA action limit of 1 µg/g wet wt and 7 offshore species exceeded the Texas State Department of Health Services (TSDHS) health-based standard of 0.7 µg/g wet wt. In comparison, gafftopsail catfish was the only nearshore fish species that had an average Hg concentration that exceeded the EPA human health criterion of 0.3 µg/g wet wt, and no shellfish species exceeded the EPA criterion. While shellfish are safe to eat due to their very low Hg concentration, offshore fish species and some nearshore fish species should still be consumed in moderation due to their high Hg concentration.

I. INTRODUCTION

1.1 Mercury as a global pollutant

Mercury (Hg) is a nonessential trace element that is toxic to marine organisms at low concentration (UNEP 2013; Ordiano-Flores et al. 2011). It is a pervasive global pollutant that can be found within the earth, in the atmosphere, terrestrial systems, and aquatic systems (Selin 2009). Elemental Hg (Hg^0) and inorganic Hg(II) (Hg^{2+}) are released into the environment through natural processes like volcanic eruptions and soil erosion (Pirrone et al. 2010; Schuster et al. 2002), as well as anthropogenic sources, including emissions from coal fired power plants and gold mining activities (UNEP 2013; Wang et al. 2004). In the atmosphere, Hg^0 is photo-oxidized into Hg^{2+} where it can travel up to a year before it is deposited onto terrestrial or aquatic surfaces via wet and dry deposition (Driscoll et al. 2013; Hall et al. 2007; Morel et al. 1998). After atmospheric fallout over aquatic environments, Hg can be converted into methylmercury (MeHg; CH_3Hg^{2+}), its most toxic organic form, primarily by sulfate reducing bacteria (De Simone et al. 2014; King et al. 2001; Morel et al. 1998) in sulfur-rich anoxic sediments as well as in the water column (Shao et al. 2012). Once methylated, Hg is more bioavailable to organisms and enters the food web via the phytoplankton and is trophically transferred to top predators (Morel et al. 1998).

1.2 Mercury bioaccumulation and biomagnification in marine organisms

Mercury is well known to bioaccumulate and biomagnify in marine organisms. Within a species of fish, Hg normally increases in concentration over time, so larger, older individuals tend to have a greater muscle Hg concentration compared to smaller, younger individuals (Vega-Sánchez et al. 2017; Teffer et al. 2014; Choy et al. 2009; Chen et al. 2008; Kojadinovic et al. 2006; Baeyens et al. 2003; Gilmour and Riedel, 2000). This is because > 84% of Hg found in muscle tissue is present as MeHg (Cabañero et al. 2007; Hammerschmidt and Fitzgerald, 2006; Duffy et al. 1999), and therefore total Hg (THg; referred to as Hg throughout this study) can be used as an indicator of MeHg concentration in fish muscle tissue. Laboratory studies have shown that ~99% of MeHg enters fish through their diet (Dutton and Fisher 2014, 2010; Wang and Wong, 2003), and due to the high assimilation efficiency (AE; ≥ 90%) and low efflux rate (0.8%/day) of MeHg from the body (Dutton and Fisher, 2014, 2010), it increases in concentration over time.

Mercury, in particular MeHg, biomagnifies up marine food webs (Lavoie et al. 2013; Bank et al. 2007; Cai et al. 2007; Hammerschmidt and Fitzgerald, 2006; Atwell et al. 1998), so that Hg body burdens increase with trophic level, resulting in top predators, such as tuna, swordfish, sharks, and marine mammals having the highest Hg body burden (De Simone et al. 2014; Seixas et al. 2014; Karimi et al. 2013; Storelli et al. 2002; Storelli and Marcitrigiano, 2001; Monteiro et al. 1996; Dietz et al. 1996). Mercury biomagnifies up marine food webs because the percentage of Hg that is present as MeHg increases with every increase in trophic step. For example, Hammerschmidt and Fitzgerald (2006) found that within the Long Island Sound, the percentage of total Hg present as MeHg was 3% in oxic water, 9% in microseston, and 84% in alewife (*Alosa pseudoharengus*). Laboratory studies have shown that the trophic transfer factor (TTF), defined here as the likelihood a contaminant will biomagnify at a particular trophic step, is much greater than 1 (7.6 – 13) for MeHg when forage fish consume crustaceans and oligochaetes containing MeHg; in comparison Hg(II) is unlikely to biomagnify because the TTF is less than 1 (0.26 – 0.38) (Dutton and Fisher 2011, 2010). An increase in MeHg concentration occurs with an increase in trophic step due to MeHg's high assimilation efficiency and low efflux rate from the body in fish, therefore the high TTF results in higher trophic predators having the highest MeHg body burden.

1.3 Negative health effects of Hg in fish

Exposure to Hg, in particular MeHg, is well known to have deleterious health effects in fish at all life stages. In embryos, exposure to MeHg can result in pericardial edema, spinal curvature and craniofacial defects (Dong et al. 2016; Samson and Shenker 2000; Weis et al. 1981). Gill deformities are common in larvae and in adults that are exposed to MeHg concentrations greater than 0.3 ppm, resulting in a disruption in gas exchange (Macirella and Brunelli, 2017; Morcillo et al. 2016). In adults, Hg(II) and MeHg exposure can cause lower testosterone production in males and estrogen in females resulting in altered mating behavior and lowered oocyte production (Guchhait et al. 2018; Sandheinrich et al. 2006). Maternal transfer studies have shown that Hg can lead to increased embryo mortality and reduced hatching success of larvae (Sackett et al. 2013; Devlin and Mottet, 1992).

1.4 Mercury and human health

The main source of Hg exposure in humans is through seafood consumption, in particular predatory fish found at the top of the food web (Li et al. 2014, 2013; Pastorelli et al. 2012; Mergler et al. 2007**)**. Mercury poisoning through consumption of contaminated seafood was first confirmed in 1956 when locals in Minamata Bay, Japan experienced tingling in their hands and feet, reduced motor control, trembling, speech impairment, and mental retardation (Fujiki and Tajima, 1992).The resulting illness, Minimata disease, was caused by the discharge of industrial chemicals that contained Hg by the Chisso Corporation into Minamata Bay (Ekino et al. 2007; Funabashi, 2006) resulting in health effects that were transgenerational (Yorifuji et al. 2011).

Mercury is a well-known neurotoxin which can cross the blood brain barrier targeting the central nervous system (Rice et al. 2014), resulting in the effects displayed with Minamata disease. Exposure to Hg can also result in cardiovascular issues such vasoconstriction and increased risk of hypertension, heart attack and stroke, and immunological health effects (Rice et al. 2014; Solenkova et al. 2014; Roman et al. 2011). Mercury exposure while *in utero* and in first year of life is of particular concern because Hg crosses the placenta and is transferred by breast milk from mother to child; studies have shown that infants have reduced brain function and lower IQ scores, and muscle growth and coordination as a result of Hg exposure (Freire et al. 2010; Oken et al. 2008, 2005; Oken and Bellinger, 2008).

Due to these deleterious health effects in humans, federal and state agencies now issue seafood consumption advisories for fish and shellfish species that contain an elevated concentration of Hg. The Food and Drug Administration (FDA) which oversees commercial fisheries will issue an advisory when the Hg concentration in muscle tissue exceeds the 1 µg/g wet weight action limit, and the Environmental Protection Agency (EPA), which oversees recreational fisheries nationwide recommends an advisory is issued when the Hg concentration exceeds the 0.3 µg/g wet weight human health criterion. However, every individual state health agency in the U.S. determines the Hg concentration at which they will issue an advisory for recreationally caught fish; in Texas, the Texas Department of State Health Services (TDSHS) issues a Hg advisory when the Hg concentration exceeds the 0.7 µg/g wet weight health-based standard.

Although people are advised to limit their consumption of certain fish species to reduce their Hg exposure, it is also advised that they include fish as part of a healthy diet since it is low in saturated fat and high in protein, omega-3 fatty acids, and selenium (Se) which is required for cardiovascular and cognitive function (Molfino et al. 2014; Burger and Gochfeld, 2009; Daviglus et al. 2002; Patterson 2002). Due to these conflicting messages, the species of fish to consume and the frequency at which to eat it is confusing for the seafood consuming public.

1.5 Mercury in the Gulf of Mexico

The Gulf of Mexico has a higher concentration of Hg compared to waters off the east coast and west coast of the United States (Evans et al. 2015; NSTC, 2004), and this higher Hg concentration has been reported in fish as well. For example, a study by the FDA (2014) found that golden tilefish (*Lopholatilus chamaeleonticeps*) in the Gulf of Mexico had a higher average Hg concentration (1.123 µg/g wet wt) compared to individuals from the Atlantic Ocean (0.144 µg/g wet wt). King mackerel and Spanish mackerel in the Gulf of Mexico have also been found to have higher Hg concentrations (0.45 µg/g and 0.73 µg/g, respectively) in comparison to Atlantic populations (0.05 µg/g and 0.18 µg/g, respectively) (Levenson and Axelrad, 2006). Harris et al. (2012a, b) calculated that 88% of Hg that enters the Gulf of Mexico is from the Loop Current, 10%

is from atmospheric deposition, and 2% is from major rivers (i.e., Mississippi and Atchafalaya Rivers). Mercury can be remobilized within sediment along the Gulf of Mexico coastline from natural disturbances like hurricanes, which can result in a short-term increase in the rate of Hg methylation within the local environment (Liu et al. 2009). Sediment surrounding drilling rigs in the Gulf of Mexico have been found to have higher rates of Hg methylation, possibly influenced by an increase in organic matter from higher densities of fish (Delaune et al. 2008; Trefry et al. 2007) which could increase the amount of Hg that is bioavailable for uptake.

In the U.S., fisheries in the Gulf of Mexico account for 16% of the national commercial catch and 41% of the national recreational catch (Harris et al. 2012a, 2012b; NOAA, 2011; Buck et al. 2015). Most commercial and recreational fisheries have a minimum harvest size in which larger individuals are targeted to allow for younger individuals to reach lengths at which they are sexually mature; therefore a higher concentration of Hg is expected in the muscle tissue and can increase the risk of toxicity in humans from consumption of these larger individuals. All 5 U.S. states bordering the Gulf of Mexico (TX, LA, MS, AL, FL) have issued Hg advisories regarding seafood consumption (Texas Department of State Health Services, 2013; Florida Department of Health, 2017; Alabama Department of Public Health, 2017; Mississippi Department of Environmental Quality, 1998; Louisiana Department of Health & Hospitals, Department of Environmental Quality, Department of Wildlife & Fisheries, 2006). States have the ability to set the concentration at which to issue a Hg advisory instead of adopting the EPA human health criterion which could lead to a lack of advisories being issued for many bodies of water (Adams et al. 2016). These differences between issued Hg advisories for federal and state agencies could potentially be confusing for recreational fisherman and possibly lead to increased Hg exposure in humans that consume larger fish species not listed under state regulations. The Hg concentration in swordfish, shark, tilefish, and king mackerel can exceed the FDA 1 μg/g action limit (Harris et al. 2012; Levenson and Axelrad, 2006) therefore, knowing the length at which Hg concentrations start to exceed these limits can be beneficial to commercial and recreational fisheries. In addition, the average per capita fish consumption is higher along the Gulf coast and that increases risk of Hg exposure (Harris et al. 2012b) because recreational fishing for marine species is a popular past time, as well as a relatively easy source of protein for low income and/or minority groups (Burger and Gochfeld, 2011b; Williams and Choo, 2003).

1.6 Mercury in Texas seafood

Due to elevated Hg concentrations in the Gulf of Mexico, Hg exposure through the consumption of contaminated fish and shellfish is a public health concern. An Hg advisory is issued by TSDHS when the Hg concentration in muscle tissue exceeds 0.7 μg/g wet weight. The current advisories state that blue marlin (*Makaira nigricans*) should not be consumed, as well as king mackerel (*Scomberomorus cavalla*) that are larger than 88.9 cm total length. It is also suggested that women under 50 and children under 12 should avoid consumption of blackfin tuna (*Thunnus atlanticus*), little tunny (*Euthynnus alletteratus*), crevalle jack (*Caranx hippos*), sharks, swordfish, and king mackerel (Texas Department of State Health Services, 2013).

The TDSHS issued Hg advisories in 2013 for certain marine fish species along the Texas coast that contain Hg concentrations above the human health-based standard of 0.7 µg/g wet wt. In total, 288 individual samples were taken from 17 species of fishes caught along the Texas Coast in 2011 by the TDSHS. In addition, only 12 of the species of fishes had the relationship between body length and Hg concentration examined in the report (TDSHS, 2012). There are issues with the current report as the number of samples (e.g. n = 8 blackfin tuna, n = 3 blue marlin, and n = 2 mangrove snapper samples) for each species investigated was relatively low, as well as the lack in representation for smaller nearshore species commonly caught along the Texas coastline. An update in Hg concentration for fishes caught along the Texas coast is warranted as the current TDSHS Hg advisory is 8 years old with a low representation of fishes that are caught and consumed along the Texas coast.

Previous studies investigating the relationship between Hg concentration and body length found a positive relationship in large pelagic species such as cobia (*Rachycentron canadum*), blue marlin, blackfin tuna, king mackerel, little tunny, dolphinfish (*Coryphaena hippurus*), yellowfin tuna (*Thunnus albacares*), wahoo (*Acanthocybium solandri*), and red snapper (*Lutjanus campechanus*), and all these species had a Hg concentration ranging above and below the TDSHS health-based standard (Zapp Sluis et al. 2013; Kuklyte 2012; Cai et al. 2007).

In addition, residential species such as red drum (*Sciaenops ocellatus*), southern flounder (*Paralichthys lethostigma*), spotted seatrout (*Cynoscion nebulosus*), black drum (*Pogonias cromis*), gafftopsail catfish (*Bagre marinus*), and spanish mackerel (*Scomberomorus maculatus*) (Nims and Walther, 2014; Anderson and Karel, 2012, 2009;

Neahr et al. 2010; Rooker et al. 2010; Mendoza-Carranza and Hernandez-Franyutti, 2005; Gold and Turner, 2002; Gold et al. 2001,1993; Finucane and Collins, 1986) could have spatial differences in Hg concentrations along the Texas coast due to local differences in environmental Hg concentrations; however; with the exception of Lavaca Bay (a remediated Hg Superfund site which is now under long term biomonitoring), Hg concentrations have remained consistently low in these species throughout the Texas coastline (Stunz and Robillard, 2011; Sager 2004).

After searching through the scientific literature, it was found that no studies have investigated either the Hg concentration and/or Hg relationship between Hg concentration and body length in crevalle jack, great barracuda (*Sphyraena barracuda*), white marlin, and many nearshore fish species like spanish mackerel, pinfish (*Lagodon rhomboides*), mangrove snapper (*Lutjanus griseus*), striped mullet (*Mugil cephalus*) Atlantic croaker (*Micropogonias* undulatus), blue crab (*Callinectes* sapidus), brief squid (*Lolliguncula* brevis), and brown shrimp (*Penaeus aztecus*) caught along the Texas coast. Determining Hg concentrations and the relationship between Hg concentration and body length can give further understanding to the size range of fish and shellfish species that is still safe to consume from recreational and commercial fisheries.

1.7 Objectives of the study

This study examined the concentration of total Hg in 26 fish and 4 shellfish species harvested along the Texas coast. The objectives can be broken down as follows:

1. Investigate the relationship between Hg concentration and body length or weight for each species, with the prediction that Hg concentration will increase with body size
2. Determine if there are any site differences in Hg concentration for red drum, black drum, speckled seatrout, and southern flounder, with the prediction that there will be differences among sites
3. Investigate interspecies differences in Hg concentration between nearshore fish, offshore fish, and shellfish with the prediction that offshore fish species will have the highest Hg concentrations
4. Determine which species have Hg concentrations that exceed federal and state Hg advisory levels.

II. METHODS

2.1 Sample collection

Tissue samples from 26 fish species and 4 shellfish species were collected for this study (Table 1). Muscle tissue samples were collected from 24 fish species at 10 fishing tournaments held in 6 locations along the Texas Coast (Texas City, Freeport, Port O'Connor, Port Aransas, Port Mansfield, and Port Isabel; Figure 1) during July and August 2016 and March, July, and August 2017. For 22 of these species, approximately 5 g of axial muscle tissue was collected with the skin attached from the left side of the body beneath the front of the dorsal fin using a stainless steel fillet knife, stored individually in trace metal clean tubes, and immediately placed on ice; fish length (fork or total) and weight data was also recorded at the time of muscle sample collection. For this study, cubera snapper (*Lutjanus cyanopterus*), remora (*Remora remora*), and rainbow runner (*Elagatis bipinnulata*) were included as nearshore species because they were caught close to shore where they can be found for part of their life cycle (Claro et al. 2009; Fertyl and Landry, 2009; Yesaki, 1979) and weighed-in during the bay fishing part of the competition.

Four fish species were collected whole, stored on ice in Ziplock bags, and later subsampled in the lab. Mangrove snapper and pinfish were obtained at the Port Mansfield Fishing Tournament, whereas Atlantic croaker and striped mullet were purchased from bait shops in Port Mansfield and Port Isabel, respectively. The four investigated shellfish species were also purchased whole, stored on ice in Ziplock bags and later processed in the lab. American oysters (*Crassotrea virginica*) were purchased in Port Isabel (however, they were harvested in Rockport), blue crab and brief squid were purchased from bait shops in Texas City, and brown shrimp were purchased from a shrimp boat in Port Aransas. Upon return to the lab, all samples were stored at -20 °C until further processing.

2.2 Sample processing

Muscle samples collected at fishing tournaments were thawed, the skin and any bone removed, and the tissue subsampled making sure that no surface tissue that was exposed during sampling at the tournament was included to reduce the risk of contamination from external sources. Samples were then blot dried to remove excess water, the wet weight recorded, and then dried at 60°C for 48 hours, after which the dry weight was recorded and the tissue homogenized into a fine powder using a pestle and Ziplock bags. For the four fish species that were collected whole, the fish were thawed, the total length and whole weight were recorded, and axial muscle was sampled from both sides of the fish to obtain enough mass. Samples were then dried and homogenized as previously described.

The shellfishes were thawed and processed as followed: American oysters were cleaned of any debris on the shell, after which all soft tissue was removed and blot dried to remove excess water. Brown shrimp were weighed whole before removing the exoskeleton, head and tail, so only the soft tissue was analyzed. The mantle length of the brief squid was recorded, and the mantle separated from the rest of the body for Hg analysis. The blue crab carapace width was recorded and the leg/claw tissue removed and dried. All samples were dried and homogenized following the method previously described.

For comparison with other studies and advisory guidelines, the water content for each species (to allow for conversion between dry and wet weight Hg concentrations) is shown in Table 1.

2.3 Mercury analysis

Samples were analyzed for total Hg using a Direct Mercury Analyzer (DMA-80; Milestone Inc., Shelton, CT) through thermal combustion, gold amalgamation, and atomic absorption spectroscopy following EPA Method 7473 (U.S. EPA, 2007). Based on the expected Hg concentration, between 5 and 50 mg of sample was analyzed; for example, species with a known high concentration of Hg, e.g., blue marlin and king mackerel had 5-15 mg of muscle tissue analyzed, whereas lower Hg concentration organisms, e.g., red drum, brief squid, and American oyster had 35-50 mg of tissue analyzed. The DMA-80 was calibrated as needed using three certified reference materials (CRMs) from the National Research Council Canada (NRCC): MESS-4, marine sediment (0.08 μg/g); TORT-3, lobster hepatopancreas (0.292 μg/g); and PACS-3, marine sediment (2.98 μg/g).

Within each analysis run, quality assurance/quality control (QA/QC) included blanks (empty quartz boats), standard reference materials (SRMs) and/or CRMs [DORM-4, fish protein (NRCC), 0.412 μg/g; ERM-CE464, tuna (European Reference Materials), 5.24 μg/g; and TORT-3], and duplicate samples. All blanks (n = 187) were below a detection limit of < 0.0001 μg/g. The recovery of SRMs/CRMs (mean ± 1 SD) was 96 ±

2% for DORM-4 (*n* = 82; range = 89 to 99%), 99 ± 4% for ERM-CE464 (*n* = 80; range = 90 to 107%) and 96 ± 5% for TORT-3 (*n* = 16; range = 84 to 99%). The average relative percentage difference between sample duplicates (*n* = 180) was 3%. One set of QA/QC was included with every 10 samples analyzed.

2.4 Statistical analysis

Linear regressions were used to examine the relationship between body length or whole weight and Hg concentration for each investigated species. If the assumptions of normality or equal variance were not met, the Hg concentrations were natural log transformed prior to analysis, after which the majority of species passed both or one of the assumptions. Glass et al. (1972) argued that the analysis of variance (ANOVA) is robust to violations of a single assumption, so as long as one assumption was met, the test was considered valid. If both assumptions were not met after natural log transformation, the relationship was examined using a Spearman's rank correlation coefficient.

Red drum, black drum, spotted seatrout, gafftopsail catfish, spanish mackerel, and southern flounder were considered as residential species. Analysis of covariance (ANCOVA), with body length used as the covariate, was used to determine whether there was a significant difference ($P < 0.05$) in Hg concentration for red drum, spotted seatrout, and southern flounder between individuals collected in Port Aransas versus individuals collected from Port Mansfield and Port Isabel; Port Mansfield and Port Isabel populations were combined due to close proximity. If the ANCOVA did not satisfy the two assumptions, the data was natural log transformed prior to analysis. Due to unequal variances and sample sizes, a Kruskal-Wallis was used as a non-parametric analysis to determine site differences in black drum populations followed by a Wilcoxon signed rank pairwise test using Bonferroni correction. Although gafftopsail catfish and spanish mackerel are considered residential species, spatial differences were not examined because of low sample size between site populations ($n = 27$ for Port Aransas and $n = 5$ for Port Mansfield/Port Isabel for gafftopsail catfish; $n = 19$ for Port Aransas and $n = 1$ for Port Mansfield/Port Isabel for spanish mackerel). All statistical analysis was performed using Sigmaplot 12 and 13 (Systat Software Inc., San Jose, CA) and Program R version 3.5.0. Confidence levels for all statistical analysis was $P < 0.050$.

III. RESULTS

This study reported Hg concentrations in 1,468 samples collected from 26 species of fish and 4 species of shellfish. Out of the total species sampled, 43% (n =13) were nearshore fish species, 43% (n =13) were offshore fish species, and 14% (n = 4) were shellfish species (Table 1). The nearshore fishes had the greatest number of samples analyzed (51.3%; n = 753), followed by offshore fishes (38.1%; n = 559), and shellfishes (10.6%; n =156).

3.1 Intraspecies variability in Hg concentration

Twenty-six of the 30 investigated species had a large enough sample size that the relationship between Hg concentration and body length could be examined. These relationships are shown in Figures 2 – 7 and the corresponding linear regression analysis results are shown in Table 2. Due to small sample sizes (n = 1), the relationship between Hg concentration and body length could not be investigated for cubera snapper, rainbow runner, remora, and white marlin (*Kajikia albida*), but Hg concentration for each species is shown in Table 3.

There was a positive relationship ($P < 0.05$) between Hg concentration and body length or weight for 18 of the investigated species, including 7 nearshore fish species (black drum, red drum, spotted seatrout, southern flounder, spanish mackerel, gafftopsail catfish, and atlantic croaker; Fig. 2 and 3), 10 offshore fish species (blackfin tuna, yellowfin tuna, wahoo, little tunny, king mackerel, red snapper, dolphinfish, cobia, great barracuda, and crevalle jack; Fig. 5 and 6), and one shellfish species (American oyster;

Fig. 7). Linear regressions were used to describe the relationship for all species (Table 2) except little tunny which was examined using the Spearman's rank correlation coefficient (Rs = 0.679; P <0.001). Striped mullet was the only species that had a negative relationship between Hg concentration and body length (P <0.001; Fig, 3). In comparison, there was no correlation between Hg concentration and body length or weight (P > 0.05) for the remaining 7 species (pinfish, mangrove snapper, blue marlin, sailfish, brown shrimp, blue crab, and brief squid; Fig. 3, 4, and 7).

3.2 Spatial differences in Hg concentrations in residential species

No site difference was found for southern flounder (P = 0.836); however, it was found for red drum, black drum, and southern flounder. Red drum (ANCOVA; n = *169*, df = 166; R^2 = 0.109; P <0.001) and spotted seatrout (ANCOVA; n = 190; df = 187; R^2 = 0.0991; P = 0.017) had a greater Hg concentration in Port Mansfield/Port Isabel (0.723 and 1.1 µg/g dry wt, respectively) than in Port Aransas (0.569 and 0.921 µg/g dry wt, respectively) when taking into account body length. The results are as follows:

Red drum:

Port Mansfield/Port Isabel: [mean Hg = 0.723 µg/g; n = 139)

Port Aransas: [mean Hg = 0.569 µg/g; n = 30)

Spotted seatrout:

Port Mansfield/Port Isabel: [mean Hg = 1.093 µg/g; n = 162)

Port Aransas: [mean Hg = 0.921 µg/g; n = 28)

The mean Hg concentration was higher in black drum from Texas City (0.682 µg/g dry wt) than from Port Aransas (0.309 µg/g dry wt) [Kruskal-Wallis rank sum test: $\chi^2 = 8.654$; df = 2; *P*-value = 0.013, (Wilcoxon rank sum test: *P*-value = 0.045)].

3.3 Interspecies variability in Hg concentrations

Concentrations were converted from dry weight to wet weight to allow for comparison to federal and state advisory limits in section 3.4, which are issued on a wet weight-basis. All nearshore fish, offshore fish, and shellfish species were combined together to calculate the mean wet weight Hg concentration for each group (Fig. 8). Offshore fish had a mean Hg concentration (1.61 µg/g; *n* = 558) which was 9.2-times higher than nearshore fish (0.175 µg/g; *n* = 750 and 46-times higher than shellfish (0.035 µg/g; *n* = 156).

The mean wet weight Hg concentration in all fish and shellfish species with a sample size > 1 is shown in Fig. 9. For the offshore fish species, the mean Hg concentration was highest in blue marlin (11.6 µg/g) and sailfish (1.21 µg/g) and lowest in red snapper (0.164 µg/g) and dolphinfish (0.157 µg/g); for the nearshore fish species the mean Hg concentration was highest in gafftopsail catfish (0.387 µg/g) and pinfish (0.346 µg/g) and lowest in Atlantic croaker (0.022 µg/g) and striped mullet (0.011 µg/g); and for the shellfish species the mean Hg concentration was highest in blue crab (0.096 µg/g) and brief squid (0.0286 µg/g) and lowest in brown shrimp (0.0086 µg/g) and American oyster (0.006 µg/g).

A detailed breakdown of Hg concentrations in each fish and shellfish species, including the mean, standard deviation, and minimum and maximum Hg values on a dry and wet weight basis, along with the corresponding body length values (mean, standard deviation, and range) are shown in Appendix A – C.

3.4 Species exceeding federal and state Hg advisory levels

Table 4 shows the species that exceeded federal and state advisory levels and the body length at which they started to exceed each one. Of the 30 investigated species, the majority of species (73.3%; n = 22) had individuals that exceeded the EPA human health criterion of 0.3 µg/g wet wt, whereas 43.3% (n = 13) of species had individuals that exceeded the TSDHS human health-based standard of 0.7 µg/g wet wt, and 36.6% (n = 11) of species had individuals that exceeded the FDA action limit of 1 µg/g wet wt. All species that exceeded the FDA advisory were offshore fish species; blue marlin and white marlin were the only species where every individual exceeded the FDA commercial action limit.

Based on the mean wet weight Hg concentration in species with a sample size greater than 1 (n = 26; Fig. 9), only 3 offshore fish species (blue marlin, sailfish, and king mackerel) had a mean Hg concentration that exceeded the FDA action limit. In comparison, gafftopsail catfish was the only nearshore fish species that had a mean Hg concentration that exceeded the EPA human health criterion, and no shellfish species exceeded the EPA advisory level, indicating that on average, these species are low in Hg.

IV. DISCUSSION

This study examined the intra- and interspecies variability in Hg concentrations in 30 species of fish (nearshore and offshore) and shellfish, as well as investigated the relationship between Hg concentrations and body length for 26 of these species caught recreationally and commercially along the Texas coast. This study also gives a 5-6 year update on Hg concentrations in fishes sampled by the TDSHS as well as species not investigated by the state. The majority of species investigated (69%) had a significant positive relationship between Hg concentration and body size. Spatial differences in the Hg concentration in muscle tissue were only seen in pelagic residential fish species and not in benthic residential fish species. Overall, offshore fish species had a higher average Hg concentration compared to nearshore fish species and shellfish. Larger and longer lived offshore fish species had more individuals that exceeded federal and state Hg advisory level than smaller, shorter lived species.

4.1 Intraspecific Variability in Hg Concentration

A positive relationship between Hg concentration and body size was observed in the majority of the species investigated in this study. Positive relationships were possibly attributed to the majority of Hg being present as MeHg in fish (> 84%; Hammerschmidt and Fitzgerald, 2006) which has a high assimilation efficiency from the diet and low efflux rate from the body (90% vs 0.8%/d; Dutton and Fisher, 2011, 2010; Wang and Wong, 2003); as a result larger individuals examined had muscle Hg concentration that built up over time. Ontogenetic dietary switchovers could also be responsible for the accumulation of Hg in individuals due to seasonality differences in food availability (Liu et al. 2012; Szczebak and Taylor, 2001) in which more contaminated prey could be consumed during those seasonal periods.

The findings of this study did not always fit the prediction that there would be a positive relationship between Hg concentrations and body length in all species as 30% of the species examined had no relationship/negative relationship between body length and Hg concentration. A negative relationship was observed between Hg concentration and body length in striped mullet, is attributed to growth dilution as the rate of growth is greater than the rate of Hg accumulation. McDonough and Wenner (2003) have found that juvenile striped mullet rapidly start to grow when body sizes average around 4 cm in South Carolina and this species have shown to have patterns of growth dilution in the Gulf of California (Ruelas-Inzunza et al. 2017). Growth dilution has also been found in other fish species including Atlantic salmon (*Salmo* salar) and tilapia (*Oreochromis niloticus*) (Wang and Wang, 2012; Ward et al. 2010). Ontogenetic dietary switchovers can also be potentially responsible for the accumulation pattern. Striped mullet have been observed to be more zooplanktivorous post-larvae at the sizes of 1 – 3 cm and begin to shift towards a more adult diet comprised of benthic organisms, detritus and sediment as juveniles grow larger than 5 – 8 cm in length (Cardona, 2015). Individuals within this study ranged from 9 to 20 cm indicating that a dietary switchover to a more benthic diet could have already started within the selected individuals.

Species that were found to have no relationship between Hg concentration and body size were pinfish, sailfish, blue marlin, mangrove snapper, brief squid, blue crab and brown shrimp. Determination of relationships can depend on sample sizes and or the size range of the species being investigated. Narrow size ranges with a large sample set can give rise to high variability in concentrations within a given species because the range in the *x*-value is limited, thus, preventing any relationship to be seen between the variables being examined just like pinfish and brown shrimp within this study. Sample sizes (> 20) increase the likelihood of a relationship due to an increase in power with an increase in sample size (Gewurtz et al. 2011; Zar, 1984); however, in the case of blue marlin (n =5) and sailfish (n = 10), a low sample size with high variation can also prevent a relationship from being detected. Fishing tournaments can also have an effect on the detection of a relationship. Tournaments have to follow federal and state landing size regulations which can skew the size distribution of the fish species being sampled. For example, larger sized fish are more likely to be opportunistically sampled during the 2nd day of a tournament than the first. If the investigated fish species were commercially caught, a wider range of sample sizes could be collected due to different landing sizes.

Seventy five percent of the shellfish species investigated did not display a positive relationship between Hg concentration and body size. Shellfishes are opportunistic omnivores that feed lower on the food web (Evans et al. 2000; Rosas et al. 1994; Lassuy, 1983; Stanley and Sellers, 1986). It has been examined that phytoplankton and zooplankton have a lower percentage of Hg present as MeHg (Hammerschmidt and Fitzgerald, 2006) and a lower overall concentration of Hg, in which shellfish consuming these prey items do not accumulate Hg to greater concentrations. Pinfish are an omnivorous species that feed on animals as juveniles and undergo an ontogenetic dietary switchover in which they consume on plant material as adults (Morris et al. 2016; Luczkovich and Stellwag, 1993). A complete switch in dietary sources from a source of higher Hg to lower could explain as to why pinfish contained varying concentrations of Hg but did not see a trend in Hg accumulation over time.

Several prior studies have investigated the relationship between Hg concentration and body length in species examined in this study. The positive relationship observed in this study was also found in yellowfin tuna in the Pacific and Indian Ocean (Jindasa et al. 2015; Ordiano-Flores et al. 2011; Kojadinovic et al. 2006) and dolphinfish, wahoo, king mackerel, cobia, and red snapper in the Indian and western Atlantic Ocean (Adams 2018; Sinkus et al. 2017; Petre et al. 2012; Adams 2009; Kojadinovic et al. 2006). While this study did not find a positive relationship for blue marlin, mangrove snapper, spotted seatrout, and blue crab, previous studies in the western Atlantic Ocean, Gulf of California, and Pacific Ocean did (Taylor and Calabrese, 2018; Drevnick and Brooks, 2017; Vega-Sanchez et al. 2017; Burger et al. 2011; MacDonald et al. 2010; Adams and Onorato, 2005); this difference is mostly likely due to a smaller sample size and/or narrow range in body size. Lastly, striped mullet has also shown to have a negative relationship in a previous study from the Gulf of California (Ruelas-Inzunza et al. 2017).

4.2 *Spatial differences in Hg concentrations in residential species*

Site differences were detected in red drum and spotted seatrout, with the Port Mansfield/Port Isabel population having higher Hg concentrations in comparison to the Port Aransas population. Red drum and spotted seatrout share similar diets that consist of crabs, shrimp and small fish (Sager 2002; Sharf and Schlight, 2000; Lorio and Perret, 1978) in which differences in Hg concentrations between the two populations are more likely due to differences in the environmental concentration of Hg rather than difference in diet. Reasons for this difference are not apparent, but could be due to possibly increased atmospheric deposition as a result of poorer environmental regulations in Mexico (Rutter et al. 2009). Black drum populations had a higher average concentration of Hg in Texas City (located in Galveston Bay) than Port Aransas. Previous studies found that Hg concentrations in Galveston Bay have remained relatively low (Al Mukaimi et al. 2018; Apeti et al. 2012; Han et al. 2006) even with the vast amount of oil refineries surrounding urban areas (Lan et al. 2014; Brooks et al. 2010), suggesting biological factors are more of an influence for Hg uptake. Black drum consume mollusks and small crustaceans (Brown et al. 2008; Perry and McIlwain, 1986), so individual biological factors such as growth rate and age (Wang, 2012; Trudel and Rasmussen, 2006) in which larger individuals (slower growing) had had more time to accumulate Hg within their tissues. Black drum are known to live up to 60 years and based off of other studies, individuals captured in this study were anywhere from 2-5 years in age (Murphy et al. 1998; Murphy and Taylor, 1989) suggesting Texas City (which had the larger size range) individuals are older and have accumulated more Hg in their tissues over time in comparison to Port Aransas individuals.

There were no site differences detected in southern flounder populations. Flounder are known to feed on small crustaceans and fish that are lower in the food web and have been found to contain consistently low Hg concentrations along the Texas coastline (Sager, 2002; Reagan and Wingo 1985). Because populations of red drum and spotted seatrout were 1.27 and 1.19-times higher, respectively, in Port Mansfield/Port Isabel populations than in Port Aransas, further studies are warranted to determine the cause of this difference in Hg concentration.

The relationship between Hg concentration and body length in the 4 residential species were examined by location and as all locations combined. Red drum, spotted seatrout, southern flounder, and black drum all showed a positive relationship when sites were combined; however, when split into both populations, red drum and spotted seatrout had a positive relationship in Port Mansfield/Port Isabel, but no relationship in Port Aransas. As a whole, the relationship for both species were most likely influenced by the Port Mansfield/Port Isabel sites as the majority of samples (82.2% and 85.3%, respectively) were taken from those locations. Black drum sites when separated had a positive relationship for Texas City, but not for Port Aransas. Port Aransas had a greater number of individuals sampled (62.5% of n = 40) than Texas City, but Texas City had a larger size range (TC: 41.3-77.5 cm and PA: 44.1-70.2 cm) which could explain why there was a relationship seen in Texas City but not in Port Aransas.

4.2 Interspecific differences in Hg concentration

To examine the differences between offshore fish species, nearshore fish species, and shellfish species, all species within each group were combined to compare average wet weight concentrations. Offshore fish species had an average concentration of 1.61 µg/g (n = 558) which was 9.2-times higher than nearshore fish (0.175 µg/g; n = 750) and 46-times higher than shellfish (0.035 µg/g; n = 156). As trophic level increases, the percentage of MeHg increases as well. Fish species that are larger and mainly piscivorous are known to contain more Hg within their tissues than those that feed more on invertebrates (Tremain and Schaefer, 2015; Adams and Onorato, 2005) due to an increase in Hg content within their prey species (Payne and Taylor, 2010; Cai et al. 2007).

However, variation of Hg concentration within a trophic level can be influenced by diet preferences, growth patterns, and longevity.

Species such as the tunas have multiple closely related species that can have varying Hg concentrations and preference in diets. For example, blackfin tuna had a higher average concentration of 0.63 µg/g wet wt. whereas the larger species, yellowfin tuna, had an average of 0.25 µg/g wet wt and little tunny had an average concentration of 0.866 µg/g wet wt within this study. The variation in Hg concentration between the 3 species could potentially be explained through differences in age and diet. Yellowfin tuna had an average length of 115 cm in which Lessa and Duarte-Neto (2004) estimated that individuals of that size were just over 2 years of age. Blackfin tuna on the other hand had an average of 66 cm in length in which the individuals of that size are estimated to be around the age of 3 or 4 (Adams and Kersetter, 2014), and at a mean length of 60 cm little tunny is estimated to be anywhere from 2-5 years in age (Adams and Kersetter, 2014; NOAA 1983). All three tuna species have similar diets consisting of small fish, shellfish, and cephalopods (Ahrabi-Nejad, 2014; Teffer et al. 2014). Within the 3 species, little tunny has the longest lifespan in which the higher concentration is most likely reflective of their age. Tuna species are just one example of species that can have Hg concentrations that are reflective of their age status. Blue marlin is another fish species that are known to be larger in size and are a longer lived species with a lifespan of 15+ years (Orbesen et al. 2008) and in that time they can accumulate higher concentrations of Hg. Blue marlin within this study had the highest average Hg concentration out of any species investigated within this study at 11.7 µg/g wet wt.

Comparisons between the average wet wt Hg concentrations measured in this study and previous findings for the Gulf of Mexico and Atlantic Ocean can be found in Table 5. When examining nearshore species, black drum, spotted seatrout, southern flounder and pinfish had average concentrations that were higher than previous findings within the Gulf of Mexico. However, spanish mackerel, Atlantic croaker, and striped mullet had lower concentrations. For offshore species, blue marlin, king mackerel, cobia and crevalle jack had a higher average Hg concentration, whereas wahoo had a lower average than previously measured in the Gulf of Mexico. American oyster and brown shrimp also contained lower concentrations. Comparable concentrations were found in blackfin tuna, yellowfin tuna, and dolphinfish. However, more accurate comparisons between the Atlantic Ocean, previous Gulf studies, and this study are lacking due to body lengths not being listed within the comparable literature.

Mercury concentrations measured in fish sampled from the Atlantic Ocean were higher in red drum, pinfish, wahoo, cobia, and blue crab than this study, whereas dolphinfish, king mackerel, and great barracuda contained concentrations similar to those in the Atlantic Ocean. Species that contained lower concentrations in the Gulf of Mexico than the Atlantic were spotted seatrout, atlantic croaker, striped mullet, mangrove snapper, and red snapper; these species were also smaller on average than those measured in the Atlantic Ocean which probably influenced the Hg concentration.

Blue marlin had an average Hg concentration of 11.7 µg/g wet wt. for this study which was much higher than a study from the Pacific Ocean with a concentration of 1.91 µg/g (Vega-Sanchez et al. 2017). Sailfish on the other hand had a slightly lower concentration of 1.2 µg/g in this study in comparison to the average concentration in the

Pacific Ocean (1.48 µg/g; Soto-Jiminez et al. 2010). These differences are most likely due to differences in the amount of atmospheric Hg deposition and diet preferences.

This study found a wide variation in Hg concentration among individuals within certain species such as pinfish and king mackerel. Pinfish had a narrow size range of 12.6–26.9 cm and concentrations varied greatly from 0.044-0.909 µg/g indicating that biological processes are most likely factors in Hg accumulation than diet and age as all individuals were caught in the same location. The migratory status of fish can also contribute to the variability in Hg concentrations as spatial differences potentially influence Hg concentrations within a particular species being investigated. King mackerel exhibited a wide variation in Hg concentrations across all sizes sampled. There are two populations of king mackerel within the Gulf of Mexico; one along the Yucatan peninsula with a higher Hg concentration and the other population around Florida with a lower Hg concentration (NOAA, 2017; Johnson et al. 1994; Grimes et al. 1990). Both populations converge along the Texas coastline (Barile, 2013; Levenson and Axelrad, 2006) which could account for the large variability in Hg concentration at a given body length, as it was not determined in this study which population individual fish belonged to.

Species such as dolphinfish, yellowfin tuna, and wahoo can have overlapping dietary niches (Teffer et al. 2015; Cai et al. 2007; Collette and Nauen, 1983) but differ in Hg values within their tissues due to differences in growth rate and age (Rudershausen et al. 2010). Dolphinfish are known to have very fast growth rates in the Gulf of Mexico and can grow up to 3.54 mm per day, can reach sexual maturity by the age of 5-7 months, and have a lifespan of 3-4 years (Young, 2014; Schwenke and Buckel, 2008). Their first year of growth can reach lengths of 80 – 150cm in TL and begins to slow by the age 3 when they reach up to 200 cm (Brewton et al. 2014; Young, 2014). Because of their fast growth rate, dolphinfish continually have lower Hg values because of growth dilution in comparison to longer lived species like yellowfin tuna and wahoo that have lifespans up to 5 and 10 years respectively (Zischke, 2012; Stequert et al. 1996). The low Hg concentration in smaller dolphinfish has also been reported in other studies (Araújo and Cedeño-Macias, 2016; Cladis et al. 2014; Adams, 2009), however, once the growth rate slows down, the concentration of Hg increases rapidly.

4.3 Species exceeding federal and state Hg advisories

Currently, the TDSHS has blue marlin listed as the only species to not be consumed at any size caught and the results from this study support this consumption ban as all individuals sampled had concentrations over 3 µg/g wet weight. Other larger species like blackfin tuna, little tunny, crevalle jack, and wahoo have been listed to only be consumed by adults over the childbearing age twice per month in 8-ounce portions (TDSHS, 2013). Average concentration for little tunny was 0.866 µg/g when compared to TDSHS average of 0.499 µg/g and Hg concentrations investigated in this study support the current Texas consumption advisory for that species. However, blackfin tuna, crevalle jack, and wahoo were found to have Hg averages within this study that were lower than TDSHS sampling averages which can be easily influenced by larger individuals and a smaller sampling size (n=6, 30.5 - 81 cm; TDSHS: n = 7, 83.8 – 105.4 cm), blackfin tuna (n = 76, 44.1- 84.2; TDSHS: n = 8, 76.2-90.8 cm) and wahoo (n = 61, 95.3-155.6 cm; TDSHS: n = 9, 101.6 – 156.8 cm) (TDSHS, 2012). It should be considered that species such as cobia, great barracuda and sailfish be added to the state Hg advisory list due mean concentrations being above 0.7 µg/g and that 97%, 100%, and 90% individuals that exceeded the EPA, 58%, 61%, and 80% individuals exceeded the TDSHS, and 42%, 26%, and 70% of individuals exceeded the FDA respectively.

The size of fish more often than not can help predict the concentration of Hg within its muscle tissue yet, many of the guidelines issued do not include size ranges that can be considered relatively safe to consume. Texas has only king mackerel listed with a size limit for individuals over 88.9cm to be safely consumed once per month. Currently, there is no fishing and consumption size limit for blackfin tuna in Texas waters. This study had a larger blackfin tuna sample size (n = 76) in comparison to the TDSHS 2011 study (n = 8), however, the TDSHS study sampled larger individuals than this study (size range = 76.2 – 90.8 cm and 44.1 – 84.2 cm, respectively). It could be suggested that there be further investigation into adding a consumption size advisory for this species as 43% of individuals sampled were over the Texas consumption guideline. The same could be suggested little tunny as 69% of individuals sampled over 42 cm were above Texas guidelines as well. Establishing size guidelines for fish consumption can be beneficial to consumers but could also cause issues in fish populations. Smaller individuals are considered to be younger and an increased selection of those smaller individuals can have negative consequences on fish populations. Any individual caught before sexual maturity, especially the longer lived species such as tuna, prevents the chance to spawn and could potentially lead to a population decline (Stergiou et al. 2009; Myers and Mertz, 1998).

4.4 Conclusions

This study provides the most up-to-date information on Hg concentrations in fish and shellfish caught commercially and recreationally along the Texas coast. The findings of this study not only provide insight into the health of the Gulf of Mexico ecosystem, but also provide valuable data to the seafood consuming public and to the medical community who advise patients what seafood to eat. This could ultimately be beneficial to the entire nation as the Gulf of Mexico supplies 16% of the commercial fish (Buck et al. 2015; Harris et al. 2012a, 2012b) and consumer preference can possibly pressure fisheries to catch more species that are lower in the food web such as shrimp, red drum, and spotted seatrout that have low tissue Hg concentrations.

The TSDHS issued consumption advisories for fish species commonly caught along the Texas Coast in 2011 (TSDHS, 2013). There has been no update to these advisories in the last several years and this study expanded on the number of species examined and increased the sampling size. Finally, the Texas Parks and Wildlife Department (TPWD) is currently ranking several of the investigated species (e.g., blue marlin, sailfish, king mackerel, cobia, yellowfin tuna red snapper, and southern flounder) to determine whether they are species of greatest conservation need, (SGCN). If they are listed, then the data in this study can be used in conservation and recovery plans since Hg is a known threat to the health of fish.

4.5 Future Directions

A large number of species were investigated in this study; however 16 of the species including blue marlin, white marlin, sailfish, crevalle jack, and blue crab had a low sample number and/or a narrow size range. An increase in sample size and a wider range of body size would help determine whether species that showed no relationship between Hg concentration and body size in this study actually have a positive relationship. Species that are not targeted by fishing tournaments but are commercially important in the Gulf of Mexico such as yellow amberjack (*Seriola lalandi*), red grouper (*Epinephelus morio*), black grouper (*Mycteroperca bonaci*), and vermillion snapper (*Rhomboplites aurorubens*) should be sampled to increase our understanding of Hg concentrations in other economically important species. Samples from these species can potentially be obtained through working with seafood processing plants located in Austin, San Antonio, and on the coast.

Currently, Hg advisories regarding seafood consumption are issued based on the concentration of Hg in muscle tissue, but several studies have shown that selenium (Se), an essential element, has an antagonistic relationship with Hg and at high enough concentration may have a protective role against Hg toxicity (Diop and Amara 2016; Polak-Juszczak 2015; Squadrone et al. 2015; Berry and Ralston, 2008; Ralston et al. 2008; Ralston et al. 2007). It has been proposed that the Se:Hg molar ratio can be used as a seafood safety criterion in risk assessment because if the molar ratio is greater than 1:1, then selenium may have a protective role against the effects of Hg exposure. Because selenium has been shown to prevent Hg toxicity, listing the molar ratios can help create a better risk assessment for fish that contain high Hg and Se concentrations. However, extensive data collection is still needed to understand the complex interaction between Hg and Se, the intra- and interspecies variability in the molar ratio, and the relationship between human health and Se:Hg molar ratios (Burger and Gochfeld, 2013, 2012, 2011a; Burger et al. 2013, 2012; Burger, 2012).

Finally, stable isotopes are a useful tool to estimate the flow of organic matter through the marine food web and the trophic position of each species (Richert et al. 2015). Nitrogen, ($\delta^{15}N$) is widely used to estimate trophic positions as consumers are enriched by 3-4‰ from their diets (DeNiro and Epstein, 1981). Carbon ($\delta^{13}C$) lends additional and more accurate source of information on the flow of organic carbon, specifically the diets of marine species (Li et al. 2016). The positioning of an organism's trophic status based on their diet as fractionation of $\delta^{15}N$ and $\delta^{13}C$ values varies as it moves up the food chain and organisms becomes more enriched as trophic levels increase (Atwell et al. 1998, Cabana and Rasmussen, 1994). Sampling on all trophic levels, especially the base of the food web, would give a baseline to determine absolute stable isotope values which would give better trophic level estimations. Isotopes are also used to estimate the biomagnification of Hg in food webs. By integrating $\delta^{15}N$ values and Hg concentrations, we can give better estimates on how Hg is transferred through trophic levels (Feng et al. 2016; Karimi et al. 2013; Adams and Paperno, 2012; Di Beneditto et al. 2011, Payne and Taylor, 2010; Chasar et al. 2009).

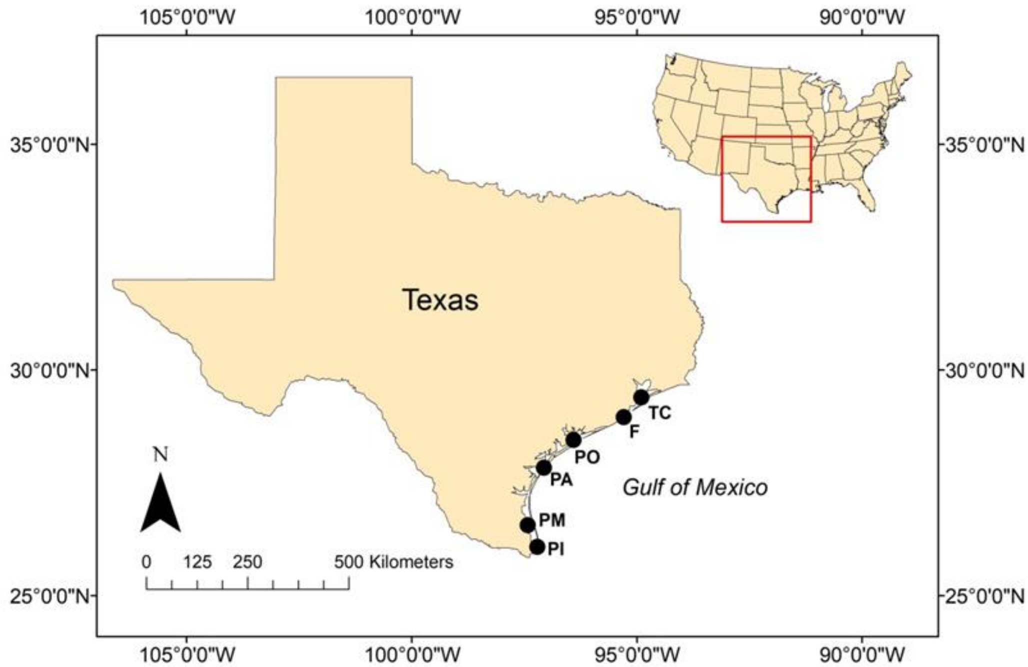

Figure 1: Fish and shellfish sampling locations on the Texas coast. TC = Texas City; F = Freeport; PO = Port O'Connor; PA = Port Aransas; PM = Port Mansfield; PI = Port Isabel.

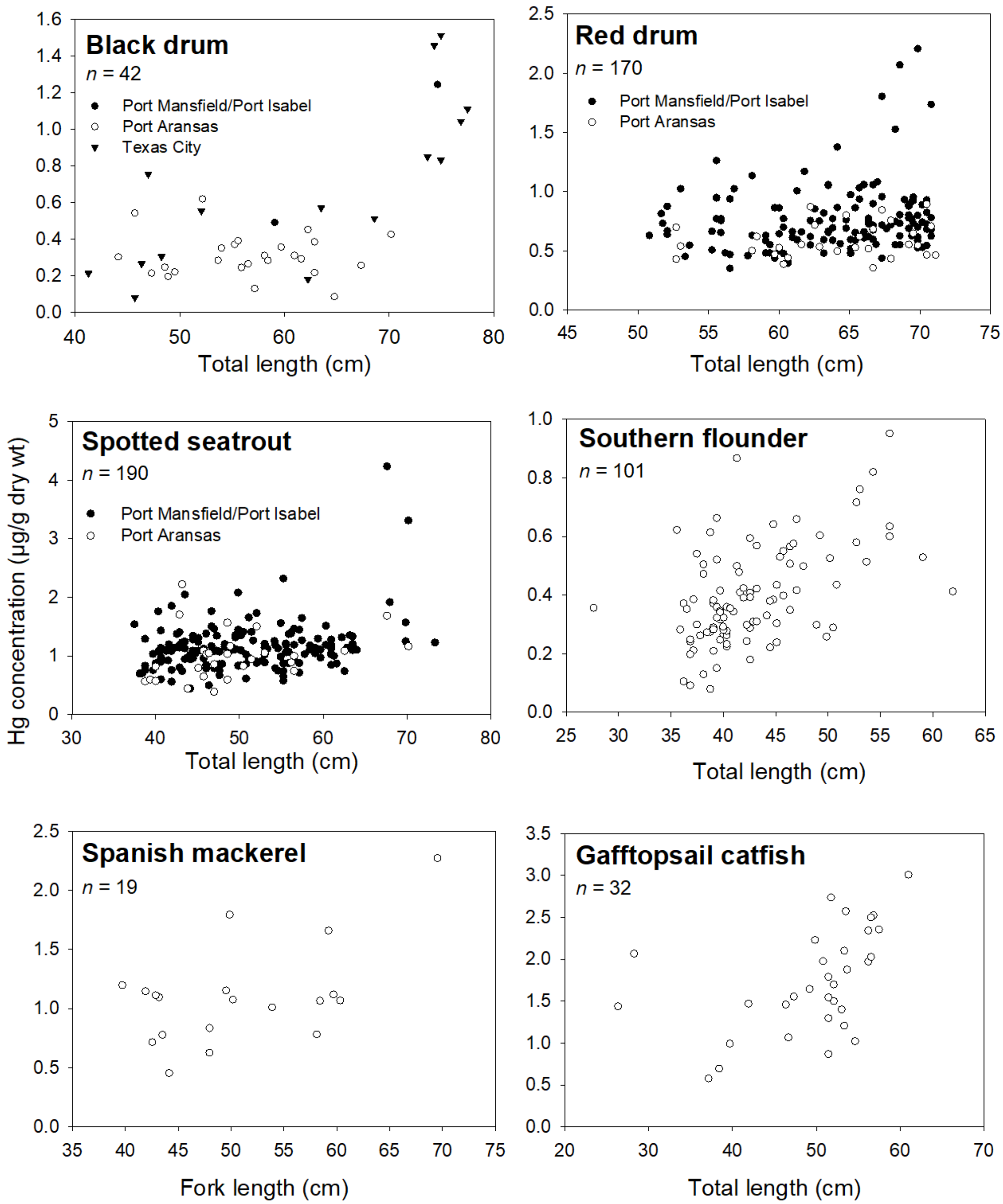

Figure 2: Relationship between Hg concentration (µg/g dry weight) and body length in commonly fished nearshore fish species. Linear regression results describing the relationships are shown in Table 2.

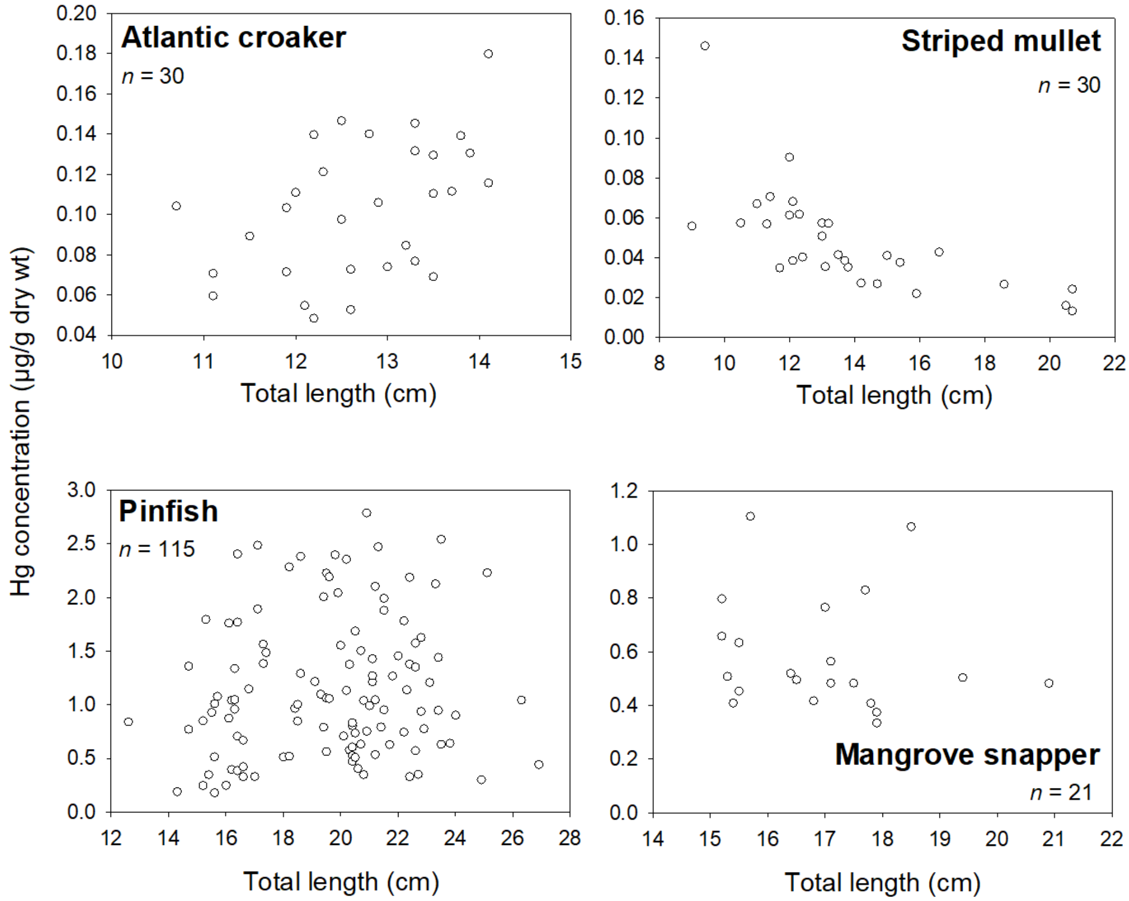

Figure 3: Relationship between Hg concentration (µg/g dry weight) and body length in small nearshore forage fish species. Linear regression results describing the relationships are shown in Table 2.

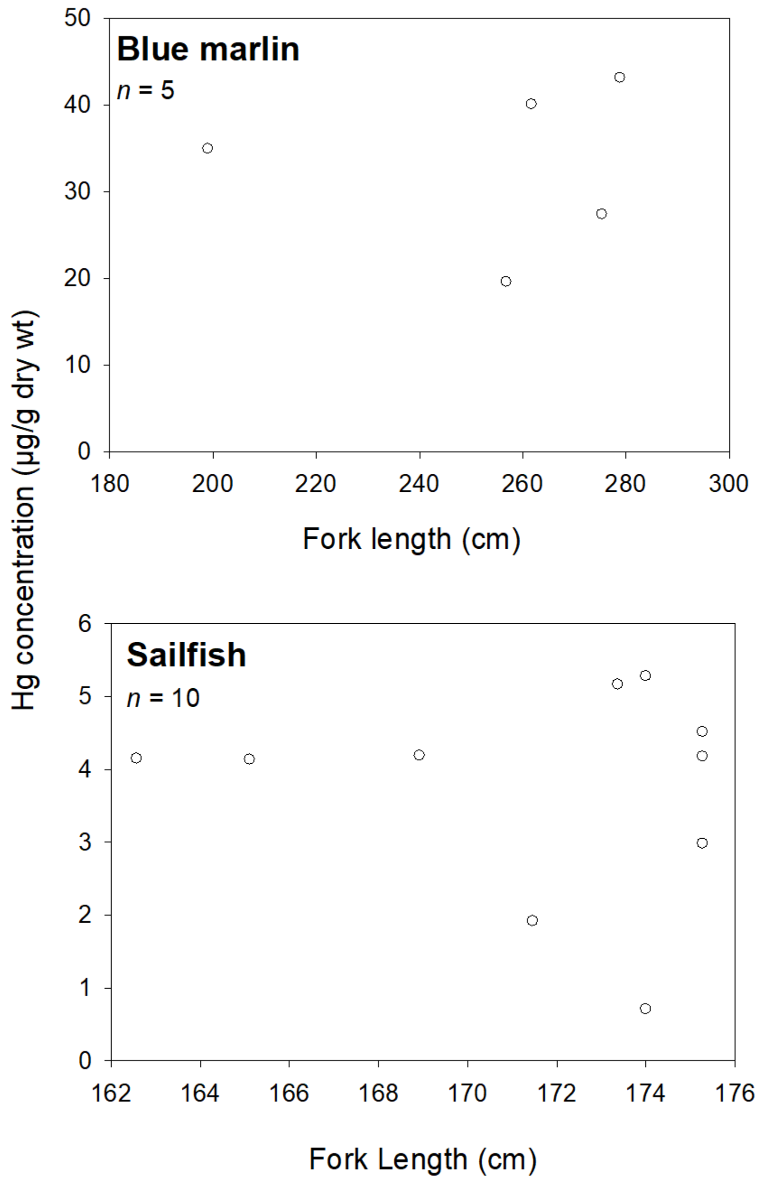

Figure 4: Relationship between Hg concentration (µg/g dry weight) and body length in billfish. Linear regression results describing the relationships are shown in Table 2.

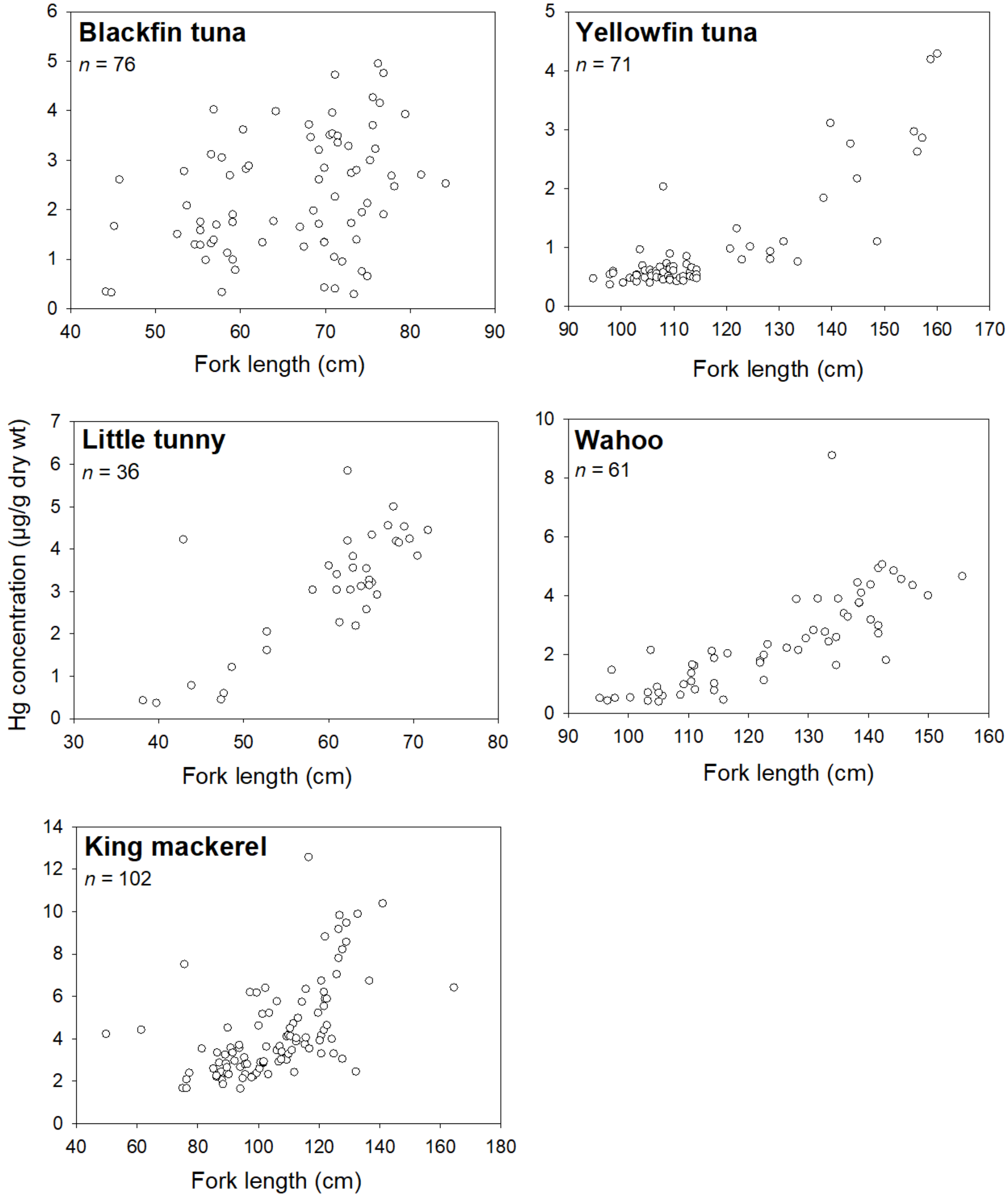

Figure 5: Relationship between Hg concentration (µg/g dry weight) and body length in offshore fish species in the family Scombridae (tunas, mackerels, and bonitos). Linear regression results describing the relationships are shown in Table 2.

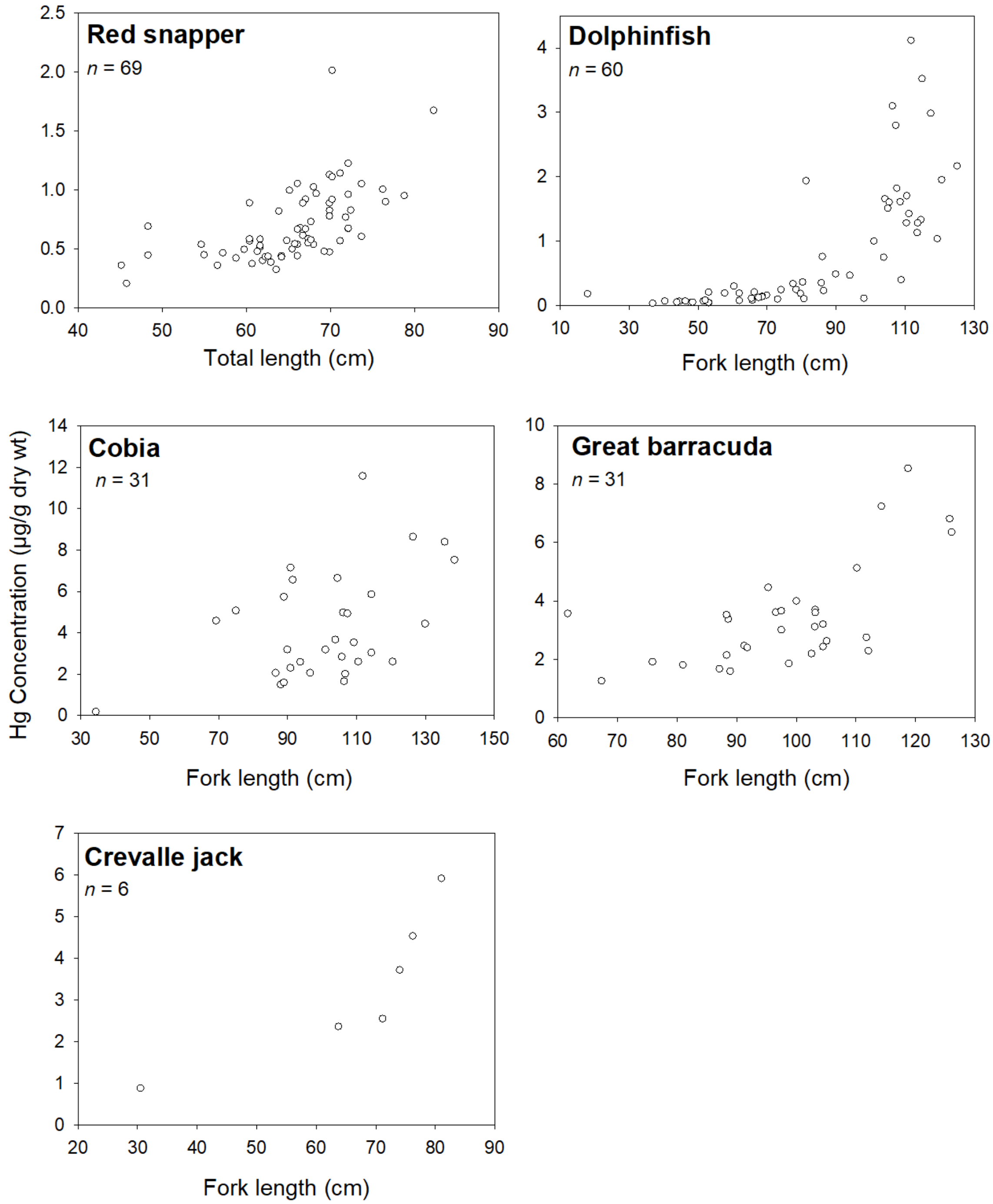

Figure 6: Relationship between Hg concentration (µg/g dry weight) and body length in other commonly fished offshore fish species. Linear regression results describing the relationships are shown in Table 2.

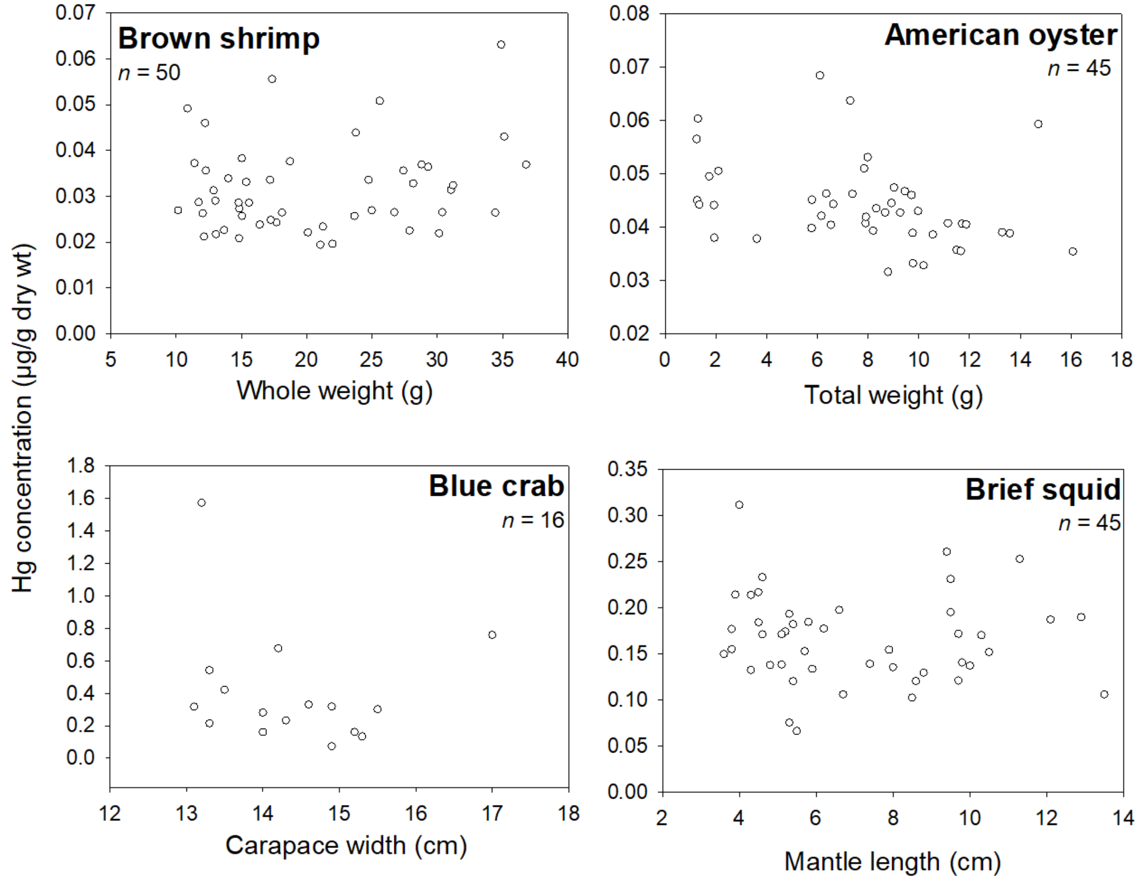

Figure 7: Relationship between Hg concentration (µg/g dry weight) and body length or weight in shellfish. Linear regression results describing the relationships are shown in Table 2.

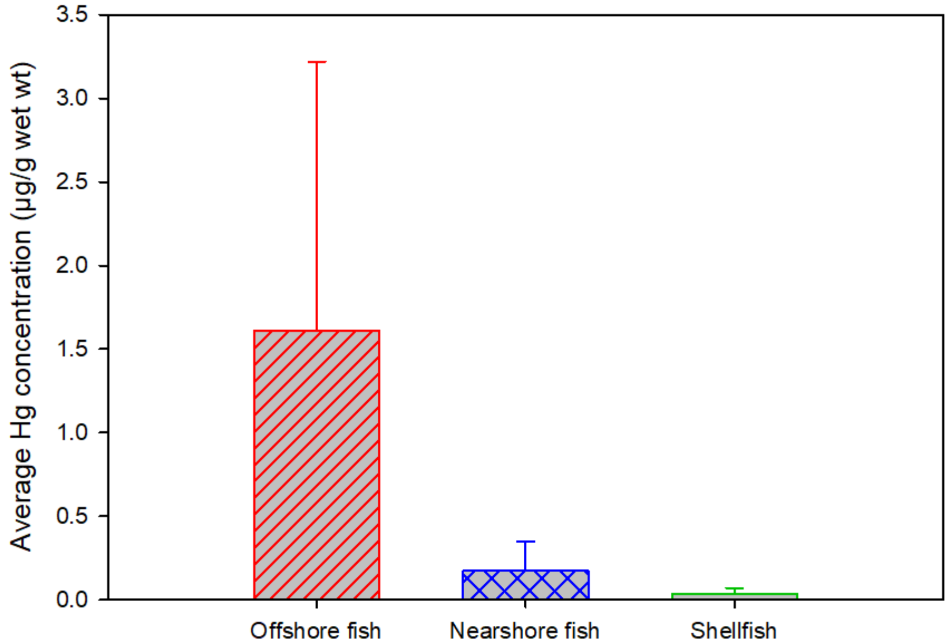

Figure 8: Average Hg concentrations (µg/g wet weight) in all species of offshore fish, nearshore fish, and shellfish caught along the Texas coast combined by group. Error bars are 1standard deviation.

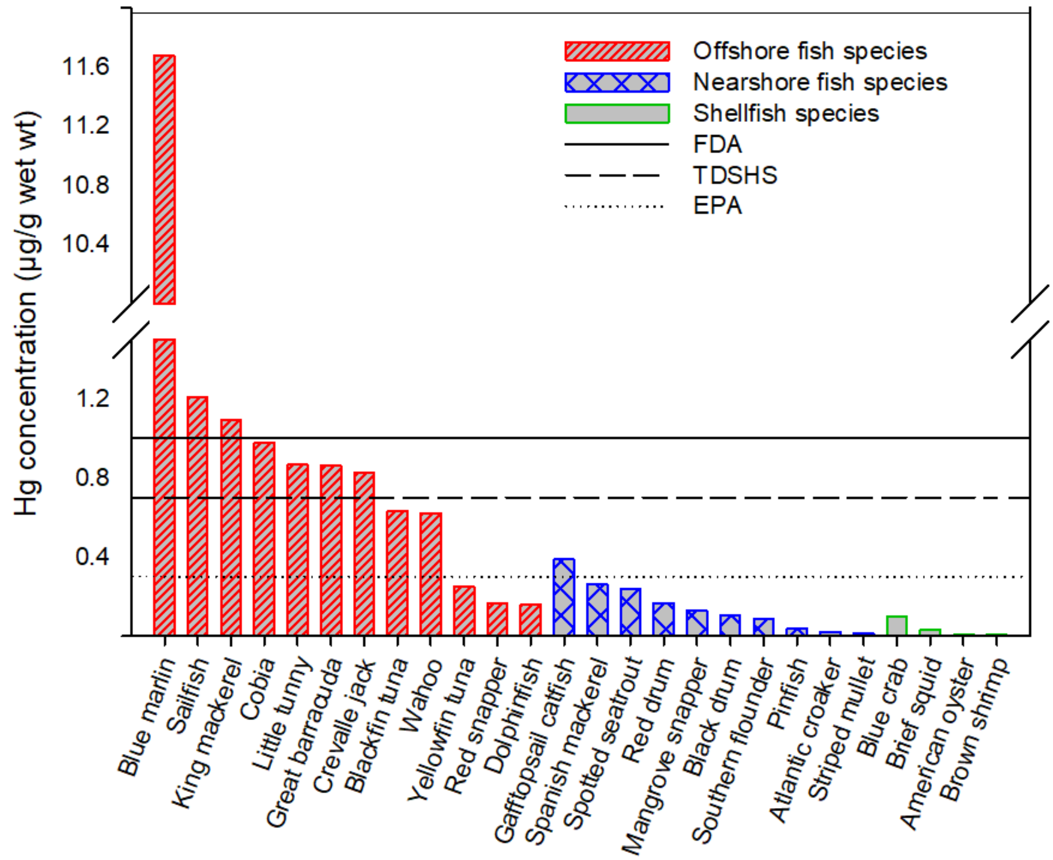

Figure 9: Average Hg concentrations (µg/g wet weight) in commercially and recreationally caught fish and shellfish species caught along the Texas coast. The federal and state Hg advisory levels are included (FDA 1 µg/g wet weight Hg action limit; TDSHS 0.7 µg/g wet weight Hg human health-based standard; and EPA 0.3 µg/g wet weight human health criterion).

Table 1: List of fish and shellfish species investigated in this study with corresponding sample size, sampling locations, and percentage water content in muscle or whole body tissue (mean ± standard deviation). TC = Texas City; F = Freeport; PO = Port O'Connor; PA = Port Aransas; PM = Port Mansfield; PI = Port Isabel.

Common name	Species	*n*	Location	% Water
Nearshore fish				
Black drum	*Pogonias cromis*	42	TC, PA, PM, PI	78 ± 1
Red drum	*Sciaenops ocellatus*	170	PA, PM, PI	78 ± 1
Spotted seatrout	*Cynoscion nebulosus*	190	PA, PM, PI	79 ± 2
Southern flounder	*Paralichthys lethostigma*	101	PA, PM, PI	78 ± 3
Spanish mackerel	*Scomberomorus maculatus*	19	PA, PI	76 ± 1
Gafftopsail catfish	*Bagre marinus*	32	PA,PI	78 ± 2
Atlantic croaker	*Micropogonias undulatus*	30	PM	79 ± 2
Striped mullet	*Mugil cephalus*	30	PI	77 ± 2
Pinfish	*Lagodon rhomboides*	115	PM	71 ± 4
Mangrove snapper	*Lutjanus griseus*	21	PM	78 ± 1
Cubera snapper	*Lutjanus cyanopterus*	1	PA	77 ± NA
Rainbow runner	*Elagatis bipinnulata*	1	PA	74 ± NA
Remora	*Remora remora*	1	PA	80 ± NA
Offshore fish				
Blue marlin	*Makaira nigricans*	5	F, PO	73 ±1
White marlin	*Kajikia albida*	1	PI	74 ± NA
Sailfish	*Istiophorus platypterus*	10	PM, PI	68 ± 4
Blackfin tuna	*Thunnus atlanticus*	76	F, PO, PA,PM,PI	72 ± 2
Yellowfin tuna	*Thunnus albacares*	71	F, PO, PA, PM, PI	73 ± 2
Little tunny	*Euthynnus alletteratus*	36	PA, PM, PI	72 ± 2
Wahoo	*Acanthocybium solandri*	61	F, PO, PA, PM, PI	74 ± 1
King mackerel	*Scomberomorus cavalla*	102	PA, PM, PI	74 ± 2
Red snapper	*Lutjanus campechanus*	69	PA,PM	77 ± 1
Dolphinfish	*Coryphaena hippurus*	60	F, PO, PA, PM, PI	78 ± 3
Cobia	*Rachycentron canadum*	31	PA, PM, PI	77 ± 1
Great barracuda	*Sphyraena barracuda*	31	PA,PI	75 ± 2
Crevalle jack	*Caranx hippos*	6	PO, PA, PI	75 ± 2
Shellfish				
Brown shrimp	*Penaeus aztecus*	50	PA	73 ± 2
American oyster	*Crassotrea virginica*	45	PI	87 ± 3
Blue crab	*Callinectes sapidus*	16	TC	78 ± 4
Brief squid	*Lolliguncula brevis*	45	TC	83 ± 2

Table 2: Linear regression results describing the relationship between Hg concentration and body length or weight for each investigated species. The relationships for black drum, red drum, and spotted seatrout are provided for each sampling location as well as all locations combined. TL = total length; FL = fork length; TW = total weight; WW = whole weight; CW = carapace width; TC = Texas City; PA = Port Aransas; PM/PI = Port Mansfield/Port Isabel. NA = not applicable because the relationship was not significant.

Species	y = a + bx	df	F	R^2	P-value
Nearshore fish					
Black drum	Hg = -0.873 + (0.0228 * TL)	40	29.7	0.426	<0.001
TC	Hg = -0.902 + (0.0256 * TL)	13	18.8	0.56	<0.001
PA	NA	NA	NA	NA	0.756
Red drum	ln(Hg) = -1.047 + (0.0107 * TL)	167	5.69	0.033	0.018
PA	NA	NA	NA	NA	0.315
PM/PI	ln(Hg) = -1.063 + (0.0116 * TL)	137	5.62	0.0395	0.019
Spotted seatrout	ln(Hg) = -0.536 + (0.0118 * TL)	188	16.6	0.0814	<0.001
PA	NA	NA	NA	NA	0.069
PM/PI	ln(Hg) = -0.432 + (0.0102 * TL)	160	12.0	0.07	<0.001
Southern flounder	ln(Hg) = -2.636 + (0.0378 * TL)	99	29.5	0.23	<0.001
PA	Hg = -0.520 + (0.0218 * TL)	16	11.5	0.419	0.004
PM/PI	ln(Hg) = -2.262 + (0.0297 * TL)	81	20.8	0.204	<0.001
Spanish mackerel	Hg = -0.281 + (0.0273 * FL)	17	6.59	0.28	0.02
Gafftopsail catfish	ln(Hg) = -0.702 + (0.0239 * TL)	30	9.08	0.233	0.005
Atlantic croaker	Hg = -0.108 + (0.0166 * TL)	28	7.32	0.207	0.011
Striped mullet	ln(Hg) = -1.315 - (0.134 * TL)	28	57.7	0.673	<0.001
Pinfish	NA	NA	NA	NA	0.079
Mangrove snapper	NA	NA	NA	NA	0.479
Offshore fish					
Blue marlin	NA	NA	NA	NA	0.957
Sailfish	NA	NA	NA	NA	0.758
Blackfin tuna	Hg = -0.707 + (0.0452 * FL)	74	10.6	0.126	0.002
Yellowfin tuna	ln(Hg) = -4.060 + (0.0325 * FL)	69	216	0.758	<0.001
Wahoo	ln(Hg) = -4.552 + (0.0417 * FL)	58	156	0.73	<0.001
King mackerel	ln(Hg) = -0.241 + (0.0151 * FL)	100	52.4	0.344	<0.001
Red snapper	ln(Hg) = -2.933 + (0.0381 * TL)	67	52.9	0.441	<0.001
Dolphinfish	ln(Hg) = -4.830 + (0.0462 * FL)	58	191	0.768	<0.001
Cobia	ln(Hg) = -1.127 + (0.0234 * FL)	29	17.9	0.383	<0.001
Great barracuda	ln(Hg) = -0.848 + (0.0201 * FL)	29	20.4	0.413	<0.001
Crevalle jack	ln(Hg) = -1.273 + (0.0351 * FL)	4	53.9	0.931	0.002
Shellfish					
Brown shrimp	NA	NA	NA	NA	0.214
American oyster	ln(Hg) = -3.009 - (0.0161 * TW)	43	6.46	0.131	0.015
Blue crab	NA	NA	NA	NA	0.537
Brief squid	NA	NA	NA	NA	0.667

Table 3: Mercury concentration (µg/g dry weight) and the corresponding body length for each fish species with a sample size of 1. TL = total length; FL = fork length.

Common Name	**Length (cm)**	**Hg (µg/g dry wt)**
Cubera snapper	64.1 (TL)	2.26
Rainbow runner	66.3 (TL)	0.117
Remora	69.9 (TL)	3.06
White marlin	176.5 (FL)	4.34

Table 4: Percentage of fish and shellfish species that exceeded the EPA Hg human health criterion of 0.3 µg/g wet wt, TDSHS Hg human health-based standard of 0.7 µg/g wet wt, and FDA Hg action limit of 1 µg/g wet wt, with the corresponding body length at which the Hg concentration began to exceed these advisory limits. *fork length; **total length; ***carapace width

Species	**Exceed EPA Advisory**	**% Exceeded**	**Body length (cm)**	**Exceed State Advisory**	**% Exceeded**	**Body length (cm)**	**Exceed FDA Advisory**	**% Exceeded**	**Body Length (cm)**
Nearshore									
Black drum	Yes	2.38	74.9	No	-	-	No	-	-
Red drum	Yes	2.96	67.3	No	-	-	No	-	-
Spotted seatrout	Yes	14.7	37.5	Yes	0.5	67.6	No	-	-
Spanish mackerel	Yes	15.8	49.9	No	-	-	No	-	-
Gafftopsail catfish	Yes	78.1	26.4	No	-	-	No	-	-
Pinfish	Yes	49.6	14.7	Yes	10.4	17.1	No	-	-
Cubera snapper	Yes	100	64.14	No	-	-	No	-	-
Remora	Yes	100	69.9	No	-	-	No	-	-

Table 4 Continued: Percentage of fish and shellfish species that exceeded the EPA Hg human health criterion of 0.3 µg/g wet wt, TDSHS Hg human health-based standard of 0.7 µg/g wet wt, and FDA Hg action limit of 1 µg/g wet wt, with the corresponding body length at which the Hg concentration began to exceed these advisory limits. *fork length; **total length; ***carapace width

Species	Exceed EPA Advisory	% Exceeded	Body length (cm)	Exceed State Advisory	% Exceeded	Body length (cm)	Exceed FDA Advisory	% Exceeded	Body length (cm)
Offshore									
Blue marlin	Yes	100	198.9	Yes	100	198.9	Yes	100	198.9
White marlin	Yes	100	176.5	Yes	100	176.5	Yes	100	176.5
Sailfish	Yes	90	162.6	Yes	80	162.6	Yes	70	162.6
Blackfin tuna	Yes	85.5	42.1	Yes	43.4	45.7	Yes	17.1	56.8
Yellowfin tuna	Yes	16.9	107.95	Yes	8.5	139.7	Yes	1.41	160.0
Little tunny	Yes	86.1	42.9	Yes	69.4	42.9	Yes	41.7	42.9
Wahoo	Yes	70	97.24	Yes	38.3	128.0	Yes	18.3	128.0
King mackerel	Yes	100	49.7	Yes	80.2	49.7	Yes	42.6	49.7
Red snapper	Yes	2.9	70.2	No	-	-	No	-	-
Dolphinfish	Yes	23.3	81.28	No	-	-	No	-	-
Cobia	Yes	96.8	69.2	Yes	58.1	69.2	Yes	41.9	74.9
Great barracuda	Yes	100	61.7	Yes	61.3	61.7	Yes	25.8	95.3
Crevalle jack	Yes	83.3	63.7	Yes	50	74.0	Yes	33.3	76.2
Invertebrate									
Blue crab	Yes	6.25	13.2	No	-	-	No	-	-

Table 5: Comparison of the average Hg concentrations (µg/g wet weight) between this study and other studies in the Gulf of Mexico (GoM) and the Atlantic Ocean (AO). NA = not applicable because the study did not report the body size.

Species	This Study	GOM	Size range (cm)	AO	Size range (cm)	Sources
Black drum	0.103	0.037	NA	-	NA	Showater 2010
Red Drum	0.162	-	NA	0.134	29.5 - 86.3	Evans and Crumley, 2005
Spotted Seatrout	0.237	0.122	NA	0.33-0.624	25.7 - 55.0	Showater 2010; Adams et al. 2010; Evans and Crumley, 2005
Southern flounder	0.089	0.03	NA	-	NA	Showater 2010
Spanish mackerel	0.263	0.264-0.415	NA	-	NA	Showater 2010; Thera and Rumbold, 2014
Atlantic Croaker	0.022	0.046	NA	0.026-0.5	12.8-28.6	Tremain and Shaefer, 2015; Cannon 2017; Showater 2010
Striped Mullet	0.011	0.052	NA	0.015	13.9-54.6	Tremain and Shaefer, 2015; Showater 2010
Pinfish	0.346	0.076	NA	0.078	12.3-21.3	Tremain and Shaefer, 2015; Thera and Rumbold 2014
Mangrove snapper	0.128	-	NA	0.183	15.9 - 49.7	Evans and Crumley, 2005
Blue Marlin	11.600	10.52	256-311	-	NA	Cai et al. 2007
Blackfin tuna	0.630	0.39 - 0.64	27-80,	-	NA	Kuklyte 2012; Cai et al. 2007
Yellowfin tuna	0.246	0.18 - 0.36	22-147	-	NA	Nicklisch et al. 2017; Kuklyte 2012; Cai et al. 2007
Wahoo	0.619	0.73-0.78	91-152	0.6	100- 150	Petre et al. 2012; Kuklyte 2012; Cai et al. 2007
King Mackerel	1.1	0.96-1.04	70-98	1.16	80 - 160	Petre et al. 2012; Kuklyte 2012; Cai et al. 2007
Red snapper	0.164	0.21	NA	0.18	18.2-90.5	Sinkus et al. 2017; Thera and Rumbold 2014
Dolphinfish	0.157	0.07-0.21	43-123	0.07-0.205	40 - 120	Petre et al. 2012; Teffer et al. 2014; Kuklyte 2012; Cai et al. 2007
Cobia	0.977	0.808-0.890	18.4-170.0	0.673	39.2-153.3	Adams 2018; Cai 2007
Great barracuda	0.864	-	NA	0.73-1.71	NA	Drescher et al. 2014; Rumbold et al. 2018
Crevalle jack	0.829	0.087-0.290	NA	-	NA	Showater 2010; Thera and Rumbold, 2014
Brown shrimp	0.0086	0.033	NA	-	NA	Harris et al. 2012b
American oyster	0.006	0.023	NA	-	NA	Apeti et al. 2012
Blue Crab	0.096	-	NA	0.078	7.5 - 19.6	Adams and Engel, 2014

APPENDIX SECTION

Table A: Hg concentrations (μg/g dry and wet wt) for each investigated nearshore fish species with corresponding range in body size. TL = total length; FL = fork length; NA = not applicable due to a sample size of 1.

	Body length (cm)			Hg (μg/g dry wt)			Hg (μg/g wet wt)		
Species	Mean	SD	Range	Mean	SD	Range	Mean	SD	Range
Black drum	58.9	10.12	41.3 – 77.5 (TL)	0.469	0.353	0.080 – 1.51	0.103	0.075	0.017 – 0.324
Red drum	63.8	5.383	50.8 – 71.1 (TL)	0.732	0.276	0.351 – 2.20	0.162	0.062	0.077 – 0.493
Spotted seatrout	50.7	7.952	37.5 – 73.3 (TL)	1.13	0.43	0.386 – 4.23	0.237	0.089	0.082 – 0.871
Southern flounder	42.8	5.759	27.6 – 61.9 (TL)	0.396	0.168	0.079 – 0.952	0.089	0.038	0.018 – 0.205
Spanish mackerel	50.7	8.235	39.7 – 69.5 (FL)	1.1	0.425	0.454 – 2.27	0.263	0.108	0.104 – 4.234
Gafftopsail catfish	49.6	8.057	26.4 – 61 (TL)	1.73	0.613	0.577 – 3.01	0.387	0.134	0.125 – 0.674
Atlantic croaker	12.7	0.917	10.7 – 14.1 (TL)	0.103	0.033	0.048 – 0.179	0.022	0.007	0.010 – 0.039
Striped mullet	13.8	3.088	9 – 20.7 (TL)	0.048	0.257	0.013 – 0.146	0.011	0.005	0.003 – 0.033
Pinfish	19.6	2.892	12.6 – 26.9 (TL)	1.15	0.646	0.179 – 2.79	0.346	0.218	0.044 – 0.909
Mangrove snapper	16.9	1.5	15.2 – 20.9 (TL)	0.586	0.214	0.334 – 1.11	0.128	0.048	0.069 – 0.246
Cubera snapper	NA	NA	64.1 (TL)	NA	NA	2.25 – NA	NA	NA	0.510 – NA
Rainbow runner	NA	NA	66.3 (TL)	NA	NA	0.117 – NA	NA	NA	0.031 – NA
Remora	NA	NA	69.9 (TL)	NA	NA	3.06 – NA	NA	NA	0.604 – NA

Table B: Hg concentrations (µg/g dry and wet wt) for each investigated offshore fish species with corresponding range in body size. TL = total length; FL = fork length; NA = not applicable due to a sample size of 1.

	Body length (cm)			Total Hg (µg/g dry wt)			Total Hg (µg/g wet wt)		
Species	Mean	SD	Range	Mean	SD	Range	Mean	SD	Range
Blue marlin	254.3	32.28	198.9 – 278.8 (FL)	33.1	9.58	19.6 – 43.2	11.7	6.052	3.86 – 18.64
White marlin	NA	NA	176.5 (FL)	NA	NA	4.34 – NA	NA	NA	1.11 – NA
Sailfish	172.5	4.544	162.6 – 175.3 (FL)	3.73	1.45	0.715 – 5.29	1.21	0.506	0.227 – 1.84
Blackfin tuna	66.1	9.412	44.1 – 84.2 (FL)	2.28	1.2	0.291 – 4.95	0.63	0.32	0.078 – 1.33
Yellowfin tuna	115.4	16.376	94.6 – 160.0 (FL)	0.938	0.87	0.370 – 4.29	0.246	0.216	0.099 – 1.03
Little tunny	60	9.083	38.1 – 71.7 (FL)	3.08	1.4	0.368 – 5.85	0.866	0.383	0.107 – 1.64
Wahoo	123.8	16.064	95.3 – 155.6 (FL)	2.41	1.65	0.399 – 8.77	0.619	0.425	0.104 – 2.29
King mackerel	105.2	17.836	49.7 – 164.5 (FL)	4.3	2.22	1.652 – 12.6	1.1	0.54	0.441 – 3.07
Red snapper	65.5	7.118	45.1 – 82.2 (TL)	0.702	0.32	0.208 – 2.02	0.164	0.071	0.048 – 0.447
Dolphinfish	81.8	26.685	17.9 – 125.1 (FL)	0.814	0.991	0.033 – 4.12	0.157	0.175	0.008 – 0.66
Cobia	101.2	20.46	34.3 – 138.4 (FL)	4.28	2.57	0.183 – 11.6	0.977	0.609	0.039 – 2.85
Great barracuda	98.1	14.881	61.7 – 126.1 (FL)	3.43	1.76	1.26 – 8.54	0.864	0.469	0.316 – 2.39
Crevalle jack	66.1	18.362	30.5 – 81 (FL)	3.33	1.78	0.886 – 5.91	0.829	0.444	0.210 – 1.48

Table C: Hg concentrations (µg/g dry and wet wt) for each investigated shellfish species with corresponding range in body size. WW = whole weight; CW = carapace width; ML = mantle length.

	Body length (cm)			**Total Hg (µg/g dry wt)**			**Total Hg (µg/g wet wt)**		
Species	Mean	SD	Range	Mean	SD	Range	Mean	SD	Range
Brown shrimp	20.5	7.691	10.2 – 36.8 (WW)	0.031	0.01	0.019 - 0.063	0.0086	0.003	0.005 – 0.017
American oyster	7.8	3.799	1.3 – 16.1 (WW)	0.044	0.007	0.032 - 0.068	0.006	0.002	0.003 – 0.011
Blue crab	14.4	1.055	13.1 – 17.0 (CW)	0.406	0.365	0.074 - 1.574	0.096	0.095	0.014 – 0.405
Brief squid	7.0	2.727	3.6 – 13.5 (ML)	0.166	0.048	0.066 - 0.312	0.0286	0.008	0.012 – 0.051

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