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Mercury bioaccumulation in offshore reef fishes from waters of the Southeastern USA☆

Wiley Sinkus [a,*], Virginia Shervette [b], Joseph Ballenger [c], Lou Ann Reed [d], Craig Plante [a], Byron White [c]

[a] College of Charleston, Grice Marine Laboratory, 205 Fort Johnson Rd, Charleston, SC 29412, USA
[b] University of South Carolina Aiken, 471 University Pkwy, Aiken, SC 29801, USA
[c] South Carolina Department of Natural Resources, Marine Resources Research Institute, 217 Fort Johnson Rd, Charleston, SC 29412, USA
[d] National Oceanic and Atmospheric Administration Hollings, Marine Laboratory, 331 Fort Johnson Rd, Charleston, SC 29412, USA

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Mercury (Hg) concentrations and nitrogen ($\delta^{15}N$) and carbon ($\delta^{13}C$) stable isotopic ratios were measured to assess differences in Hg bioaccumulation in four predatory fish species (*Mycteroperca microlepis, Lutjanus campechanus, Caulolatilus microps,* and *Serioli dumerili*) of high commercial and recreational importance in Atlantic waters of the southeastern US. Positive relationships existed between Hg and length, weight, and age, for all species, strongest for *M. microlepis* and *L. campechanus*. Intraspecific Hg concentrations also strongly correlated with $\delta^{15}N$ for all species, and $\delta^{13}C$ for only *L. campechanus*, and *S. dumerili*. Comparisons of stable isotopes between species and their impact on mean Hg concentration were inconclusive. This study is the first to report Hg concentrations for *C. microps*. The current study provides data for an under-sampled region, explores how feeding ecology impacts Hg uptake in commonly co-occurring fishes, and raises questions of the importance of sex and reproduction in Hg accumulation for marine fishes.

1. Introduction

Mercury (Hg), a naturally occurring element, is ubiquitous in the environment throughout the world (US EPA, 1997). After being released into the atmosphere by both natural and anthropogenic sources, with anthropogenic accounting for 70% of total Hg input over last 100 years, it is deposited on land, locally, regionally or globally (Pirrone et al., 2010). Mercury eventually makes its way to aquatic ecosystems through runoff and coastal erosion (Schuster et al., 2002), where it can be transformed to the water- and fat-soluble methylmercury (MeHg; Compeau and Bartha, 1985) and can enter the aquatic food web at its base through microbial assimilation (Kojadinovic et al., 2006). Through bioaccumulation and biomagnification, Hg builds to levels in organisms significantly higher than concentrations found in their surrounding environments, typically to greater concentrations in higher-level consumers (Thera and Rumbold, 2014).

Mercury accumulates in tissues of predatory fishes mainly as MeHg, which often comprises more than 90% of the total Hg (THg) in fish muscle of carnivorous and omnivorous species (Bloom, 1992; Bank et al., 2007; Senn et al., 2010). The methylated form of Hg is considered to be extremely toxic due to the ease that it can penetrate membrane barriers in the brain and placenta, acting as a neurotoxin (Magos, 1968; Yang et al., 1997). A major source of MeHg in humans comes from the consumption of fish and other seafood (WHO, 1976).

While consuming large quantities of fish with high Hg levels can be harmful, the recognition of health benefits from eating fish is becoming more publicized. Understanding Hg concentrations in fish vital to consumers who need to make well–informed choices about eating fish with lower Hg concentrations. Fish provide the world, especially coastal areas, with a major source of a protein that has lower levels of saturated fats compared to red meat (Giovannucci et al., 1994; FAO, 2014). Many fish species also contain high quantities of long-chain omega–3 fatty acids, which are suggested to have beneficial effects for cardiovascular and cancerous diseases (König et al., 2005; Kim et al., 2009). Fish muscle tissue is also known to contain a micronutrient, selenium, utilized by the

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* Corresponding author.
E-mail address: sinkusw@dnr.sc.gov (W. Sinkus).

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human nervous system. Research suggests that selenium works to counteract harmful effects of Hg (Feroci et al., 2005).

In an effort to protect the public from chronic over−exposure to Hg, government and non-government entities, such as the US Environmental Protection Agency (US EPA), the US Food and Drug Administration (US FDA), and the National Resources Defense Council (NRDC), issue advisories to seafood consumers, directed towards women of childbearing age and children. The most recent Hg advisory, issued jointly by the US EPA and the US FDA (2014), recommends the target demographic avoid consuming shark, swordfish, king mackerel, and tilefish from the Gulf of Mexico (GOM), which are commonly found to have Hg concentrations above 1.0 ppm. The advisory also recommends limiting consumption of fish commonly found above the US EPA screening level of 0.3 ppm to one meal a month, but does encourage consuming at least one and up to three servings a week of fish with low Hg concentrations (US EPA, 2000; US EPA and US FDA, 2014; NRDC, 2014; US EPA, 2017).

While Hg is pervasive throughout the world, environmental concentrations are highly variable. Differences in fish muscle tissue Hg concentrations between the GOM and Atlantic waters of the southeastern US (ASEUS) have been documented for several species (Adams and Onorato, 2005; Adams and McMichael, 2007). Historically, regional and national advisories have not distinguished among the locations where marine fish species are caught. Only recently have the differences in Hg contamination between the GOM and the ASEUS been identified by a government−issued advisory (US EPA and US FDA, 2014). Hall et al. (1978) reported that Golden Tilefish *Lopholatilus chamaeleonticeps* from the GOM ($n = 60$) had high Hg concentrations (mean Hg > 1.25 ppm). Harris et al. (2012) provided a comparison of the relationship between fish size and Hg concentration in *L. chaeaeleonticeps* for the GOM and the ASEUS, documenting significantly lower concentrations of Hg from the ASEUS samples for all size classes. A similar issue of elevated Hg concentration in the GOM compared to the ASEUS may occur for other species, but sufficient samples have not been analyzed from across the ranges.

Mercury concentrations in fish is thought to increase with feeding at higher trophic levels (Senn et al., 2010). Determination of relative trophic position using nitrogen stable isotope values ($\delta^{15}N$) is possible due to the selective excretion of the lighter isotope (^{14}N) by organisms, leading to an accumulation of ^{15}N at higher trophic levels (Deniro and Epstein, 1981; Post, 2002). Relative carbon stable isotope values ($\delta^{13}C$ can be used to differentiate ultimate source of carbon (terrestrial, benthic, or pelagic) because prey feeding primarily on different carbon sources exhibit different levels of ^{13}C enrichment (Deniro and Epstein, 1978; Fry and Sherr, 1984; France, 1995). The consumer typically has a $\delta^{15}N$ that is enriched by 3−4‰ (parts per thousand) compared to its prey; however, minimal changes occur to the $\delta^{13}C$ as carbon moves through the food web ($\delta^{13}C$ trophic enrichment = 0.4‰; Post, 2002). Results from Bank et al. (2007) show a model integrating $\delta^{15}N$ and $\delta^{13}C$ to predict Hg concentration in two snapper species was most effective ($r^2 = 0.83$), suggesting that a significant amount of the variability in Hg concentrations within these snapper species is related to trophic position or sources of carbon.

The compounding effects on Hg emissions of burning fossil fuels and climate change consequences, such as melting of polar ice sheets, leave future reductions in Hg emissions uncertain (Outridge et al., 2008). Changes in atmospheric concentration from Hg emissions can take from three to ten years to be reflected in the ocean and wildlife Hg concentrations (Harris et al., 2007; Sunderland and Mason, 2007), thus continued monitoring of Hg in seafood is key in keeping the public well informed.

1.1. Study species

The present study focused on four species of commercially and recreationally important marine fishes that were chosen based on similarities or differences in characteristics such as habitat selection, feeding habits, longevities, and growth rates. These four species are Gag Grouper *Mycteroperca microlepis*, Red Snapper *Lutjanus campechanus*, Greater Amberjack *Seriola dumerili*, and Blueline Tilefish *Caulolatilus microps*. Mercury concentrations in these species, excluding *C. microps*, have been reported prior to the current study (Bank et al., 2007; Cai et al., 2007; Petre et al., 2012; Tremain and Adams, 2012; Thera and Rumbold, 2014). Many of those studies, however, focused on samples from the GOM, had relatively low samples sizes, or were limited by a combination of these two factors.

The species *M. microlepis,* from the family Serranidae, is a long-lived, slow-growing species reaching a maximum age of 26 years, a maximum size of 1275 mm TL, with a Brody growth constant (k) from von Bertalanffy growth function of 0.354 in the Atlantic (Harris and Collins, 2000; NMFS, 2014). Adult *M. microlepis* are associated with inshore reef and shelf break habitats occurring at depths up to 110 m (Bullock and Smith, 1991). *Mycteroperca microlepis* exhibit variable movement, moving on average 150 km in the ASUES, yet 1/3 of the fish studied moved less than 2 km (McGovern et al., 2005). Diet of *M. microlepis* is composed of 78% fish and 22% decapod crustaceans using an index of relative importance (IRI) based on stomach content analysis in the GOM (Tremain and Adams, 2012). *Mycteroperca microlepis* are protogynous hermaphrodites with 50% of individuals transitioning from female to male at 9.7 years and 1049 mm TL (Reichert and Wyanski, 2005).

Lutjanus campechanus, family Lutjanidae, is a demersal species that lives in association with low- and high-relief hard bottom and reef ledge habitats occurring at depths over 80 m (Mitchell et al., 2014). This is a long-lived, slow-growing, gonochoristic species that attains a maximum documented age of 54 years in the Atlantic (McInerny, 2007), however, the maximum age reported from fisheries−independent sampling efforts is 26 years (Wyanski et al., 2015). The maximum size reported for the Atlantic population is 997 mm TL with a Brody growth constant (k) from von Bertalanffy growth function of 0.168 ($n = 3019$; Wyanski et al., 2015). Little is known about the movement of *L. campechanus* in the ASEUS, however studies in the GOM documented a mean moved distance of 29.6 km, a maximum moved distance of 352 km, and a large portion of fish remaining within 2 km of their tagged location (Szedlmayer and Shipp, 1994; Patterson et al., 2001). The diet of *L. campechanus* is divers; they are opportunistic predators with an observed shift in prey type with increasing fish size, from a prevalence of planktonic zooplankton to increased consumption of benthic crustaceans (Bradley and Bryan, 1975; Wells et al., 2008).

Caulolatilus microps, family Malacanthidae, is a demersal species associated with deepwater hard bottom habitat at depths up to 236 m (Parker and Ross, 1986) and occupies burrows in sandy sediments (Able et al., 1987). This is a long−lived, slow growing, gonochoristic species that can reach a maximum age of 43 years and maximum size of 884 mm TL with a Brody growth constant (k) from von Bertalanffy growth function of 0.08 (Harris et al., 2004). Dietary analyses for *C. microps* have found prey items from high relief, rocky outcroppings, and gently sloping areas (Ross and Huntsman, 1982). While adults seem to exhibit a high amount of variability in habitat selection, they have relatively high site fidelity (Ross and Huntsman, 1982; SEDAR, 2013).

Seriola dumerili is a coastal and pelagic species, that has been observed at depths up to 350 m and observed feeding on and near