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Bioaccumulation and trophic transfer of mercury in striped bass (*Morone saxatilis*) and tautog (*Tautoga onitis*) from the Narragansett Bay (Rhode Island, USA)

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ABSTRACT

We examined the bioaccumulation and trophic transfer of mercury in two marine finfish species, striped bass (*Morone saxatilis*) and tautog (*Tautoga onitis*), collected from the Narragansett Bay (Rhode Island, USA). For each of these target fish, white muscle tissue was analyzed for total mercury (Hg) and results were evaluated relative to fish age, body size, and Hg content of preferred prey. Dietary and stable isotope analysis was also used to elucidate the effect of trophic processes on Hg concentrations in fish. The Hg content of muscle tissue was positively correlated with fish age and length for both species, although striped bass accumulated Hg faster than tautog. Accelerated Hg bioaccumulation in striped bass is consistent with its high trophic level (trophic level = 4.07) and Hg-enriched prey (forage fish and macrocrustaceans; mean Hg content = 0.03 mg Hg kg wet wt^{-1}). In contrast, tautog maintain a lower trophic status (trophic level = 3.51) and consume prey with lower Hg levels (mussels and crabs; mean Hg content = 0.02 mg Hg kg wet wt^{-1}). Despite differences in Hg bioaccumulation between target fish, the mean Hg concentration of tautog exceeded levels in striped bass (0.24 and 0.16 mg Hg kg wet wt^{-1}, respectively) due to a disparity in age-at-catch between sampled groups (mean age of tautog and bass = 11.3 and 4.3 yr, respectively). Taking into account legal minimum catch lengths further revealed that 75.0% of legal-size striped bass (>70.2 cm TL; $n = 4$) and 44.8% of tautog (>40.6 cm TL; $n = 29$) had Hg levels beyond the US EPA regulatory threshold of 0.3 mg Hg kg wet wt^{-1}. Moreover, Hg-length relationships suggest that each target fish meets this threshold near their minimum legal catch length. Our findings reiterate the value of species ecology to improve predictions of fish Hg and permit better management of human contamination by this important dietary source.

1. Introduction

Methylmercury (MeHg) is widely recognized as one of the most widespread and toxic environmental contaminants affecting human health. For example, MeHg exposure has been linked to neurological and cardiovascular disorders, immune deficiencies, and reproductive deficits in humans (Moszczynski et al., 1990; Salonen et al., 1995; Grandjean et al., 1997; Sorensen et al., 1999). Dietary uptake of contaminated fish and shellfish is the most important mechanism by which humans are exposed to MeHg (Fitzgerald and Clarkson, 1991; US EPA, 1997; Hightower and Moore, 2003), and MeHg constitutes the majority of total mercury (Hg) in fish muscle tissue (>95%; Grieb et al., 1990; Bloom, 1992).

The uptake and accumulation of MeHg in aquatic food webs is affected by several biological and environmental variables (Weiner et al., 2003). The main biotic factors contributing to the MeHg burden in fish are age, body size, dietary preference, and trophic position, such that MeHg bioaccumulation and magnification increases in larger/older fish and those feeding at higher trophic levels (Weiner et al., 2003). To this end, understanding the human risk to MeHg exposure requires insight into: (1) the trophic transfer of contaminants through biotic receptors, including fish, and (2) the variability in fish MeHg concentrations as a function of life history (e.g., ontogenetic shifts in diet and habitat use, somatic growth, and longevity).

Visual estimates of a predator's stomach contents have traditionally been used to elucidate trophic relationships in aquatic communities. While providing valuable information on diet composition, stomach content analysis is limited because it only reflects immediate feeding activity. Conversely, stable isotope analysis is routinely used to quantify the relative trophic position of a species as a function of its time-integrated diet history (Michener and Schell, 1994). For example, nitrogen isotopic signatures ($^{15}N/^{14}N$) are effective at quantifying the trophic position of an organism because enrichment of the heavier isotope (^{15}N) occurs incrementally across trophic levels at a constant rate (~3–4‰; Michener and Schell, 1994). Conversely, carbon isotopic signatures ($^{13}C/^{12}C$) are consistent across trophic levels (<1‰ change between primary producer and consumer; Fry and Sherr, 1984), but are

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valuable biomarkers for identifying different sources of primary production (e.g., salt marsh grasses, macroalgae, benthic microalgae, and phytoplankton) (Peterson and Howarth, 1987), and therefore are effective at distinguishing between benthic and pelagic trophic linkages (France, 1995). Moreover, recent studies have utilized stable isotope techniques to determine patterns of trophic transfer of contaminants in food webs, yet the majority of this research has focused on freshwater systems (Jarman et al., 1996; Bowles et al., 2001; Power et al., 2002; Bank et al., 2007; Cai et al., 2007).

The purpose of this study was to examine the bioaccumulation and trophic transfer of total mercury (Hg) in striped bass (*Morone saxatilis*) and tautog (*Tautoga onitis*): two species of marine fish that support lucrative recreational and, to a lesser extent, commercial fisheries along the northeastern United States. Striped bass and tautog were collected from the Narragansett Bay (Rhode Island, USA), and therefore this study represents a comprehensive analysis of Hg contamination at relatively small spatial scales (<400 km^2). Furthermore, observed patterns of Hg concentration in striped bass and tautog were analyzed relative to the individual's age and body size. Conventional dietary analyses (i.e., visual observation of stomach contents) were also coupled with nitrogen and carbon stable isotope analyses to define key trophic pathways in the estuarine system and quantify the transfer of Hg contaminants in the food web.

2. Materials and methods

2.1. Target species

Striped bass are an estuarine-dependent species with a geographic distribution ranging from the Gulf of St. Lawrence to Florida (Collette and Klein-MacPhee, 2002). As obligate users of estuarine systems (Able, 2005), striped bass exhibit large plasticity in migration strategies that range from local seasonal movements within estuaries to extensive coastal migrations (Secor and Piccoli, 2007). Striped bass of various life history stages utilize multiple habitat-types, which are governed by the availability of preferred food resources, e.g., small forage fish and macrocrustaceans (Harding and Mann, 2003; Nelson et al., 2003, 2006). Moreover, somatic growth rates of striped bass are consistent with other temperate, long-lived fishes (~20–30 yr; Secor et al., 1995; Secor, 2000; Collette and Klein-MacPhee, 2002), but growth rates of striped bass demonstrate significant latitudinal differences (Welsh et al., 2003).

Tautog are a temperate wrasse distributed along the northwestern Atlantic coast extending from Nova Scotia to South Carolina, with peak concentrations occurring in coastal regions from Cape Cod to the Chesapeake Bay (Collette and Klein-MacPhee, 2002). In contrast to striped bass, tautog exhibit strong site fidelity (Olla et al., 1979; Able and Fahay, 1998) and only limited seasonal migrations that are attributed to estuarine residency during spring spawning events and the subsequent migration to nearshore wintering habitats (Olla et al., 1974; Briggs, 1977). The diet of tautog consists of epibenthic and encrusting invertebrates (e.g., brachyuran crabs and bivalves; Steimle et al., 2000), and the longevity of this slow-growing fish exceeds 30 yr (Cooper, 1967; Steimle and Shaheen, 1999).

2.2. Sample collection, processing, and preservation

Striped bass, tautog, and "bioavailable" prey were collected from the Narragansett Bay in 2006 and 2007 using bottom trawls, seines, fish traps, and hook and line (Fig. 1). "Bioavailable" prey are defined as small forage fish and invertebrates that were captured in the field and represent common food items of striped bass and tautog (Steimle et al., 2000; Nelson et al., 2003, 2006), i.e., river herring (*Alosa* spp.), bay anchovy (*Anchoa mitchilli*), Atlantic menhaden (*Brevoortia tyrannus*), scup (*Stenotomus chrysops*), green crab (*Carcinus maenas*), black-finger mud crab (*Panopeus herbstii*), sand shrimp (*Crangon septemspinosa*), and blue mussel (*Mytilus edulis*). For a complete description of gear specifications, collection locations, and sample frequency, refer to the sampling procedures identified in Lynch (2000) and Collie et al. (2008).

Fish and invertebrates captured in the field were immediately placed on ice for transportation and frozen at −4 °C after returning to the laboratory. Individuals were then partially thawed and measured for wet weight (g) and total length (TL cm; fish and shrimp), carapace width (CW cm; crabs), or shell height (SH cm; blue mussels) (Tables 1 and 2). Bioavailable prey were subsequently processed and analyzed as whole-body samples, with the exception of mussels that had their shells removed. For striped bass and tautog, ~2.5 g wet weight of white muscle tissue (with scales and skin removed) was excised from the dorsal region above the operculum using a stainless-steel scalpel (D_0 = 1 biopsy per left and right side of the fish; Fig. 2A). To ensure that the total mercury concentration of this biopsy was indicative of the whole-body filet, a sub-sample of bass and tautog (n = 18 and 14, respectively) had additional biopsies excised from the dorsal and lateral tissue along the anterior–posterior axis (D_{1-3} and L_{1-3}; 6 biopsies per left and right side of the fish; Fig. 2A). The stomachs of bass and tautog were also removed, and the contents were examined. The "recovered" prey items from dissected stomachs were identified to the lowest practical taxon, and where possible, measured for wet weight (g) and TL (cm; fish and shrimp) or CW (cm; crabs). For final preservation, all samples were freeze-dried for 48 h (Labconco FreeZone 4.5L Benchtop Freeze-Dry System), and subsequently weighed (g dry weight), homogenized with a mortar and pestle, and stored at room temperature in clear borosilicate 40-mL vials (Peterson et al., 2005).

2.3. Mercury analysis

Total mercury (Hg) was measured in the muscle biopsies and stomach contents (i.e., "recovered" prey) of striped bass and tautog, and whole-body samples of "bioavailable" prey using a DMA-80 Direct Mercury Analyzer (Cizdziel et al., 2002). For all target fish and prey, a sub-sample of freeze-dried/homogenized tissue (~40 mg) was added to the mercury analyzer. The instrument has a detection limit of 0.01 ng Hg (typical working range 0.05–600 ng), and employs thermal decomposition, amalgamation, and atomic absorption spectrophotmetry (EPA Method 7473; US EPA, 1998). The mercury analyzer was calibrated using standard reference materials (SRMs) of known Hg content and prepared by the National Research Council Canada, Institute of Environmental Chemistry (Ottawa, Canada), i.e., TORT-1 (lobster hepatopancreas) and DORM-2 (dogfish muscle). Calibration curves were highly significant (mean R^2 = 1.00; range R^2 = 0.99–1.00; $p < 0.0001$), and the recovery of TORT-1, DORM-2, and PACS-2 (marine sediment) SRMs ranged from 91.1% to 108.3% (mean = 97.3%). For quality control, all samples were analyzed as duplicates (acceptance criteria = 10% error), and an additional 10% of the samples were analyzed as blind replicates (acceptance criteria = 10% error). For further quality assurance, blanks (i.e., empty quartz boat) were analyzed every 10 samples to assess instrument accuracy and potential drift.

Toxic methylmercury (MeHg) typically accounts for the majority of total Hg in fish tissue (>95%; Grieb et al., 1990; Bloom, 1992). To ensure the accuracy of this approximation, a sub-sample of striped bass and tautog tissue (D_0 biopsy; n = 11 for each species) were analyzed for MeHg (and inorganic Hg) concentration by isotope dilution gas chromatography inductively coupled plasma mass spectrometry (GC–ICP-MS) at the Trace Element Analysis Laboratory, Dartmouth College (Hanover, New Hampshire, USA). Fish