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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)

Running head: Mercury in striped bass and tautog

Key words: bioaccumulation, food web, mercury, Morone saxatilis, stable isotope, striped bass, tautog, Tautoga onitis, trophic transfer

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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 $(\mathrm{Hg})$ 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 $=0.03 \mathrm{mg} \mathrm{Hg} \cdot \mathrm{kg}^{\mathrm{wet}} \mathrm{wt}^{-1}$ ). 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 $=$ $0.02 \mathrm{mg} \mathrm{Hg} \cdot \mathrm{kg}$ wet $\mathrm{wt}^{-1}$ ). Despite differences in Hg bioaccumulation between target fish, the mean Hg concentration of tautog exceeded levels in striped bass ( 0.24 and $0.16 \mathrm{mg} \mathrm{Hg} \cdot \mathrm{kg}$ wet $\mathrm{wt}^{-1}$, 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 ( $>70.2 \mathrm{~cm} \mathrm{TL} ; n=4$ ) and $44.8 \%$ of tautog ( $>40.6 \mathrm{~cm} \mathrm{TL} ; ~ n=29$ ) had Hg levels beyond the U.S. EPA regulatory threshold of 0.3 mg Hg . kg wet $\mathrm{wt}^{-1}$. 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 (Moszcynski et al., 1995; 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; U.S. 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

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position of a species as a function of its time-integrated diet history (Michener and Schell, 1994). For example, nitrogen isotopic signatures $\left({ }^{15} \mathrm{~N} /{ }^{14} \mathrm{~N}\right)$ are effective at quantifying the trophic position of an organism because enrichment of the heavier isotope $\left({ }^{15} \mathrm{~N}\right)$ occurs incrementally across trophic levels at a constant rate (approx. 3-4\%; Michener and Schell, 1994). Conversely, carbon isotopic signatures $\left({ }^{13} \mathrm{C} /{ }^{12} \mathrm{C}\right)$ are consistent across trophic levels ( $<1 \%$ o change between primary producer and consumer; Fry and Sherr, 1984), but are 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 $\left(<400 \mathrm{~km}^{2}\right)$. 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 ( $\sim 20-30 \mathrm{yr}$; 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 \& 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 $-4^{\circ} \mathrm{C}$ 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, $\sim 2.5 \mathrm{~g}$ 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 ( $\mathrm{D}_{0}=1$ 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 ( $\mathrm{D}_{1-3}$ and $\mathrm{L}_{1-3}$;

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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 hr (Labconco FreeZone 4.5 L Benchtop Freeze-Dry System), and subsequently weighed (g dry weight), homogenized with a mortar and pestle, and stored at room temperature in clear borosilicate $40-\mathrm{mL}$ 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 ( $\sim 40 \mathrm{mg}$ ) was added to the mercury analyzer. The instrument has a detection limit of 0.01 ng Hg (typical working range $0.05-600 \mathrm{ng}$ ), and employs thermal decomposition, amalgamation, and atomic absorption spectrophotmetry (EPA Method 7473; U.S. 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 $(\mathrm{MeHg})$ 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 ( $\mathrm{D}_{0}$ 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 tissue (30-50 mg dry weight), previously freeze-dried and homogenized, was added to amber glass vials. Approximately 2.65 ng of aqueous $\mathrm{CH}_{3} \mathrm{Hg}^{201}$ was then added as the enriched isotope spike to the sample, followed by 10 ml of $4 \mathrm{~mol} \mathrm{l}^{-1} \mathrm{HNO}_{3}$ (Hintelmann and Nguyen, 2005). The entire sample was heated over night at $55^{\circ} \mathrm{C}$, after which a $50-\mu \mathrm{l}$ sub-sample of the extract was analyzed by purge and trap, thermal desorption GC-ICP-MS (Rodriguez Martin-Doimeadios et al., 2002). This process includes the ethylation and trapping of volatile Hg species on Tenax traps (Brooks Rand, Seattle, Washington, USA), followed by thermal desorption onto open column GC and Hg isotope detection by ICP-MS (Element2, Thermo-Finnegan, Bremen, Germany). The recovery of MeHg in TORT SRM was $94.8 \%$ (range $=91.5-96.2 \%$ ) and sample precision for duplicate runs was $99.2 \%($ range $=96.6-102.2 \%)$.

Total Hg levels measured by the DMA-80 Direct Mercury Analyzer were in good agreement with the isotope dilution ICP-MS method $(\mathrm{MeHg}+$ inorganic $\mathrm{Hg}=$ total Hg$)$ (Linear regression: $\left.R^{2}=0.902 ; p<0.0001 ; y=0.7659 x+0.0325\right)$. Total Hg was higher (10.7\%) in samples analyzed by the DMA-80 Direct Mercury Analyzer (mean $=0.255 \pm 0.032 \mathrm{mg} \mathrm{Hg} \cdot \mathrm{kg}$

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wet weight ${ }^{-1}$ ) relative to the ICP-MS method (mean $=0.227 \pm 0.026 \mathrm{mg} \mathrm{Hg} \cdot \mathrm{kg}$ wet weight ${ }^{-1}$ ). This difference was not statistically significant (one-way ANOVA: $F=0.16, d f=1,43, p=$ 0.693), however, and total Hg concentrations are hereafter reported for the DMA-80 Analyzer only.

### 2.4. Stable isotope analysis

Nitrogen and carbon isotope measurements of a sub-sample of freeze-dried target fish ( $\mathrm{D}_{0}$ biopsy; $n=18-20$ ) and bioavailable prey (whole-body; $n=18-20$ ) were performed by the Boston University Stable Isotope Laboratory (Boston, Massachusetts, USA). Samples of animal ( $\sim 1 \mathrm{mg}$ dry weight) were analyzed using automated continuous-flow isotope ratio mass spectrometry (Preston and Owens, 1983). Samples were combusted in a EuroVector Euro EA elemental analyzer (Eurovector, Milan, Italy), after which $\mathrm{N}_{2}$ and $\mathrm{CO}_{2}$ gases were separated on a GC column, passed through a reference gas box, and introduced to an GV Isoprime isotope ratio mass spectrometer (GV Instruments, Manchester, UK). Ratios of ${ }^{13} \mathrm{C} /{ }^{12} \mathrm{C}$ and ${ }^{15} \mathrm{~N} /{ }^{14} \mathrm{~N}$ were expressed as the relative per mil (\%o) difference between the samples and international standards (Vienna Peedee Belemnite carbonate, ${ }^{13} \mathrm{C}_{\mathrm{V}-\mathrm{PDB}}$, and atmospheric nitrogen, ${ }^{15} \mathrm{~N}_{\text {air }}$, respectively):

$$
\begin{equation*}
\delta X=\left(R_{\text {sample }} / R_{\text {standard }}-1\right) \times 1000(\% o) \tag{1}
\end{equation*}
$$

where, $X={ }^{13} \mathrm{C}$ or ${ }^{15} \mathrm{~N}$ and $R={ }^{13} \mathrm{C} /{ }^{12} \mathrm{C}$ or ${ }^{15} \mathrm{~N} /{ }^{14} \mathrm{~N}$. The sample isotope ratio was compared to a secondary gas standard, whose isotope ratio was calibrated to international standards ( $\mathrm{R}_{\text {standard }}$ ). For ${ }^{13} \mathrm{C}_{\text {V-PDB, }}$, the gas was calibrated against NBS 20 (Solenhofen limestone). For ${ }^{15} \mathrm{~N}_{\text {air }}$, the gas

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was calibrated against atmospheric $\mathrm{N}_{2}$ and ammonium sulfate standards (IAEA standards $\mathrm{N}-1$, $\mathrm{N}-2$, and N-3). All international standards were obtained from the National Bureau of Standards in Gaithersburg, Maryland, USA.

### 2.5. Statistical analyses

Prior to parametric statistical analysis, total Hg data were converted to wet weight using a wet/dry ratio measured for a sub-sample of striped bass, tautog, and bioavailable prey (Tables 1 and 2). Data were then $\log _{10}$-transformed or arc-sin square-root transformed (proportional data) to meet assumptions of normality and homogeneity of variance. Mean Hg concentrations among muscle biopsies taken from striped bass $(n=18)$ and tautog $(n=14)$ were statistically compared with independent one-way analysis of variance (ANOVA) models using biopsy location ( $\mathrm{D}_{0-3}$ and $\mathrm{L}_{1-3}$ biopsies; Fig. 2A) as a fixed factor. Note that the Hg content of a single muscle biopsy represents the average calculated for the left and right side of the fish. Interspecies differences in muscle mercury levels (total Hg and $\% \mathrm{MeHg}$ ), stomach content Hg levels (recovered prey), and isotopic signatures ( $\delta^{15} \mathrm{~N}$ and $\delta^{13} \mathrm{C}$ ) were analyzed with one-way ANOVA models using species (striped bass and tautog) as a fixed factor. The effects of target fish TL (cm) and age (yr) on total Hg concentration were analyzed with least-squares (non-linear) exponential regressions. The ages of individual striped bass and tautog were estimated from species-specific age-size relationships reported in the literature (Hostetter and Munroe, 1993; Nelson et al., 2006). To assess differences in Hg bioaccumulation rates between striped bass and tautog, two-way ANOVA models were used to test for homogeneity of slopes (i.e., interaction effects between size/age and species) of the linearized exponential regressions (semi-logarithmic transformation).

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The total Hg concentration and isotopic signature of bioavailable prey were analyzed with a one-way ANOVA models using prey species as a fixed factor. Mean differences in total Hg levels and isotopic signatures across 8 and 7 levels of prey, respectively, were contrasted with a Ryan-Einot-Gabriel-Welsch (Ryan's Q) multiple comparison test. Note that direct statistical comparisons of the Hg content and isotope values of target fish and bioavailable prey were not made because of the different tissues analyzed between the groups (i.e., dorsal muscle versus whole-body tissues, respectively) (Gray, 2002). Non-linear exponential regression analysis was used, however, to test for the effect of trophic position $\left(\delta^{15} \mathrm{~N}\right)$ and carbon sources $\left(\delta^{13} \mathrm{C}\right)$ on the transfer total Hg through the estuarine food web.

## 3. Results

### 3.1. Evaluation of mercury analysis methods

There was no significant difference in the mean total Hg concentration among muscle biopsies from an individual striped bass or tautog (one-way ANOVA; bass: $F=0.15, d f=6,125$, $p=0.989$; tautog: $F=0.12, d f=6,97, p=0.994$ ) (Fig. 2). The Hg concentration of tissue located above the operculum ( $\mathrm{D}_{0}$ biopsy; Fig. 2 A ) is therefore indicative of the contaminant present in the whole-body white muscle filet, and Hg levels in target fish are hereafter reported for the $\mathrm{D}_{0}$ biopsy only. Mercury speciation analysis also revealed that MeHg accounts for $>98 \%$ (range $=$ 96.3-99.6\%) of the total Hg present in the muscle tissue of striped bass and tautog (Table 1). To this end, measurements of total Hg are a suitable approximation of MeHg in the white muscle tissue of target fish.

### 3.2. Mercury analysis of target fish

Total Hg levels in the dorsal muscle tissue of target fish $(n=119)$ ranged from 0.03 to $0.58 \mathrm{mg} \mathrm{Hg} \cdot \mathrm{kg}^{2}$ wet $\mathrm{wt}^{-1}$ (Table 1). Of these samples, the mean total Hg concentration of tautog was $33.3 \%$ greater than values measured in striped bass (Table 1). To account for differences in age-at-catch, total Hg data were standardized by the estimated age of each fish. Accordingly, age-normalized Hg levels of striped bass were $47.4 \%$ higher than concentrations measured in tautog.

There was a significant positive correlation between total Hg concentration and TL for both target fish (Exponential regression; bass: $F=50.35, d f=1,65, p<0.0001$; tautog: $F=$ 86.28, $d f=1,52, p<0.0001$ ) (Fig. 3A). However, the slope of the linearized exponential regression model for tautog $(\beta=0.022)$ was greater than striped bass $(\beta=0.013)$ (two-way ANOVA; species $\times$ length: $F=7.98, d f=3,118, p<0.01$ ), indicating that tautog have higher Hg concentrations at a given body length. Total Hg concentration also increased significantly with fish age, irrespective of species (Exponential regression; bass: $F=56.15, d f=1,65, p<0.0001$; tautog: $F=82.13, d f=1,52, p<0.0001$ ) (Fig. 3B). In contrast to the mercury-length relationship, however, Hg bioaccumulation rates as a function of age were significantly faster in striped bass than tautog ( $\beta=0.133$ and 0.065 , respectively) (two-way ANOVA; species $\times$ age: $F$ $=13.17, d f=3,118, p<0.0005)$.

Of the target fish analyzed in this study, $12.1 \%$ of striped bass and $30.2 \%$ of tautog exceeded the U.S. Environmental Protection Agency advisory level of $0.3 \mathrm{mg} \mathrm{MeHg} \cdot \mathrm{kg}$ wet $w t^{-1}$ (U.S. EPA, 2006). Data were also analyzed with respect to legal-size limits for each
species, i.e., minimum catch size for striped bass and tautog in Rhode Island equal 70.2 and 40.6 cm , respectively (RIDEM, 2008). The mean total Hg concentration of legal-size striped bass was $0.342 \pm 0.040 \mathrm{mg} \mathrm{Hg} \cdot \mathrm{kg}^{2}$ wet $\mathrm{wt}^{-1}(n=4)$, of which $75.0 \%$ exceeded the U.S. EPA advisory level. For legal-size tautog, the mean total Hg concentration was $0.319 \pm 0.027 \mathrm{mg} \mathrm{Hg} \cdot \mathrm{kg}$ wet $\mathrm{wt}^{-1}(n=29)$, and $44.8 \%$ of these fish had Hg levels above $0.3 \mathrm{mg} \mathrm{MeHg} \cdot \mathrm{kg}$ wet $\mathrm{wt}^{-1}$. The mercury-length exponential regression model also predicted that striped bass and tautog attain Hg concentrations equal to the U.S. EPA advisory level near their minimum legal catch size (75.7 and 48.6 cm TL, respectively; Fig. 3A).

### 3.3. Stomach content analysis

The stomach contents of striped bass and tautog revealed interspecies differences in diet (Table 3). The results suggest that striped bass are opportunistic foragers, as bass consumed 10 different recovered prey species. Teleost fish and decapod crustaceans (sand shrimp and green crab) comprised the largest fraction of the striped bass diet. Collectively, these prey items were identified in $29.4 \%$ of the stomachs analyzed. Only 5 novel prey taxa were positively identified in tautog stomachs (Table 3); hence, relative to bass, tautog demonstrate a selective foraging strategy. The blue mussel and decapod crabs (green, mud, and unidentified crabs) were the preferred prey of tautog, occurring in $43.0 \%$ of the stomachs.

### 3.4. Mercury analysis of prey

The principal prey of striped bass and tautog, as determined by dietary analysis, were collected from the Narragansett Bay and subsequently analyzed for whole-body total Hg . Mean total Hg levels of these bioavailable prey varied significantly as a function of prey-type (one-way ANOVA; $F=29.70, d f=7,835, p<0.0001$ ) (Table 2). With the exception of the Atlantic menhaden, prey that were exclusive to striped bass (anchovy, herring, scup, and sand shrimp) had significantly higher Hg concentrations than the prey of tautog (blue mussel and mud crab) (Ryan's Q multiple comparison test) (Fig. 4). On average, the total Hg concentration of tautog prey was $32.0 \%$ lower than the diet items of striped bass. The mean total Hg concentration of prey removed from tautog stomachs was also $13.2 \%$ lower than the recovered prey of striped bass, but this difference was not significant at $p<0.05$ (Table 1).

### 3.5. Stable isotope analysis

Stable nitrogen $\left(\delta^{15} \mathrm{~N}\right)$ isotope signatures were used to delineate the trophic positioning of target fish and bioavailable prey. Mean $\delta^{15} \mathrm{~N}$ values varied considerably among taxonomic groups (Tables 1 and 2; Fig. 5). Striped bass had a significantly higher $\delta^{15} \mathrm{~N}$ value relative to tautog, and both target fish had higher $\delta^{15} \mathrm{~N}$ values than their preferred prey. Among prey, $\delta^{15} \mathrm{~N}$ values varied significantly across species (one-way ANOVA; $F=57.99, d f=6,136, p<0.0001$ ). Specifically, significant ${ }^{15} \mathrm{~N}$ enrichment was observed in scup and bay anchovy, whereas the most depleted $\delta^{15} \mathrm{~N}$ values were calculated for Atlantic menhaden and blue mussel (Table 2; Fig. 5) (Ryan's Q multiple comparison test).

The $\delta^{15} \mathrm{~N}$ value of the blue mussel was used to assign distinct trophic levels for each consumer species analyzed in this study (modified from Hobson et al., 2002).

Trophic level $=2+\frac{\left(\delta^{15} N_{\text {consumer }}-\delta^{15} N_{\text {mussel }}\right)}{3.4}$
where, " 2 " is the assumed trophic level of the blue mussel, a phytoplanktivore (Newell, 1989), $\delta^{15} \mathrm{~N}_{\text {mussel }}$ and $\delta^{15} \mathrm{~N}_{\text {consumer }}$ are the respective nitrogen isotope signatures of blue mussel ( $10.62 \%$; this study) and the consumer species of interest, and " 3.4 " is the constant nitrogen isotope enrichment (\%o) per trophic level (Post, 2002). Accordingly, striped bass and tautog were designated a trophic level of $4.07 \pm 0.08$ and $3.51 \pm 0.03$, respectively, whereas prey species (excluding mussels) occupied trophic levels between 2.43 and 3.26 (Tables 1 and 2).

Stable carbon $\left(\delta^{13} \mathrm{C}\right)$ isotope signatures were used to determine the varying sources of primary production to the estuarine food web (e.g., benthic algae versus phytoplankton), and thus, distinguish between benthic and pelagic trophic linkages (Peterson and Howarth, 1987; France, 1995). Measured $\delta^{13} \mathrm{C}$ values ranged from $-20.16 \pm 0.17 \%$ o to $-15.13 \pm 0.25 \%_{o}$ and varied significantly between target fish (Table 1) and among prey (one-way ANOVA; $F=40.66$, $d f=6,136, p<0.0001$ ) (Table 2). Moreover, two distinct isotopic groups were demarcated by a $\delta^{13} \mathrm{C}$ value of approximately $-18 \%$ (Fig. 5). Relative to this demarcation point, more depleted $\delta^{13} \mathrm{C}$ values indicate a phytoplankton-based (pelagic) food web, and in this study were represented by the striped bass, planktivorous forage fish, and blue mussels. Conversely, $\delta{ }^{13} \mathrm{C}$ signatures greater than $-18 \%$ (i.e., more enriched) suggest benthic sources of primary production and were characteristic of tautog, scup, and decapod crustaceans.

Stable isotope analysis was coupled with total Hg data to provide insight into the effects of trophic structure on species contamination. There was a significant positive correlation between mean total Hg concentration and $\delta^{15} \mathrm{~N}$ values measured for target fish and bioavailable
prey (Exponential regression; $F=38.79, d f=1,8, p<0.0005$ ) (Fig. 6); hence verifying an increase in Hg contamination at higher trophic levels in the estuarine food web. Conversely, no significant relationship was observed between mean total Hg concentration and $\delta^{13} \mathrm{C}$ values measured across all species (Exponential regression; $F=0.20, d f=1,7, p=0.670$ ).

## 4. Discussion

### 4.1. Mercury analysis of target fish

This study provides information on Hg levels in the muscle tissue of striped bass and tautog collected from the Narragansett Bay, and also examines the effect of several biotic factors on interspecies Hg bioaccumulation patterns. A significant positive correlation between Hg concentration and target fish age (and size) was observed for striped bass and tautog, and similar patterns have been previously documented for each species (Hammerschmidt and Fitzgerald, 2006a; Mason et al., 2006). The Hg -age relationship confirms that striped bass and tautog effectively bioaccumulate Hg , which is further explained by a low rate of Hg depuration relative to the rapid accumulation of Hg in fish muscle tissue (Clarkson, 1992; Trudel and Rasmussen, 1997). It is well established that dietary uptake is the dominant source of Hg in fish (Hall et al., 1997); however, striped bass bioaccumulate Hg at an accelerated rate relative to tautog. For example, the slope of the linearized exponential (Hg-age) regression provides an estimate of the Hg bioaccumulation rate for a given species. The comparison of these slope values between the target fish predicts that striped bass $(\beta=0.133)$ accumulate Hg in muscle tissue twice as fast as tautog $(\beta=0.065)$. As discussed in the subsequent section, differences in diet and trophic
positioning in the estuarine food web likely account for the observed patterns of Hg bioaccumulation in the target fish (Weiner et al., 2003; Bank et al., 2007).

Although striped bass accumulate Hg more rapidly than tautog, results from this study indicate that the mean Hg concentration of tautog exceeded levels in bass. This apparent discrepancy is attributed to the greater age-at-catch of tautog (Cooper, 1967), and thus, protracted period in which this species is exposed to Hg . Specifically, the average age of tautog in this study was 2.6 times greater than striped bass; hence, at a given body length, tautog had a higher Hg tissue concentration than bass. A similar explanation was provided by Hammerschmidt and Fitzgerald (2006a) to reconcile the difference in Hg concentrations observed in tautog and bluefish (Pomatomus saltatrix), an apex piscivore, from the Long Island Sound. To this end, the authors recommended that fish longevity be considered when assessing their potential risk to human consumers (Hammerschmidt and Fitzgerald, 2006a).

In this investigation, Hg concentrations of tautog were comparable to values observed in conspecifics from other coastal ecosystems. Hammerschmidt and Fitzgerald (2006a), using cold-vapor atomic fluorescence spectrometry (CVAFS), analyzed the mercury content of tautog collected from the Long Island Sound (mean TL $=41.4 \mathrm{~cm} ; n=32$ ), and reported an average MeHg concentration of $0.19 \mathrm{mg} \mathrm{MeHg} \cdot \mathrm{kg}$ wet $\mathrm{wt}^{-1}$ for axial muscle tissue. Moreover, for tautog collected from the New York Bight Apex (mean TL $=31.0 \mathrm{~cm} ; n=14$ ), Deshpande et al. (2000) estimated a mean total Hg concentration of $0.08 \mathrm{mg} \mathrm{Hg} \cdot \mathrm{kg}^{\mathrm{m}}$ wet $\mathrm{wt}^{-1}$ for white muscle tissue. Using the exponential regression presented in this study, tautog of comparable sizes (41.4 and 31.0 cm ) from the Narragansett Bay had projected Hg values of 0.21 and $0.12 \mathrm{mg} \mathrm{Hg} \cdot \mathrm{kg}$ wet $\mathrm{wt}^{-1}$, respectively.

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In contrast to tautog, geographic variability in Hg contamination was apparent in striped bass. In the Chesapeake Bay, for example, the dorsal muscle tissue of striped bass (mean TL ~ $66 \mathrm{~cm} ; n=70$ ) had a mean MeHg concentration of $0.12 \mathrm{mg} \mathrm{MeHg} \cdot \mathrm{kg}$ wet $\mathrm{wt}^{-1}$ (analyzed using CVAFS; Mason et al., 2006). This level of contamination is considerably lower then the estimated MeHg content of striped bass from the Narragansett Bay that are of the same TL ( 0.22 $\mathrm{mg} \mathrm{MeHg} \cdot \mathrm{kg}$ wet $\mathrm{wt}^{-1}$; using Hg -size relationship and $\% \mathrm{MeHg}$ data). It is also noteworthy that the mean proportional contribution of MeHg to total Hg in striped bass from the Chesapeake Bay (65\%; Mason et al., 2006) was lower than the value reported in this study $(98.7 \%)$.

Spatial variations in the Hg burden of marine and estuarine fish are poorly understood, but are presumably a function of geographic differences in the leyel of Hg input to the ecosystem and biogeochemical conditions that affect MeHg production and mobilization (Rolfhus and Fitzgerald, 1995; Benoit et al., 2003; Kraepiel et al., 2003; Hammerschmidt and Fitzgerald, 2006b). Moreover, spatially-explicit physicochemical variables and predator-prey interactions influence the initial incorporation of Hg into the food web and its subsequent transfer through trophic assemblages (Lindqvist et al., 1991; Watras and Bloom, 1992; Mason et al., 1996; Bloom et al., 1999; Sveinsdottir and Mason, 2005). The assessment of spatial differences in fish Hg levels, however, are complicated for those species that exhibit large-scale seasonal migrations and ontogenetic shifts in habitat use and diet, as reported for striped bass (Harding and Mann, 2003; Nelson et al., 2003, 2006; Secor and Piccoli, 2007).

### 4.2. Diet and trophic effects on Hg bioaccumulation

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Visual analysis of the stomach contents of tautog and striped bass revealed differences in their feeding strategy and diet composition. Tautog exhibited a strict durophagous foraging strategy, and therefore fed exclusively on hard-bodied epibenthic and encrusting invertebrates. Macrocrustaceans were also numerically dominant prey for striped bass, although dietary overlap with tautog was minimal. Further, striped bass were piscivorous, with teleosts accounting for a large proportion of the identified diet. These results are consistent with previous analyses of the food habits of tautog and striped bass collected from other coastal Atlantic regions, including the Chesapeake Bay (Hildebrand and Schroeder, 1928; Hartman and Brandt, 1995; Harding and Mann, 2003; Walter and Austin, 2003), Delaware Bay (Steimle and Shaheen, 1999), New YorkNew Jersey estuarine waters (Olla et al., 1974; Festa, 1979; Steimle and Ogren, 1982), and Massachusetts coastal waters (Nelson et al., 2003, 2006).

Measurements of interspecies $\delta^{13} \mathrm{C}$ signatures corroborated the findings of the conventional dietary analysis of target fish. Specifically, tautog had a $\delta^{13} \mathrm{C}$ signature consistent with a benthic carbon source and presumably derived from the consumption of epibenthic crustaceans. It was evident, however, that tautog maintained a lower $\delta^{13} \mathrm{C}$ value than its principal crustacean prey, the green crab. This discrepancy is attributed to the substantial contribution of the blue mussel, a phytoplanktivore (Newell, 1989), to the diet of tautog. In contrast to tautog, striped bass had a $\delta^{13} \mathrm{C}$ signature characteristic of a pelagic carbon source, which in turn resulted from bass feeding on forage fish. The slightly more enriched $\delta^{13} \mathrm{C}$ signature of striped bass relative to planktivorous forage fish, however, may reflect the additional contribution of decapod crustaceans to the diet of bass.

The diet and trophic ecology of striped bass and tautog are the main factors impacting their respective Hg bioaccumulation rates. In this study, species-specific $\delta^{15} \mathrm{~N}$ signatures
indicate that striped bass occupy a higher trophic position than tautog (trophic level $\sim 4.1$ and 3.5, respectively). The variation in trophic status between target fish is adequately explained by the trophic positioning of their prey. Striped bass, for example, feed on relatively high trophic level prey (forage fish and decapod crustaceans; mean trophic level $=2.86$ ), whereas tautog consume prey that occupy lower trophic levels (mussel and green crab; mean trophic level $=$ 2.33). The analysis of Hg levels in bioavailable prey further revealed that striped bass feed on Hg -enriched prey relative to the dietary items of tautog (mean Hg content of prey $=0.03$ and $0.02 \mathrm{mg} \mathrm{Hg} \cdot \mathrm{kg}$ wet $\mathrm{wt}^{-1}$, respectively). The Atlantic menhaden, an important food resource of striped bass (Walter and Austin, 2003; Nelson et al., 2006; this study), was an exception to this observed pattern. The low Hg content of menhaden was attributed to this species occupying a low trophic position in the estuary ( $\sim 2.4$ ). This is further supported by a depleted $\delta^{13} \mathrm{C}$ signature in menhaden, which implies that this species consumes phytoplankton (e.g., pelagic diatoms; Collette and Klein-MacPhee, 2002).

The positive correlation between Hg levels and $\delta^{15} \mathrm{~N}$ signatures of sampled biota indicates that Hg is trophically transferred through the estuarine food web, as reported in other aquatic systems (Jarman et al., 1996; Bowles et al., 2001; Power et al., 2002; Bank et al., 2007; Cai et al., 2007). Precaution must be exercised in this study, however, when evaluating the rate of Hg increase across trophic levels. This is particularly warranted for direct comparisons between target fish and bioavailable prey because different tissues were analyzed (dorsal muscle and whole-body tissues, respectively). Previous studies have demonstrated that certain body tissues (e.g., muscle, liver) disproportionately concentrate Hg , and thus, have higher Hg levels than whole-body samples (Gray, 2002; Peterson et al., 2005). If applicable to this investigation, then the muscle tissue of target fish is expected to have modestly inflated Hg levels relative to

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the entire body. Notwithstanding this limitation, the cumulative data presented herein offers compelling evidence that time-integrated diet history and trophic relationships are the dominant factors impacting Hg bioaccumulation in the Narragansett Bay estuary.

### 4.3. Human health perspective

Cursory examination of this study's full data set indicates that Hg concentrations in target fish are relatively low, i.e., only $12.1 \%$ of striped bass and $30.2 \%$ of tautog exceeded the U.S. EPA criterion of $0.3 \mathrm{mg} \mathrm{MeHg} \cdot \mathrm{kg}$ wet $\mathrm{wt}^{-1}$ (U.S. EPA, 2006). To accurately assess the possible human health risks associated with eating fish, however, contaminant data must be analyzed relative to state-imposed legal-size limits for target species. In this study, reevaluating the Hg data for only legal-size fish revealed that $75.0 \%$ of striped bass and $44.8 \%$ of tautog accumulated Hg levels beyond the U.S. EPA regulatory threshold. Moreover, the mercury-length exponential model presented in this study suggests that striped bass and tautog obtain Hg levels of 0.3 mg Hg - kg wet $\mathrm{wt}^{-1}$ near their minimum legal catch size ( 75.7 and 48.6 cm TL , respectively). It is important to note, however, that the low sample size of legal-size target fish, particularly for striped bass $(n=4)$, requires that additional studies be conducted on the Hg contamination of these edible species. Nevertheless, the results presented in this study indicate that frequent consumption of striped bass and tautog could pose a human health risk, and thus, justify their inclusion in state-specific consumption advisories. Moreover, this exercise underscores the importance of coupling species-specific Hg data with state-imposed minimum catch requirements; hence, focusing on fishery resources that represent Hg exposure pathways to human consumers.

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## Figure captions

Fig. 1. Map of the Narragansett Bay (Rhode Island, USA) with points denoting collection sites of striped bass (Morone saxatilis), tautog (Tautoga onitis), and bioavailable prey (forage fish and invertebrates).

Fig. 2. (A) Sub-sampling locations in the dorsolateral muscle tissue of striped bass (Morone saxatilis) and tautog (Tautoga onitis) (top and bottom fish image, respectively). (B) Relationship between the total mercury concentration $\left(\mathrm{Hg} ; \mathrm{mg} \mathrm{Hg} \cdot \mathrm{kg}\right.$ wet weight ${ }^{-1}$ ) of a multiple biopsies removed from the muscle tissue of striped bass $(n=18)$ and tautog $(n=14)$. Each muscle biopsy was excised from the dorsal and lateral tissue along the anterior-posterior axis and represents the mean ( $\pm$ standard error) Hg concentration of the biopsy removed from the left and right side of each fish.

Fig. 3. Total mercury concentration ( $\mathrm{Hg} ; \mathrm{mg} \mathrm{Hg} \cdot \mathrm{kg}$ wet weight ${ }^{-1}$ ) of the dorsal muscle tissue of striped bass $(n=66)$ and tautog $(n=53)$ as a function of fish total length, $T L$ (A) and age, $a(\mathrm{~B})$. Exponential models were fit to the data and the equations are presented.

Fig. 4. Mean total mercury concentration $\left(\mathrm{Hg} ; \mathrm{mg} \mathrm{Hg} \cdot \mathrm{kg}\right.$ wet weight $\left.{ }^{-1}\right)\left(+\right.$ standard error) of $^{\text {a }}$ prey collected from the Narragansett Bay, i.e., "bioavailable prey". Prey include bay anchovy (Anchoa mitchilli; $n=39$ ), scup (Stenotomus chrysops; $n=56$ ), green crab (Carcinus maenas; $n$ $=110$ ), river herring (Alosa spp.; $n=183$ ), sand shrimp (Crangon septemspinosa; $n=241$ ),
back-finger mud crab (Panopeus herbstii; $n=15$ ), Atlantic menhaden (Brevoortia tyrannus; $n=$ 32), and blue mussel (Mytilus edulis; $n=160$ ).

Fig. 5. Stable nitrogen $\left(\delta^{15} \mathrm{~N}\right)$ and carbon $\left(\delta^{13} \mathrm{C}\right)$ isotope signatures (mean $\pm$ standard error) of striped bass (Morone saxatilis), tautog (Tautoga onitis), and their prey ( $n=18-20$ per species). Prey species include river herring (Alosa spp.), scup (Stenotomus chrysops), bay anchovy (Anchoa mitchilli), Atlantic menhaden (Brevoortia tyrannus), green crab (Carcinus maenas), sand shrimp (Crangon septemspinosa), and blue mussel (Mytilus edulis). Ovals represent organisms that derive their carbon from pelagic or benthic sources, as determined by interspecies $\delta^{13} \mathrm{C}$ signatures.

Fig. 6. Relationship between total mercury concentration $\left(\mathrm{Hg} ; \mathrm{mg} \mathrm{Hg} \cdot \mathrm{kg}\right.$ wet weight $\left.{ }^{-1}\right)$ and stable nitrogen $\left(\delta^{15} \mathrm{~N}\right)$ isotope signatures (mean $\pm$ standard error) of striped bass (Morone saxatilis), tautog (Tautoga onitis), and their prey ( $n=18-20$ ). Prey species include river herring (Alosa spp.), scup (Stenotomus chrysops), bay anchovy (Anchoa mitchilli), Atlantic menhaden (Brevoortia tyrannus), green crab (Carcinus maenas), sand shrimp (Crangon septemspinosa), and blue mussel (Mytilus edulis).


| Parameter | Striped bass |  |  | Tautog |  |  | $F$ | df | $p$ |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
|  | $n$ | Mean $\pm$ SE | Range | $n$ | Mean $\pm$ SE | Range |  |  |  |
| Total length ( cm$)^{\text {a }}$ | 66 | $49.2 \pm 1.8$ | 26.2-102.0 | 53 | $39.3 \pm 1$. | 10.7-56.4 | 16.84 | 118 | < 0.0001 |
| Wet wt (kg) ${ }^{\text {a }}$ | 66 | $1.40 \pm 0.14$ | 0.18-6.82 | 53 | $1.61 \pm 0.16$ | 0.10-3.88 | 0.01 | 118 | 0.918 |
| Water content (\%) ${ }^{\text {b }}$ | 42 | $77.2 \pm 0.7$ | 50.4-79.3 | 45 | $78.6 \pm 0.5$ | 63.6-87.7 | 3.44 | 86 | 0.067 |
| Age (yr) ${ }^{\text {a,c }}$ | 66 | $4.3 \pm 0.2$ | 1.7-9.6 | 53 | $11.3 \pm 0.6$ | 3.6-18.6 | 168.18 | 118 | < 0.0001 |
| $\mathrm{Hg}\left(\mathrm{mg} \cdot \mathrm{kg} \text { wet } \mathrm{wt}^{-1}\right)^{\text {a }}$ | 66 | $0.16 \pm 0.01$ | 0.03-0.45 | 53 | $0.24 \pm 0.02$ | 0.03-0.58 | 6.79 | 118 | $<0.05$ |
| Age-normalized $\mathrm{Hg}^{\text {a,d }}$ | 66 | $0.038 \pm 0.003$ | 0.012-0.159 | 53 | $0.020 \pm 0.001$ | 0.005-0.046 | 39.47 | 118 | < 0.0001 |
| $\% \mathrm{MeHg}^{\text {b,e }}$ | 11 | $98.7 \pm 0.3$ | 96.3-99.6 | 11 | $98.0 \pm 0.2$ | 96.8-98.9 | 5.57 | 21 | $<0.05$ |
| Recovered prey Hg ${ }^{\text {a,f }}$ | 54 | $0.060 \pm 0.005$ | 0.016-0.190 | 50 | $0.053 \pm 0.005$ | 0.010-0.176 | 1.91 | 103 | 0.170 |
| $\delta^{15} \mathrm{~N}(\%)$ | 20 | $17.66 \pm 0.27$ | 15.67-19.62 | 18 | $15.76 \pm 0.12$ | 14.94-16.80 | 38.92 | 37 | < 0.0001 |
| $\delta^{13} \mathrm{C}(\%)$ | 20 | $-18.27 \pm 0.42$ | -24.08--15.99 | 18 | $-16.63 \pm 0.16$ | -18.36--15.67 | 12.21 | 37 | $<0.005$ |
| Trophic level ${ }^{\text {g }}$ | 20 | $4.07 \pm 0.08$ | 3.49-4.65 | 18 | $3.51 \pm 0.03$ | 3.27-3.82 | 39.07 | 37 | < 0.0001 |

Table 1
Comparison of total length, whole-body wet weight, muscle tissue water content, age, mercury levels [total mercury ( Hg ) and percent methylmercury (\% MeHg)], stable nitrogen $\left(\delta^{15} \mathrm{~N}\right)$ and carbon $\left(\delta^{13} \mathrm{C}\right)$ isotope signatures, and trophic level status of striped bass (Morone saxatilis) and tautog (Tautoga onitis) collected from the Narragansett Bay. Statistical summaries are from one-way analysis of variance models using species (striped bass and tautog) as a fixed factor. Sample sizes ( $n$ ), mean values $[ \pm$ standard error (SE)], and ranges are also presented.

[^0]Table 2
Comparison of length, whole-body wet weight, whole-body water content, stable nitrogen $\left(\delta^{15} \mathrm{~N}\right)$ and carbon $\left(\delta^{13} \mathrm{C}\right)$ isotope signatures, and total mercury ( Hg ) in prey species collected from the Narragansett Bay. Sample sizes ( $n$ ), mean values ( $\pm$ standard error), and ranges (in parentheses) are presented.

| Species | Length <br> $(\mathrm{cm})^{\mathrm{a}}$ | Wet weight <br> $(\mathrm{g})$ | Water content <br> $(\%)$ | $\delta^{15} \mathrm{~N}$ <br> $(\% o)^{\mathrm{b}}$ | $\delta^{13} \mathrm{C}$ <br> $(\% o)^{\mathrm{b}}$ | Trophic level ${ }^{\mathrm{c}}$ |
| :--- | :---: | :---: | :---: | :---: | :---: | :---: |

Division Teleostei

| Atlantic menhaden | $4.2 \pm 0.2$ | $0.72 \pm 0.13$ | $79.1 \pm 1.4$ | $12.08 \pm 0.13$ | $-18.77 \pm 0.11$ | $2.43 \pm 0.04$ | $0.016 \pm 0.002$ |
| :--- | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
| $(n=32)$ | $(3.0-7.5)$ | $(0.14-3.77)$ | $(71.6-83.7)$ | $(11.03-13.71)$ | $(-20.03--17.96)$ | $(2.12-2.91)$ | $(0.006-0.047)$ |
|  | $7.3 \pm 0.1$ | $2.72 \pm 0.12$ | $77.2 \pm 0.5$ | $14.39 \pm 0.13$ | $-18.34 \pm 0.11$ | $3.11 \pm 0.04$ | $0.035 \pm 0.001$ |
| Bay Anchovy <br> $(n=39)$ | $(5.3-8.2)$ | $(0.98-4.01)$ | $(74.2-79.1)$ | $(13.72-15.74)$ | $(-19.23--16.99)$ | $(2.91-3.51)$ | $(0.029-0.055)$ |
|  |  |  |  |  |  |  |  |
| River herring | $6.2 \pm 0.2$ | $2.53 \pm 0.16$ | $78.1 \pm 0.3$ | $13.42 \pm 0.19$ | $-19.73 \pm 0.47$ | $2.82 \pm 0.06$ | $0.027 \pm 0.001$ |
| $(n=183)$ | $(2.9-11.2)$ | $(0.08-11.15)$ | $(71.0-83.7)$ | $(12.31-15.47)$ | $(-25.67--17.04)$ | $(2.50-3.43)$ | $(0.008-0.188)$ |
|  |  |  |  |  | $-17.09 \pm 0.22$ | $3.26 \pm 0.07$ | $0.033 \pm 0.002$ |
| Scup | $10.3 \pm 0.5$ | $23.75 \pm 2.24$ | $75.6 \pm 0.3$ | $14.91 \pm 0.22$ | $(-18.12--13.61)$ | $(2.66-3.68)$ | $(0.015-0.066)$ |

Class Bivalvia

| Blue mussel $(n=160)^{\mathrm{d}}$ | $\begin{aligned} & 3.9 \pm 0.1 \\ & (1.8-8.2) \end{aligned}$ | $\begin{gathered} 7.20 \pm 0.46 \\ (1.07-38.66) \end{gathered}$ | $\begin{gathered} 83.0 \pm 0.3 \\ (72.1-38.66) \end{gathered}$ | $\begin{gathered} 10.62 \pm 0.20 \\ (9.15-92.7) \end{gathered}$ | $\begin{gathered} -20.16 \pm 0.17 \\ (-22.12--19.27) \end{gathered}$ | $\begin{aligned} & 2.00 \pm 0.06 \\ & (1.57-2.68) \end{aligned}$ | $\begin{aligned} & 0.015 \pm 0.006 \\ & (0.005-0.040) \end{aligned}$ |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
| S Crustacea |  |  |  |  |  |  |  |
| Green crab $(n=110)$ | $\begin{aligned} & 4.2 \pm 0.2 \\ & (1.2-7.8) \end{aligned}$ | $\begin{gathered} 24.21 \pm 2.62 \\ (0.32-104.28) \end{gathered}$ | $\begin{gathered} 73.4 \pm 1.0 \\ (61.3-89.8) \end{gathered}$ | $\begin{gathered} 12.90 \pm 0.20 \\ (11.10-14.48) \end{gathered}$ | $\begin{gathered} -15.13 \pm 0.25 \\ (-17.15--13.25) \end{gathered}$ | $\begin{gathered} 2.67 \pm 0.06 \\ (11.10-14.48) \end{gathered}$ | $\begin{aligned} & 0.032 \pm 0.003 \\ & (0.006-0.156) \end{aligned}$ |
| Mud crab $(n=15)$ | $\begin{aligned} & 1.7 \pm 0.2 \\ & (0.9-2.6) \end{aligned}$ | $\begin{aligned} & 2.58 \pm 0.61 \\ & (0.20-7.37) \end{aligned}$ | $\begin{gathered} 65.1 \pm 3.3 \\ (54.5-82.8) \end{gathered}$ | - | - | - | $\begin{aligned} & 0.016 \pm 0.001 \\ & (0.006-0.025) \end{aligned}$ |
| Sand shrimp $(n=241)$ | $\begin{aligned} & 3.8 \pm 0.1 \\ & (0.5-6.2) \end{aligned}$ | $\begin{aligned} & 0.49 \pm 0.02 \\ & (0.08-1.94) \end{aligned}$ | $\begin{gathered} 78.5 \pm 0.4 \\ (65.4-85.9) \end{gathered}$ | $\begin{gathered} 13.52 \pm 0.23 \\ (11.51-15.13) \end{gathered}$ | $\begin{gathered} -15.67 \pm 0.55 \\ (-21.09--12.47) \end{gathered}$ | $\begin{aligned} & 2.85 \pm 0.07 \\ & (2.26-3.33) \end{aligned}$ | $\begin{aligned} & 0.020 \pm 0.001 \\ & (0.006-0.123) \end{aligned}$ |

758
$759{ }^{\text {a }}$ Length measured as total length for fish and shrimp, carapace width for crabs, and shell height for mussels.
$760 \quad{ }^{\mathrm{b}}$ Sample size for stable nitrogen and carbon isotope analysis was $18-20$ for all species.
${ }^{\mathrm{c}}$ Trophic levels of individual prey species were calculated using Equation 2.
762

## ${ }^{d}$ Mussel wet weight, water content, $\delta^{15} \mathrm{~N}, \delta^{13} \mathrm{C}$, and Hg were measured for soft tissue only (shell removed).

763
764
765 766

Table 3
Stomach contents of striped bass (Morone saxatilis) and tautog (Tautoga onitis) collected from the Narragansett Bay. Values represent the percent of striped bass or tautog stomachs that contained a given prey item.

| Recovered prey |  | Target fish |  |
| :---: | :---: | :---: | :---: |
| Species | Common name | Striped bass | Tautog |
| Division Teleostei |  |  |  |
| Alosa spp. | Herring | 2.5 | 0 |
| Anchoa mitchilli | Bay anchovy | 2.5 |  |
| Brevoortia tyrannus | Atlantic menhaden | 1.3 | 0 |
| Gobiosoma bosc | Naked goby |  | 0 |
| Peprilus triacanthus | Butterfish | 1.3 | 0 |
| Stenotomus chrysops | Scup | 1.3 | 0 |
| Syngnathus fuscus | Northern pipefish | 1.3 | 0 |
| Class Bivalvia |  |  |  |
| Mytilus edulis | Blue mussel | 0 | 19.1 |
| Class Crustacea |  |  |  |
| Carcinus maenas | Green crab | 5.1 | 1.6 |
| Crangon septemspinosa | Sand shrimp | 9.0 | 0 |
| Panopeus herbstii | Black-finger mud crab | 0 | 4.8 |
| - | Unidentified crab | 2.6 | 17.5 |

Class Echinoidea

Strongylocentrotus droebachiensis Green sea urchin 0
1.6

## Class Gastropoda

## Crepidula fornicata

- 

Atlantic slipper snail

0
3.2

Unidentified gastropod
0
Class Ulvophyceae

| Ulva lactuca | Sea lettuce | 1.3 | 0 |
| :--- | :---: | :---: | :---: |
| Unidentified contents | 50.6 | 49.2 |  |
| Empty stomachs | 17.7 | 3.2 |  |



Figure 2


Figure 3


Figure 4

## ACCEPTED MANUSCRIPT



Figure 5


Figure 6


[^0]:    ${ }^{\text {a }}$ Data were $\log _{10}$-transformed prior to statistical analysis, but raw data values are presented in the table.
    ${ }^{\mathrm{b}}$ Data were arc-sin square-root transformed prior to statistical analysis, but raw data values are presented in the table.
    ${ }^{\mathrm{c}}$ Ages for individual fish were predicted from published age-size relationships (Hostetter and Munroe, 1993; Nelson et al., 2006).
    ${ }^{\text {d }}$ Age-normalized $\mathrm{Hg}=\mathrm{Hg}\left(\mathrm{mg} \mathrm{Hg} \cdot \mathrm{kg}\right.$ tissue wet $\left.\mathrm{wt}^{-1}\right) /$ age (yr).
    ${ }^{\mathrm{e}} \% \mathrm{MeHg}=\left[\mathrm{MeHg}\left(\mathrm{mg} \mathrm{MeHg} \cdot \mathrm{kg}\right.\right.$ tissue wet $\left.\mathrm{wt}^{-1}\right) /$ total $\mathrm{Hg}\left(\mathrm{mg} \mathrm{Hg} \cdot \mathrm{kg}\right.$ tissue wet $\left.\left.\mathrm{wt}^{-1}\right)\right] \times 100$.
    ${ }^{\mathrm{f}}$ Recovered prey $\mathrm{Hg}=\mathrm{Hg}\left(\mathrm{mg} \mathrm{Hg} \cdot \mathrm{kg}\right.$ wet $\left.\mathrm{wt}^{-1}\right)$ of food items removed from fish stomachs.
    ${ }^{\mathrm{g}}$ Trophic levels of individual target fish were calculated using Equation 2.

