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Mercury bioaccumulation in cartilaginous fishes from Southern New England coastal waters: Contamination from a trophic ecology and human health perspective

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Abstract

This study examined total mercury (Hg) concentrations in cartilaginous fishes from Southern New England coastal waters, including smooth dogfish (*Mustelus canis*), spiny dogfish (*Squalus acanthias*), little skate (*Leucoraja erinacea*), and winter skate (*L. ocellata*). Total Hg in dogfish and skates were positively related to their respective body size and age, indicating Hg bioaccumulation in muscle tissue. There were also significant inter-species differences in Hg levels (mean \pm 1 SD, mg Hg/kg dry weight, ppm): smooth dogfish (3.3 ± 2.1 ppm; $n = 54$) > spiny dogfish (1.1 ± 0.7 ppm; $n = 124$) > little skate (0.4 ± 0.3 ppm; $n = 173$) ~ winter skate (0.3 ± 0.2 ppm; $n = 148$). The increased Hg content of smooth dogfish was attributed to its upper trophic level status, determined by stable nitrogen ($\delta^{15}\text{N}$) isotope analysis (mean $\delta^{15}\text{N} = 13.2 \pm 0.7\text{‰}$), and the consumption of high Hg prey, most notably cancer crabs (0.10 ppm). Spiny dogfish had depleted $\delta^{15}\text{N}$ signatures ($11.6 \pm 0.8\text{‰}$), yet demonstrated a moderate level of contamination by foraging on pelagic prey with a range of Hg concentrations, e.g., in order of dietary importance, butterfish (Hg = 0.06 ppm), longfin squid (0.17 ppm), and scup (0.11 ppm). Skates were low trophic level consumers ($\delta^{15}\text{N} = 11.9\text{--}12.0\text{‰}$) and fed mainly on amphipods, small decapods, and polychaetes with low Hg concentrations (0.05–0.09 ppm). Intra-specific Hg concentrations were directly related to $\delta^{15}\text{N}$ and carbon ($\delta^{13}\text{C}$) isotope signatures, suggesting that Hg biomagnifies across successive trophic levels and foraging in the benthic trophic pathway increases Hg exposure. From a human health perspective, 87% of smooth dogfish, 32% of spiny dogfish, and < 2% of skates had Hg concentrations exceeding the US Environmental Protection Agency threshold level (0.3 ppm wet weight). These results indicate that frequent consumption of smooth dogfish and spiny dogfish may adversely affect human health, whereas skates present minimal risk.

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Keywords

bioaccumulation; diet; dogfish; food web; mercury; skate; stable isotope

1. Introduction

Mercury (Hg) is a pervasive toxicant that is introduced into the environment mainly through anthropogenic activities (US EPA, 1997). In the northeastern US, for example, Hg levels are elevated in marine coastal environments due to local point sources and distant non-point sources of contamination. The latter is dominated by the long-range transport of emissions from fossil-fuel combustion, after which the Hg by-product enters a water body through direct atmospheric deposition and watershed transport (Clarkson, 1992). Estuarine and coastal sediments are repositories for Hg (Balcom et al., 2004), and are the principal location for methylation, a bacterial-mediated process that converts inorganic Hg to its more toxic organic form, methylmercury (MeHg) (Gilmour et al., 1992; Benoit et al., 2003). MeHg derived from surface sediments may then be mobilized to the water column and transferred to biota through several physical and biological processes (Chen et al., 2008), including the bio-concentration of the contaminant in phytoplankton. This functional group, in turn, effectively transfers MeHg to both pelagic and benthic food webs (Mason et al., 1996; Moye et al., 2002), after which MeHg biomagnifies across successive trophic levels, concentrating in the tissues of top-level predators, including fish and other consumers (Wiener et al., 2003).

Fish are exposed to MeHg mainly through diet (Hall et al., 1997), and MeHg exposure causes numerous health deficits in fish. The severity of these adverse effects depends on the interplay between intra-species life history traits and the concentration and duration of MeHg exposure. Fish MeHg concentrations, for example, are positively related to body size and age when dietary intake exceeds depuration rates of the contaminant (Trudel and Rasmussen, 1997). Prey preferences and foraging ecology also impact MeHg dynamics, such that toxicant concentrations are elevated in fish feeding at higher trophic levels (Piraino and Taylor, 2009; Payne and Taylor, 2010; Szczebak and Taylor, 2011). Accordingly, fish MeHg bioaccumulation rates need to be examined concurrently with species-specific characteristics, such as body size, age, and ontogenetic shifts in diet and habitat use. Acute MeHg toxicity in fish (tissue MeHg = 6.0 mg/kg wet weight) may lead to neurological disease, including reduced swimming activity and loss of equilibrium, and possibly death (Armstrong, 1979; Wiener and Spry, 1996; Smith and Weis 1997). Chronic, low-dose MeHg exposure in fish may also cause more subtle end points of toxicity (tissue MeHg = 0.1 mg/kg wet weight), with possible deleterious effects to both the individual (e.g., compromised metabolism, osmoregulation, oxygen exchange, reproduction, and growth) and population (e.g., reduced survival and recruitment success) (Candelmo et al., 2010).

The majority of Hg-related research in marine ecosystems has focused on bony fishes (Division Teleostei) because of their importance as a human food resource (US EPA, 1997). Comparatively, there is a paucity of information on Hg burdens in cartilaginous fishes (Subclass Elasmobranchii), although a number of these species support important

commercial and recreational fisheries. Previous investigations on this topic have almost exclusively targeted sharks and have been limited geographically to coastal Pacific regions (Endo et al., 2009), tropical and sub-tropical waters (Heuter et al., 1995; de Pinho et al., 2002; Garcia-Hernandez et al., 2007; Maz-Courrau et al., 2012), and the pelagic-oceanic realm (Estrada et al., 2003; Branco et al., 2007; Suk et al., 2009). These prior studies revealed that many sharks experience contaminant concentrations above which adverse effects are realized, and further, may constitute a potential health risk for human consumers. There remains a lack of Hg research on a broad range of cartilaginous fishes from coastal regions of the Northwest Atlantic (e.g., sharks and skates), despite the ecological importance of these species in near-shore environments (Collette and Klein-MacPhee, 2002). Moreover, the broad distribution, high abundance, and upper trophic level status of cartilaginous fishes suggest that they strongly influence the fate of Hg in coastal habitats.

The primary objective of this study was to examine Hg bioaccumulation rates in cartilaginous fishes from Southern New England coastal waters, including the smooth dogfish, (*Mustelus canis*), spiny dogfish, (*Squalus acanthias*), little skate, (*Leucoraja erinacea*), and winter skate, (*L. ocellata*). Total Hg concentrations were measured in these target species and results were analyzed relative to intra-specific life history traits, including body size, age, sex, habitat use, and feeding habits. For the latter, conventional stomach content analysis was coupled with stable isotope (nitrogen and carbon) measurements to assess the effect of diet history and trophic processes on Hg contamination (Shiffman et al., 2012). Representative prey of each target fish were also analyzed for Hg content to determine their effect on Hg exposure. Lastly, given the potential value of each target species as a human dietary resource (NEFSC, 1999; 2006; NMFS, 2010), Hg results were evaluated relative to the US Environmental Protection Agency (US EPA) and US Food and Drug Administration (US FDA) criteria for the safe consumption of fishery products.

2. Methods

2.1. Target fishes

Smooth dogfish (Family Triakidae) and spiny dogfish (Family Squalidae) are the most numerically dominant shark species in the coastal western Atlantic (Collette and Klein-MacPhee, 2002). Smooth dogfish are a demersal species that occupy shallow near-shore waters (< 18 m to 200 m) from the Bay of Fundy to Florida, with maximal abundances in the Mid-Atlantic region (Cape Hatteras, North Carolina to New Jersey). Similarly, spiny dogfish are principally located from Nova Scotia southward to Cape Hatteras, but this species occupies a broader range of continental shelf waters (shallows to 900 m) and utilizes both epibenthic and pelagic habitats (Stehlik, 2007). Both smooth and spiny dogfish undergo pronounced latitudinal migrations in response to seasonal temperature changes, as well as onshore-offshore movements that are governed by prey availability and the onset of reproductive events, e.g., inshore pupping (Collette and Klein-MacPhee, 2002). Smooth and spiny dogfish also have varying life history strategies. Smooth dogfish are moderately sized [maximum size ~ 150 cm total length (TL)] and have relatively fast growth rates (15-20 cm TL/year) and short life spans (10-15 years). Conversely, spiny dogfish are characterized by a smaller maximum body size (~ 100 and 125 cm TL for males and females, respectively),

slower growth rate (1.5-3.5 cm TL/year), and extended longevity (35-40 to perhaps 100 years) (Collette and Klein-MacPhee, 2002; Stehlik, 2007). Finally, smooth dogfish are mainly benthic foragers with decapod crustaceans (crabs) representing the dominant prey, whereas the use of pelagic waters by spiny dogfish is reflected in their diet, such that fish, squid, and ctenophores are the most important food resources (Smith and Link, 2010).

Little skates and winter skates are sympatric species (Family Rajidae) with contrasting life history characteristics. Little skates are relatively small-bodied [maximum size ~ 30 cm disk width (DW)] and short-lived (~ 12 years), whereas winter skates have a longer life span (~ 20 years) and achieve an appreciable larger body size (maximum size ~ 60 cm DW) (Packer, 2003a, 2003b). The diet of both skates consist primarily of macro-invertebrates (e.g., amphipods, polychaetes, and decapod crustaceans), with fish becoming an increasingly important prey item for larger individuals (Smith and Link, 2010). Little skates and, to a lesser extent, winter skates often numerically dominant the demersal fish community in the Northwest Atlantic (Collette and Klein-MacPhee, 2002). Their specific geographic ranges are comparable and encompass coastal waters from southeastern Newfoundland to Cape Hatteras.

2.2. Sample collection and preparation

Dogfish, skates, and their prey were collected from Rhode Island/Block Island Sound and Narragansett Bay from May to October (2009-2012) using bottom trawls and hook & line (Fig. 1), and specimens were identified by their time (year and day of collection) and location (latitude-longitude) of capture. Prey were selected for analysis based on their recognized dietary contribution to dogfish and skates (Smith and Link, 2010), and included scup (*Stenotomus chrysops*), butterfish (*Peprilus triacanthus*), longfin inshore squid (*Doryteuthis pealeii*), and cancer crabs (*Cancer irroratus* and *C. borealis*). Dogfish, skates, and prey collected in the field were either processed immediately after capture or put on ice for transportation and frozen at -20°C in the laboratory for subsequent analysis.

The processing of dogfish, skates, and prey in the field and laboratory included assessing the sex of each shark and skate (presence/absence of claspers), measuring individual body size, excising muscle tissue samples, and extracting stomachs (trawl-collected dogfish and skates only). Size was measured as whole-body wet weight (g) and length or width (mm): dogfish, scup, and butterfish = TL; skate = DW; squid = mantle length (ML); and crab = carapace width (CW). The age (years) of each dogfish and skate was also estimated from sex-specific age-length relationships reported in the literature (Soldat, 1982; Collette and Klein-MacPhee, 2002; Conrath et al., 2002; Sulikowski et al., 2003; Cicia et al., 2009). Muscle tissue samples (2-5 g wet weight with skin removed) were excised from the dorsal axial musculature of dogfish and scup, pectoral wings of skates, and mantle dorsal body wall of squid using stainless-steel surgical blades, whereas crabs were processed as whole bodies. All muscle-tissue and whole-body samples were freeze-dried for 48 h (Labconco FreeZone 4.5-L Benchtop Freeze-Dry System), homogenized with clean stainless-steel spatulas, and stored at room temperature in borosilicate vials. The stomachs of dogfish and skates (~ 5 individuals/size class/species/trawl tow; assumed to have contents) were extracted

immediately after capture and preserved in Normalin® following the procedures of Smith and Link (2010).

2.3. Mercury analysis

Total Hg concentrations in mg/kg dry weight (ppm) was measured in homogenized muscle-tissue and whole-body samples of dogfish, skates, and prey (~ 0.03 g dry weight) using automated combustion atomic absorption spectrometry (AC-AAS) (DMA-80 Direct Hg Analyzer, Milestone, Inc., Shelton, Connecticut, USA), with a detection limit of 0.01 ng Hg (US EPA, 1998). The Hg analyzer was calibrated using certified reference materials (CRMs) of known Hg concentrations, and included solid standards (TORT-1: lobster hepatopancreas; DORM-2: dogfish muscle) and aqueous standards prepared by the National Research Council Canada, Institute of Environmental Chemistry (Ottawa, Canada) and the National Institute of Standards and Technology (Gaithersburg, Maryland, USA), respectively. Calibration curves were highly linear (mean $R^2 = 1.00$; range $R^2 = 0.99-1.00$; $p < 0.0001$), and the recovery of independently analyzed samples of TORT-1, DORM-2, and PACS-2 (marine sediment) CRMs ranged from 91.9% to 107.5% (mean = 96.2%). All samples were analyzed as duplicates, and an acceptance criterion of 10% was implemented. Duplicate samples with < 10% error were averaged for subsequent analysis (mean absolute difference between duplicates = 3.8%). Samples with > 10% error were reanalyzed to achieve the acceptance criterion or were eliminated from further analysis. For additional quality control, blanks were analyzed every 10 samples to assess instrument accuracy and potential drift. Further, two previous studies determined that AC-AAS used in this study produced statistically equivalent results to isotope dilution gas chromatography-inductively coupled plasma mass spectrometry, with R^2 values ranging from 0.902 and 0.946 between the two methods (Piraino and Taylor, 2009; Payne and Taylor, 2010).

2.4. Diet and trophic ecology analysis

The contents of preserved dogfish and skate stomachs were extracted in the laboratory and the total weight of these contents was measured with analytical balances (mg wet weight). All recovered prey items were identified to the lowest practical taxon with the aid of stereomicroscopes, and when possible, the length or width of individual prey was measured as defined above. The contribution of each prey taxon to the overall diet of dogfish and skates was expressed as the frequency of occurrence (% F) and percent weight (% W). Frequency of occurrence was calculated as the number of stomachs containing a specific prey taxon divided by the total number of stomachs with prey contents, whereas percent weight was calculated as the weight of a specific prey taxon divided by the total weight of all prey types. The relative importance of each prey taxon to the diet of dogfish and skates was also assessed using a modified alimentary index (% IA):

$$\%IA = \frac{IA_i}{\sum_{i=1}^n IA_i} \times 100 \quad (1)$$

where, IA is calculated for each prey taxon i as the product of % F_i and % W_i , and n is the total number of prey taxa identified in the stomach contents of dogfish and skates.

Stable isotope analysis was used to complement stomach content data and assess the effect of trophic processes on dogfish and skate Hg concentrations (Shiffman et al., 2012). Specifically, stable nitrogen ($^{15}\text{N}/^{14}\text{N}$) isotope signatures were used to estimate time-integrated feeding history (Michener and Kaufman, 2007), whereas carbon ($^{13}\text{C}/^{12}\text{C}$) isotopes were used as indicators of the initial carbon source to the marine food web, thus allowing for the differentiation between pelagic and benthic trophic pathways (Fry, 2006). A sub-sample of dogfish and skates previously selected for stomach content analysis were used for stable isotope measurements (smooth dogfish: $n = 41$; spiny dogfish: $n = 107$; little skate: $n = 90$; winter skate: $n = 82$). Isotope measurements of a sub-sample of muscle tissue (~ 1 mg dry weight) were performed by the Boston University Stable Isotope Laboratory (Boston, Massachusetts) using automated continuous-flow isotope ratio mass spectrometry (CF-IRMS). Previous studies on the stable isotope signatures of cartilaginous fishes indicated that their muscle tissue has isotopic turnover rates of 11-14 months (MacNeil et al., 2006; Logan and Lutcavage, 2010), and in this study, muscle samples were not pre-treated for lipid extraction owing to the relatively low lipid content of this tissue (Hussey et al., 2010; Kim et al., 2011). Ratios of $^{15}\text{N}/^{14}\text{N}$ and $^{13}\text{C}/^{12}\text{C}$ are described using the standard delta notation (δ), expressed as the relative per mil (‰) difference between the samples and international standards (atmospheric nitrogen, $^{15}\text{N}_{\text{air}}$, and Vienna Pee Dee Belemnite, $^{13}\text{C}_{\text{V-PDB}}$, respectively), and calculated using the following equation:

$$\delta X = (R_{\text{sample}}/R_{\text{standard}} - 1) \times 1000 \quad (\text{‰}) \quad (2)$$

where, X is ^{15}N or ^{13}C and R is $^{15}\text{N}/^{14}\text{N}$ or $^{13}\text{C}/^{12}\text{C}$. The recovery of internal reference materials (peptone and glycine) for the CF-IRMS method was 99.6% and 99.7% for nitrogen and carbon, respectively. The mean sample precision determined from duplicate analyses was 98.6% (range = 90.2-100.0%) and 99.5% (range = 93.1-100.0%) for nitrogen and carbon, respectively.

2.5. Data analysis

Inter-species differences in mean total Hg concentrations and isotopic signatures ($\delta^{15}\text{N}$ and $\delta^{13}\text{C}$) among dogfish and skates were analyzed with two-way analysis of variance (ANOVA) models using species and sex as fixed factors, whereas differences in prey Hg content across taxa were examined with a one-way ANOVA model. The post hoc separation of mean differences in Hg and isotope values across 4 levels of target species and 4 levels of prey species (Hg only) were contrasted with independent Ryan-Einot-Gabriel-Welsch (Ryan's Q) multiple comparison tests. Prior to these analyses, data were \log_{10} -transformed to meet assumptions of normality and homogeneity of variance. The effects of body size, age, and isotopic values on the Hg concentration of dogfish, skates, and prey (size only) were analyzed with least-squares exponential regressions (size and age) and linear regressions ($\delta^{15}\text{N}$ and $\delta^{13}\text{C}$). Also for dogfish and skates, analysis of covariance (ANCOVA) models were used to assess the effect of sex and species on Hg bioaccumulation rates, with age as the covariate and sex or species as the discrete explanatory variable. Lastly, multiple linear regression analysis was used to assess the effects of several biotic and abiotic variables on Hg concentrations and isotopic signatures of dogfish and skates. Five regression models were employed that either examined: (1) the independent effects of body size, location of

catch (latitude and longitude; decimal degrees), and time of catch (year and day of collection) on the response variables, or (2) the cumulative effects of all predictor variables, as determined from global and stepwise linear regression models. Small-sample, bias-corrected Akaike's Information Criterion (AIC_c) and Akaike's weights (w_i) were used to evaluate and select the optimal regression model (Burnham and Anderson, 2002):

$$AIC_c = -2 \log [\mathcal{L}(\hat{\theta})] + 2K + \frac{2K(K+1)}{n-K-1} \quad (3)$$

$$w_i = \exp(-0.5\Delta_i) / \sum_{k=1}^5 \exp(-0.5\Delta_k) \quad (4)$$

where, \mathcal{L} is the likelihood function of the model parameters, n is the sample size, K is the number of regression parameters, and Δ_i is equal to $AIC_{c,i} - AIC_{c,\min}$. The model with the smallest AIC_c value ($AIC_{c,\min}$) had the most support, and the w_i value quantified the probability that model i was the best among the candidate models.

3. Results and Discussion

3.1. Mercury concentrations and bioaccumulation in cartilaginous fishes

Mean total Hg concentrations varied significantly among dogfish and skate species, but not as a function of their sex (Tables 1 and 2). Smooth dogfish had the highest mean Hg concentration (mean \pm 1 SD = 3.3 ± 2.1 ppm), followed by spiny dogfish (1.1 ± 0.7 ppm) and skates (little: 0.4 ± 0.3 ppm; winter: 0.3 ± 0.2 ppm) (Fig. 2A). The Hg concentrations reported in this study are consistent with literature values for conspecifics and congeners from other geographic areas, including those collected from European and Asian waters (Table 3). Conversely, there is also evidence of spatial and temporal variations in target fish Hg content. In US coastal environments, for example, spiny dogfish collected from the Pacific during the early to mid-1970s had markedly higher Hg concentrations relative to Atlantic conspecifics (Table 3). In the northwestern Atlantic, the mean Hg content of spiny dogfish has ostensibly declined $\sim 35\%$ over the last several decades. Winter skate Hg concentrations in this study were also $\sim 50\%$ greater than levels measured in equally-sized conspecifics from New York commercial markets, where skates were collected from a relatively broad geographic area (mid- to north Atlantic; U.S. EPA, 2013). The observed spatio-temporal differences in fish Hg burdens are attributed to geographic variability and historical changes in contaminant inputs to coastal habitats (Benoit et al., 2003); the latter includes biota directly responding to recent reductions in point sources of Hg in the northeastern US (Sunderland et al., 2012). Moreover, geochemical, physicochemical, and ecological processes in marine coastal habitats vary over relatively small spatial and temporal scales, thus affecting Hg mobilization and its eventual incorporation and transfer through trophic pathways (Chen et al., 2008).

Total Hg concentrations in dogfish and skates were directly related to their body size and age (Table 4; Figs. 3 and 4), confirming the accumulation of Hg in muscle tissue. Multivariate regression analysis and estimates of the AIC_c and w_i further revealed that body

size was the most significant factor affecting the Hg content of all species (Table 5), although the level of influence varied between dogfish ($R^2 = 0.27-0.29$) and skates ($R^2 = 0.08-0.10$). There were also significant seasonal and annual variations in spiny dogfish and winter skate Hg concentrations (Table 5), but these factors contributed minimally to the cumulative R^2 -values for each model (day and year partial R^2 -values = 0.03-0.04). Previous studies purported Hg bioaccumulation in dogfish (Forrester et al., 1972; Hall et al., 1977; de Pinho et al., 2002; Endo et al., 2009; 2013), which is attributed to the disproportionate uptake of Hg relative to its low depuration rate (Maz-Courrau et al., 2012). In contrast, Storelli et al. (1998) found no correlation between the Hg content of three skate species (*Raja* spp.) and their respective body weight.

Total Hg concentrations varied significantly by sex in little skates when age was included as a covariate, such that females had increased contaminant levels at a given age relative to males (Table 6). Conversely, Hg concentrations did not differ between sexes in dogfish and winter skates (Table 6). The absence of gender-specific Hg burdens in dogfish is inconsistent with prior studies, and to the knowledge of the authors, has not been examined in skates. Endo et al. (2009; 2013) observed that male spiny dogfish (16-41 years) and star-spotted dogfish (*Mustelus manazo*) (4-8 years) had higher Hg concentrations relative to females (*S. acanthias*: 18-61 years; *M. manazo*: 2-14 years), which is caused by the cessation of somatic growth in mature male dogfish. In this study, the lack of gender effects on dogfish Hg concentrations were likely due to the comparatively narrow age ranges examined in target fishes (Table 1); noting that Endo et al. (2009; 2013) reported negligible differences in sex-specific Hg concentrations at younger age classes.

Hg bioaccumulation rates did not differ between smooth and spiny dogfish, and similarly, between little and winter skates (ANCOVA: Age \times Species, $p = 0.06-0.16$; Table 6). At a defined age, however, smooth dogfish and little skates had higher Hg concentrations than spiny dogfish and winter skates, respectively (Table 6; Fig. 4). It is well documented that rapid somatic growth in bony fishes reduces overall Hg burdens through growth dilution; a response caused by the disproportionate increase in fish size relative to Hg dietary intake (Wang, 2012 and references therein). Results from this study contradict growth dilution because of the increased Hg content of faster growing species (Collette and Klein-MacPhee, 2002; Frisk et al., 2006). Endo et al. (2013) also obtained results that refuted Hg biodilution in cartilaginous fishes, such that the star-spotted dogfish had higher Hg concentrations compared to slower growing spiny dogfish. Differences in the growth rates between mustelid and squalid dogfish (*M. canis*: 15-20 cm TL/year; *S. acanthias*: 1.5-3.5 cm TL/year; Collette and Klein-MacPhee, 2002; Stehlik, 2007) are also reflected in their metabolic expenditures (*Mustelus norrisi*: 161 mg O₂/kg/hr at 26°C; *S. acanthias*: 92 mg O₂/kg/hr at 22°C; Brett and Blackburn, 1978; Bushnell et al., 1989; Carlson and Parsons, 2001). To sustain their comparatively high bioenergetic demands (i.e., growth and metabolism), mustelid dogfish have elevated feeding rates, expressed as % body weight consumed per day (*Mustelus californicus*: 1.3-1.6%; *S. acanthias* = 0.4-1.3%; Holden, 1966; Jones and Green, 1977; San Filippo, 1996), thus concurrently increasing their dietary uptake and exposure to Hg. Further, differences in the food habits and Hg content of preferred prey, as discussed in

subsequent sections, likely obscure the Hg growth dilution response in dogfish and skates (Stafford and Haines, 2001).

3.2. Diet and trophic ecology of cartilaginous fishes

Direct visual analysis of smooth dogfish ($n = 34$), spiny dogfish ($n = 57$), little skate ($n = 125$), and winter skate ($n = 92$) stomach contents revealed inter-species differences in feeding habits (Tables 7 and 8), and the results herein are consistent with previous diet analyses of the target fishes (Smith and Link, 2010). The mean standardized weight of stomach contents was greater in dogfish than skates (stomach content weight/whole-body wet weight ~ 2 -3% and 0.3-0.4%, respectively), whereas skates generally had more distinct prey taxa in a single stomach compared to dogfish (4.2-4.5 and 2.0-3.5 unique prey/stomach, respectively). The results also suggest that spiny dogfish have a more restricted overall diet relative to smooth dogfish and skates (Tables 7 and 8); spiny dogfish consumed a total of 13 different prey taxa, whereas 18-21 novel prey taxa were identified in the stomachs of smooth dogfish and skates (excluding “unidentified” categories).

Spiny dogfish mostly consumed pelagic prey (bony fish and squid), whereas smooth dogfish fed almost exclusively on benthic invertebrates (crustaceans and polychaetes) (Table 7). The dominant prey of spiny dogfish, with respect to frequency of occurrence, were butterflyfish, scup, unidentified fish, longfin squid, and cancer crabs. Butterflyfish also accounted for the largest percentage by weight of spiny dogfish stomach contents, followed by longfin squid, unidentified fish, scup, and silver hake. The order of importance of prey to the diet of spiny dogfish, as expressed by the alimentary index (%IA), were butterflyfish, longfin squid, and unidentified fish, while the remaining prey groups had %IA less than 3%. Cancer crabs, polychaetes, and unidentified animal tissue (including fish and crabs) were the most frequency encountered prey in smooth dogfish stomachs, followed by algae/detritus, and other decapod crustaceans. According to other measures of dietary importance, cancer crabs and unidentified fish were the most utilized prey resource by smooth dogfish, while all remaining prey were of lesser importance (%IA < 1%).

Little skates and winter skates mostly consumed benthic prey (Table 8). Amphipods were the most frequently observed prey in skate stomachs, followed by polychaetes and other crustaceans, including sand shrimp, cancer crabs, and isopods. Amphipods and polychaetes also accounted for a high percentage of the stomach weight in both skate species, while fish also contributed considerably to the stomach weight of larger winter skates (> 350 mm DW), including sand lance and scup. The order of prey dietary importance for skates, expressed as %IA, were amphipods, polychaetes, sand shrimp, and cancer crabs, while the remaining identifiable prey taxa had %IA less than 3%.

Stable nitrogen ($\delta^{15}\text{N}$) isotope signatures were used to approximate the trophic status of dogfish and skates (Michener and Kaufman, 2007), and mean $\delta^{15}\text{N}$ values varied significantly among target fish (Table 2). Smooth dogfish had a significantly enriched $\delta^{15}\text{N}$ signature relative to spiny dogfish (13.2 ± 0.7 and $11.6 \pm 0.8\text{‰}$, respectively), which corresponds to smooth dogfish occupying a higher trophic position (Fig. 2B). The depleted $\delta^{15}\text{N}$ signature of spiny dogfish is attributed to the dietary contribution of low trophic level prey, including planktivorous forage fish (e.g., butterflyfish; %IA = 48.4%; Table 7) and

possibly ctenophores (Smith and Link, 2010), although ctenophores were not detected in dogfish stomachs due to their fragile tissues. Moreover, female spiny dogfish ($11.7 \pm 0.7\text{‰}$) had a significantly higher $\delta^{15}\text{N}$ value relative to male conspecifics ($10.8 \pm 0.4\text{‰}$), thus explaining the species-sex interaction effect in the twoway ANOVA model (Table 2). Little and winter skates had significantly lower $\delta^{15}\text{N}$ signatures than smooth dogfish, but the mean $\delta^{15}\text{N}$ values between skate species did not differ (little: $12.0 \pm 0.7\text{‰}$; winter: $11.9 \pm 1.0\text{‰}$; Fig. 2B), suggesting that these species occupy the same, relatively low, trophic level.

Stable carbon ($\delta^{13}\text{C}$) isotope signatures were used to differentiate among varying sources of primary production to the coastal food web, and thus, distinguish between benthic and pelagic trophic linkages (Peterson and Howarth, 1987; France, 1995). Mean $\delta^{13}\text{C}$ values ranged from -22.0 to -16.5‰ and varied significantly among dogfish and skates, but not as a function of their sex (Table 2; Fig. 2C). Moreover, two distinct isotopic groups were evident that further corroborated this study's stomach content analysis. The depleted $\delta^{13}\text{C}$ value of spiny dogfish ($\sim -22\text{‰}$) indicated a phytoplankton-based (pelagic) food web, which is consistent with their preferred consumption of butterfish and squid. Conversely, the more enriched $\delta^{13}\text{C}$ signatures ($\sim -17\text{‰}$) of smooth dogfish and skates suggest benthic sources of primary production, which is presumably derived from their consumption of epibenthic crustaceans and infaunal polychaetes.

Body size, location of catch (latitude and longitude), and time of catch (day of year and year) were the most significant factors affecting dogfish and skate isotopic signatures (Table 5). The estimated slope coefficients for the size- $\delta^{15}\text{N}$ relationship were positive for spiny dogfish and skates, indicating that older individuals forage at higher trophic levels. There was also an inverse relationship between body size and $\delta^{13}\text{C}$ values in winter skates, which is attributed to larger skates (> 350 mm DW) consuming pelagic prey, including butterfish, sand lance, and squid (Table 8). Dogfish and skate isotopic signatures also exhibited significant spatio-temporal variations (Table 5). Specifically, $\delta^{15}\text{N}$ and $\delta^{13}\text{C}$ values increased in a northeasterly direction, in closer proximity to the mainland (“+” and “-” coefficient for latitude and longitude, respectively) (Fig. 1), as well as becoming more enriched as the sampling season progressed (“+” coefficient for day of year and year). It is unlikely that differences in dogfish and skate foraging ecology accounted for the observed spatio-temporal variations in isotopic values. It is more probable that these differences are the result of species-specific seasonal and annual migrations, and thus, spatio-temporal variability in habitat use. This is especially relevant to dogfish because of their expansive coastal migrations (Collette and Klein-MacPhee, 2002). Moreover, the added effect of site-specific environmental conditions (e.g., spatially-explicit salinity profiles, terrestrially-derived and increased nutrient loading along the immediate coastline) can have pronounced effects on the isotopic signatures of the target fishes (Pruell et al., 2006; Michener and Kaufman, 2007).

3.3. Trophic effects on cartilaginous fish mercury concentrations

Representative prey of dogfish and skates were measured for total Hg content to assess their potential effect on target fish Hg levels (Table 1). Mean prey Hg concentrations varied significantly across taxa (Table 2; Fig. 2D), such that scup had the highest Hg content (0.29

± 0.18 ppm), followed by crabs (0.16 ± 0.11 ppm), squid (0.11 ± 0.04 ppm), and butterfish (0.07 ± 0.4 ppm). Prey Hg concentrations were also positively related to body size (Table 4; Fig. 5), again indicating the bioaccumulation of the contaminant. Inter-species differences in dogfish and skate Hg concentrations are adequately explained by the Hg content of their preferred prey. In this study, smooth dogfish had the highest Hg concentration among target fishes, and this species preferentially consumed cancer crabs with a mean body size of 55 mm CW, i.e., the mean size of crabs recovered from smooth dogfish stomachs (Table 7). Crabs of this relatively large size have an estimated Hg content of 0.10 ppm, as determined from the Hg-size regression model (Table 4). It is important to reiterate that crabs in this study were processed as whole bodies, whereas the Hg content of other prey was measured in excised muscle tissue. This discrepancy in tissue-type likely underestimates the Hg concentration of cancer crabs. For example, the Hg content of blue crab (*Callinectes sapidus*) muscle tissue is ~ 30% greater than the Hg burden of whole-body samples (Adams and Engel, 2014). Spiny dogfish had a moderate level of Hg contamination, and this species relied on prey that had both low and high Hg concentrations; in order of dietary importance, butterfish (Hg = 0.06 ppm for 108 mm TL fish), longfin squid (Hg = 0.17 ppm for 168 mm ML squid), and scup (Hg = 0.11 ppm for 94 mm TL fish) (Tables 4 and 7). Finally, little and winter skates had the lowest Hg concentrations observed in this study, and these species fed upon amphipods, polychaetes, and small cancer crabs (< 20 mm CW). Payne and Taylor (2010) measured the Hg content of amphipods and polychaetes collected from the Narragansett Bay estuary (Fig. 1), each with a mean Hg concentration of 0.09 ppm (converted from wet weight). It is important to note that such prey species would likely have reduced Hg burdens in Rhode Island/Block Island Sound, as the environment is further removed from anthropogenic influences (Taylor et al., 2012). Moreover, the estimated Hg content of cancer crabs consumed by skates was 0.05 ppm (8-18 mm CW) (Tables 4 and 8). These collective results indicate that dogfish and skates are highly responsive to the Hg content of their preferred prey.

Stable isotope ($\delta^{15}\text{N}$ and $\delta^{13}\text{C}$) analysis was coupled with total Hg data to provide insight into the effects of trophic processes on Hg contamination in dogfish and skates. There were significant positive relationships between Hg and $\delta^{15}\text{N}$ values measured for spiny dogfish and skates (Table 4; Fig. 3); hence verifying an increase in intra-specific Hg concentrations at higher trophic levels. These results are consistent with previous analyses of the trophic effects on marine fish Hg concentrations, including several species of coastal and oceanic sharks (Adams et al., 2003; Cai et al., 2007; Suk et al., 2009; Maz-Courrau et al., 2012). Significant positive relationships were also observed between Hg and $\delta^{13}\text{C}$ values measured for dogfish and little skates (Table 4; Fig. 3). This result suggests that benthic associations may increase biotic Hg concentrations via dietary intake or proximate contact with Hg-contaminated sediments, as reported elsewhere (Storelli et al., 1998; Hosseini et al., 2013). Other studies, however, indicate that pelagic-feeding organisms (depleted $\delta^{13}\text{C}$ signatures) have increased Hg exposure (Chen et al., 2009; Kim et al., 2012). Ultimately, the fate of Hg through feeding pathways depends on the number of trophic steps and consumer's utilization of different prey assemblages (Kim et al., 2012), which likely vary across discrete aquatic ecosystems.

3.4. Cartilaginous fish mercury contamination from a human health perspective

The Hg concentrations in some fish may be sufficiently high to adversely affect human health (McMichael and Butler, 2005; Willett, 2005), and human exposure to Hg occurs mainly through dietary intake of contaminated fish (Hightower and Moore, 2003). Public health officials affiliated with federal and state agencies respond to the threat of fish Hg contamination by reporting criteria for the safe consumption of fishery products, including the US FDA and US EPA threshold levels of 1.0 and 0.3 ppm Hg wet weight, respectively. Cartilaginous fishes are becoming increasingly utilized as a human food source given the precipitous declines in traditional fisheries (NEFSC, 1999; 2006; NMFS, 2010), thus underscoring the importance of research focused on Hg contamination in these species. For the purposes of comparing the results of this study to the US FDA and US EPA action levels, Hg data originally expressed as dry weight were converted to wet weight assuming 75% water content of the muscle tissue (Bosch et al., 2013). Accordingly, 24.1% of smooth dogfish, 1.6% of spiny dogfish, and 0.0% of skates exceeded the US FDA criterion of 1.0 ppm Hg wet weight. For the more conservative US EPA threshold (0.3 ppm Hg), 87.0% of smooth dogfish, 32.0% of spiny dogfish, and < 2% of skates were above this value. The Hg-length exponential regression models also estimated that smooth and spiny dogfish obtain Hg concentrations of 0.3 ppm at relatively small body sizes (57.3 and 70.9 cm TL, respectively), whereas skates do not approach this threshold level even at their maximum body sizes (39.0 and 133.9 cm DW, respectively) (Table 4).

4. Conclusions

In this study, total Hg concentrations were measured in dogfish and skates, and results were evaluated relative to species-specific life history traits. The total Hg content of dogfish and skate muscle tissue was positively related to body size and age, indicating the bioaccumulation of the contaminant. Moreover, Hg concentrations were significantly elevated in smooth dogfish, followed by spiny dogfish and skates. The increased Hg content of smooth dogfish was attributed to its upper trophic level status, consumption of Hg-enriched prey (e.g., large cancer crabs), and relatively high bioenergetic requirements. Comparatively, spiny dogfish had a reduced trophic status owing to the dietary contribution of butterfish, yet demonstrated a moderate level of Hg contamination because this species also consumed high Hg prey (e.g., squid and scup). Little and winter skates were basal consumers and had the lowest Hg concentrations because they fed exclusively on Hg-depleted prey (e.g., amphipods, small decapods, and polychaetes). The collective results from the analysis of stable isotopes further indicated that Hg biomagnifies across successive trophic levels and foraging in the benthic trophic pathway may increase Hg exposure. From a human health perspective, the consumption of smooth dogfish and, to a lesser extent, spiny dogfish pose a human health risk, and therefore, justifies stringent consumption advisories for these species. Conversely, the consumption of skates does not present a significant risk to human health. It is the recommendation of the authors that this information be effectively communicated to the general public so that citizens can make informed decisions regarding the safe consumption of fishery resources.

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Research highlights

- ▶ Mercury bioaccumulates in the cartilaginous fishes examined in this study.
- ▶ Mercury content was higher in smooth dogfish, followed by spiny dogfish and skates.
- ▶ Trophic structure and diet preferences affect dogfish and skate mercury content.
- ▶ Eating dogfish may adversely affect human health, whereas skates present low risk.

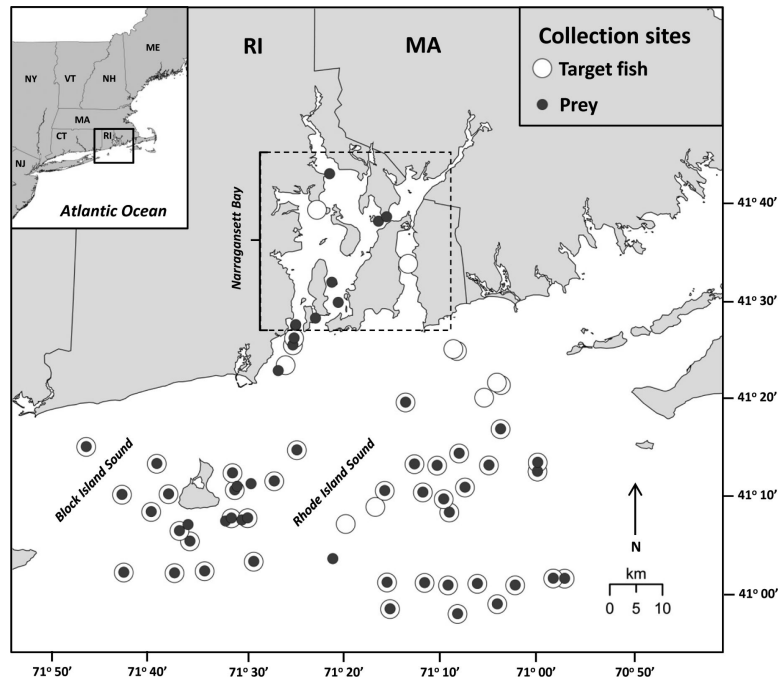


Fig 1. Map of Rhode Island Sound, Block Island Sound, and Narragansett Bay (Rhode Island, USA) with points denoting collection sites of target fish (smooth dogfish, spiny dogfish, little skate, and winter skate) and prey (scup, butterfish, longfin squid, and cancer crabs).

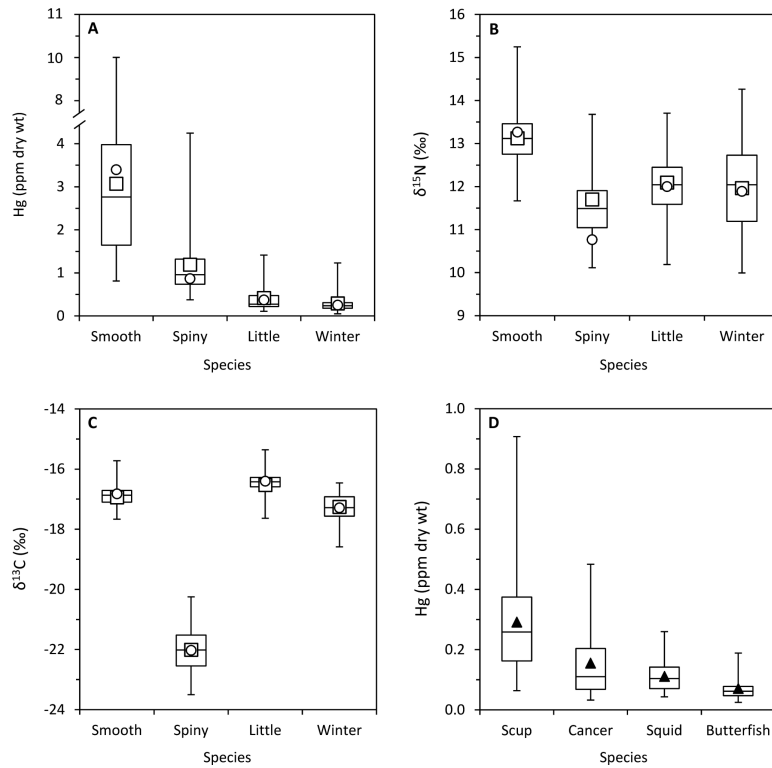


Fig. 2. Total mercury (Hg) concentrations (ppm dry weight) of target fish (A) and prey (D), and stable nitrogen (B) and carbon (C) isotope signatures ($\delta^{15}\text{N}$ and $\delta^{13}\text{C}$; ‰) of target fish. Box plots illustrate the median, 1st and 3rd quartiles, and maximum and minimum values. Open squares and circles represent means for female and male target fish, respectively, and solid triangles represent means for prey. Target fish include smooth dogfish, spiny dogfish, little skate, and winter skate, and prey include scup, cancer crabs, longfin squid, and butterfish.

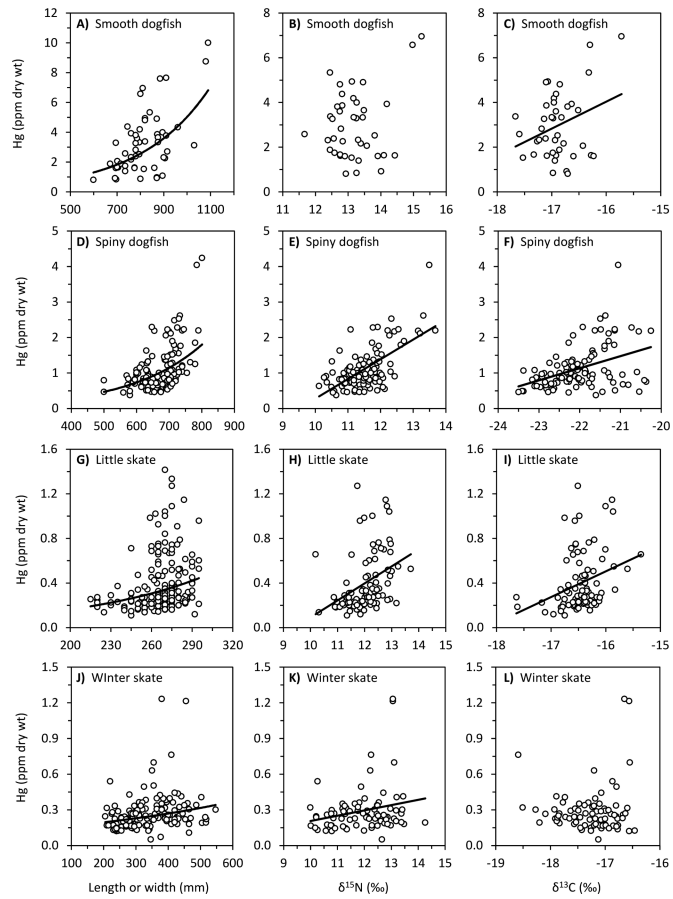


Fig. 3. Total mercury (Hg) concentrations (ppm dry weight) of target fish as a function of body size (total length or disk width; mm), and stable nitrogen and carbon isotope signatures ($\delta^{15}\text{N}$ and $\delta^{13}\text{C}$; ‰). Target fish include smooth dogfish (A-C), spiny dogfish (D-F), little skates (G-I), and winter skates (J-L). For significant relationships, exponential and linear regression models were fit to size and isotope data, respectively.

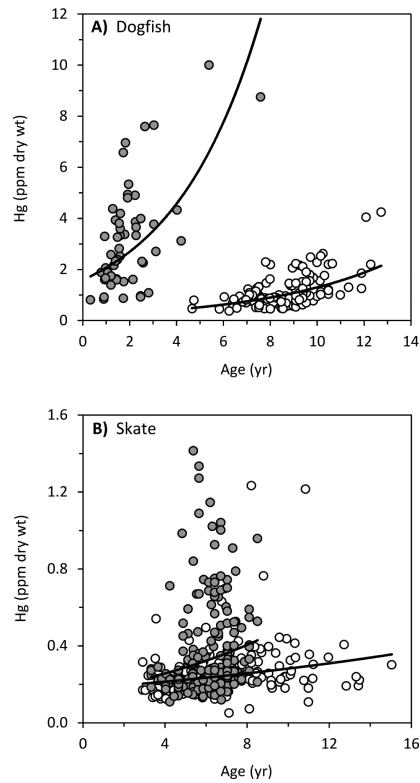


Fig. 4. Total mercury (Hg) concentrations (ppm dry weight) of dogfish (A) and skates (B) as a function of age. Smooth dogfish and little skates are denoted by solid circles, whereas spiny dogfish and winter skates are represented by open circles. Exponential regression models were fit to species-specific data.

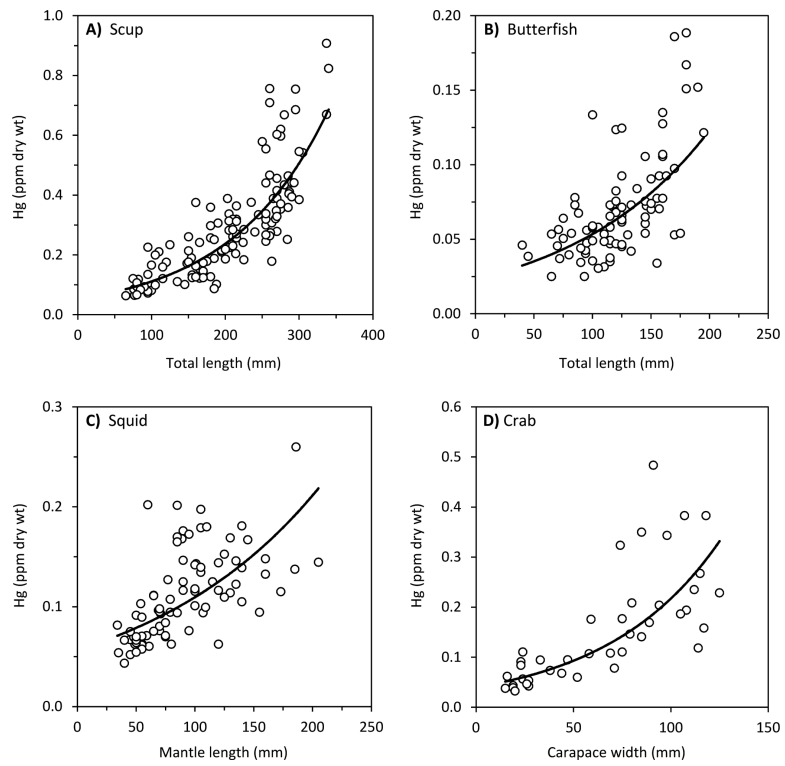


Fig. 5. Total mercury (Hg) concentrations (ppm dry weight) of scup (A), butterfish (B), longfin squid (C), and cancer crabs (D) as a function of body size (total length, mantle length, or carapace width; mm). Exponential regression models were fit to the data.

Comparison of total length, disk width, whole-body wet weight, and age among smooth dogfish, spiny dogfish, little skates, and winter skates. Sample sizes (n), means (± 1 SD), and ranges are reported for males, females, and combined data sets. Size data are also reported for representative prey.

Table 1

Species	n	Length/Width (mm)		Weight (kg)		Age (year)	
		Mean \pm SD	Range	Mean \pm SD	Range	Mean \pm SD	Range
Smooth dogfish							
Male	30	800.9 \pm 95.8	670-1080	2.73 \pm 0.69	1.48-4.62	1.97 \pm 1.34	0.78-7.59
Female	24	829.3 \pm 101.6	599-1090	4.03 \pm 1.10	1.43-6.00	2.00 \pm 1.04	0.33-5.38
Combined	54	813.5 \pm 98.5	-	3.31 \pm 1.10	-	1.98 \pm 1.20	-
Spiny dogfish							
Male	19	599.4 \pm 31.2	500-645	1.60 \pm 0.23	0.98-2.00	7.13 \pm 0.81	4.74-8.46
Female	105	685.1 \pm 49.6	500-801	2.83 \pm 0.74	1.04-5.78	8.91 \pm 1.38	4.66-12.73
Combined	124	672.1 \pm 56.4	-	2.65 \pm 0.82	-	8.64 \pm 1.45	-
Little skate							
Male	114	267.3 \pm 15.6	215-295	0.60 \pm 0.10	0.34-0.83	6.61 \pm 0.91	4.04-8.50
Female	59	264.5 \pm 18.4	220-295	0.60 \pm 0.10	0.32-0.77	5.19 \pm 0.88	3.32-6.97
Combined	173	266.2 \pm 16.6	-	0.60 \pm 0.10	-	6.13 \pm 1.12	-
Winter skate							
Male	80	326.6 \pm 86.0	210-547	1.20 \pm 1.07	0.23-5.17	6.31 \pm 2.90	2.91-15.05
Female	68	350.7 \pm 70.2	205-505	1.40 \pm 0.84	0.25-4.60	7.39 \pm 2.21	3.17-12.82
Combined	148	338.4 \pm 79.7	-	1.31 \pm 0.98	-	6.84 \pm 2.65	-
Prey							
Scup	126	203.3 \pm 69.7	65-340	0.249 \pm 0.201	0.006-0.872	-	-
Butterfish	87	121.8 \pm 33.8	40-195	0.045 \pm 0.033	0.004-0.174	-	-
Squid	84	90.6 \pm 38.7	34-205	0.038 \pm 0.033	0.002-0.154	-	-
Cancer crab	41	65.4 \pm 35.9	1.5-125	0.075 \pm 0.083	0.001-0.269	-	-

Table 2

Summary statistics for two-way analysis of variance (ANOVA) models used to examine target fish total mercury (Hg) concentrations (ppm dry weight) and stable nitrogen ($\delta^{15}\text{N}$) and carbon ($\delta^{13}\text{C}$) isotope signatures as a function of species and sex. Target fish include smooth dogfish (Smo), spiny dogfish (Spi), little skate (Lit), and winter skate (Win). Also reported are the summary statistics for a one-way ANOVA model used to examine prey Hg as a function of species, which include scup (Scu), cancer crabs (Can), squid (Squ), and butterfish (But). Results of Ryan's Q multiple comparisons tests that contrast Hg or isotope values across 4 levels of dogfish/skate and prey are reported.

Factor	Species		Sex		Species \times Sex		Ryan's Q test
	F (df)	p	F (df)	p	F (df)	p	
Target fish							
Hg	344.7 (3)	<0.0001	1.88 (1)	0.171	0.77 (3)	0.509	Smo > Spi > Lit ~ Win
$\delta^{15}\text{N}$	54.41 (3)	<0.0001	7.65 (1)	<0.01	6.36 (3)	<0.0005	Smo > Lit ~ Win > Spi
$\delta^{13}\text{C}$	1506.0 (3)	<0.0001	0.75 (1)	0.389	0.60 (3)	0.614	Lit > Smo > Win > Spi
Prey							
Hg	109.8 (3)	<0.0001	-	-	-	-	Scu > Can ~ Squ > But

Table 3

Literature review of dogfish and skate total mercury (Hg) concentrations (ppm dry weight). The following information is provided for each source document: fish species, location of sample, sample size (n), mean total length (TL; cm), and mean Hg concentrations reported in the “literature”. For comparative purposes, the total Hg content of *Mustelus canis*, *Squalus acanthias*, and *Leucoraja ocellata* (pre-defined size) were estimated from the exponential (Hg-size) regression models presented in “this study” (Table 4).

Species	Location	n	TL	Total Hg ¹		Literature source
				Literature	This study	
Dogfish						
<i>Mustelus canis</i>	southern coastal Brazil	79	89.2	1.6	1.7	de Pinho et al. (2002)
<i>Mustelus manazo</i>	northern coastal Japan	61	94.0	3.6	-	Endo et al. (2013) ²
<i>Mustelus mustelus</i>	Langebaan Lagoon, South Africa	12	112.5	8.4	-	Bosch et al. (2013) ^{2,3}
<i>Squalus acanthias</i>	northwestern Atlantic, US	80	76.6	1.6	1.2	Greig et al. (1977) ⁴
<i>S. acanthias</i>	northeastern Pacific, US	127	94.2	3.7	1.7	Hall et al. (1977) ⁵
<i>S. acanthias</i>	northeastern Pacific, US	88	86.8	2.4	1.5	^{1,3}
<i>S. acanthias</i>	coastal Japan	75	86.9	1.4	1.5	Endo et al. (2009) ²
<i>S. acanthias</i>	eastern Mediterranean Sea	47	58.0	8.3	0.84	Kousteni et al. (2006) ³
Skate						
<i>Leucoraja ocellata</i>	commercial markets, New York, US	14	36.6	0.24	0.50	US EPA (2013)
<i>Raja asterias</i>	Adriatic Sea	120	44	2.9	-	Storelli et al. (2003)
<i>Raja clavata</i>	mouth of River Tyne, England	22	45.6	0.39	-	Dixon & Jones (1994)
<i>R. clavata</i>	Bay of Biscay, France	11	93.5	2.0	-	Chouvelon et al. (2012)
<i>Raja eglanteria</i>	lower Delaware Bay, US	3	26	0.86	-	Gerhart (1977) ^{3,6}
<i>Raja kenoi</i>	eastern China Sea	2	39	0.68	-	Asante et al. (2008)
<i>Raja kwangtungensis</i>	eastern China Sea	3	35	<0.2	-	Asante et al. (2008)
<i>Raja microocellata</i>	Bay of Biscay, France	5	69.4	0.17	-	Chouvelon et al. (2012)

¹Literature Hg values expressed as wet weight were converted to dry weight assuming 75% water content (Bosch et al., 2012)

²Literature values averaged across female and male dogfish

³Median length

⁴ Samples collected from New York Bight to Gulf of Maine

⁵ Female dogfish only

⁶ Body size = disk width (cm)

Table 4

Summary statistics for univariate (non-linear) exponential and linear regression models used to examine the effect of body size [mm; total length (TL), disk width (DW), mantle length (ML), and carapace width (CW)], age (years), and stable nitrogen ($\delta^{15}\text{N}$) and carbon ($\delta^{13}\text{C}$) isotope signatures on biota total mercury (Hg) concentrations (ppm dry weight).

Species/Relationship	Regression model	F (df)	p	R ²
Smooth dogfish				
Hg-TL	$\log(\text{Hg}) = 1.47\text{E}^{-3} \times \text{TL} - 0.763$	19.56 (1, 53)	< 0.0001	0.273
Hg-Age	$\log(\text{Hg}) = 0.115 \times \text{Age} + 0.200$	17.74 (1, 53)	< 0.0001	0.252
Hg- $\delta^{15}\text{N}$	$\text{Hg} = 0.516 \times \delta^{15}\text{N} - 3.841$	2.53 (1, 40)	0.120	0.061
Hg- $\delta^{13}\text{C}$	$\text{Hg} = 1.205 \times \delta^{13}\text{C} + 23.32$	4.10 (1, 40)	< 0.05	0.095
Spiny dogfish				
Hg-TL	$\log(\text{Hg}) = 1.99\text{E}^{-3} \times \text{TL} - 1.332$	49.14 (1, 123)	< 0.0001	0.287
Hg-Age	$\log(\text{Hg}) = 0.081 \times \text{Age} - 0.696$	56.70 (1, 123)	< 0.0001	0.317
Hg- $\delta^{15}\text{N}$	$\text{Hg} = 0.555 \times \delta^{15}\text{N} - 5.276$	72.16 (1, 105)	< 0.0001	0.410
Hg- $\delta^{13}\text{C}$	$\text{Hg} = 0.342 \times \delta^{13}\text{C} + 8.654$	21.82 (1, 105)	< 0.0001	0.173
Little skate				
Hg-DW	$\log(\text{Hg}) = 4.56\text{E}^3 \times \text{DW} - 1.698$	18.51 (1, 172)	< 0.0001	0.098
Hg-Age	$\log(\text{Hg}) = 0.050 \times \text{Age} - 0.792$	9.66 (1, 172)	< 0.005	0.054
Hg- $\delta^{15}\text{N}$	$\text{Hg} = 0.153 \times \delta^{15}\text{N} - 1.439$	15.01 (1, 87)	< 0.0005	0.149
Hg- $\delta^{13}\text{C}$	$\text{Hg} = 0.231 \times \delta^{13}\text{C} + 4.203$	8.93 (1, 87)	< 0.005	0.094
Winter skate				
Hg-DW	$\log(\text{Hg}) = 6.93\text{E}^{-4} \times \text{DW} - 0.849$	13.24 (1, 147)	< 0.0005	0.083
Hg-Age	$\log(\text{Hg}) = 0.020 \times \text{Age} - 0.750$	11.95 (1, 147)	< 0.001	0.076
Hg- $\delta^{15}\text{N}$	$\text{Hg} = 0.044 \times \delta^{15}\text{N} - 0.238$	4.41 (1, 79)	< 0.05	0.054
Hg- $\delta^{13}\text{C}$	$\text{Hg} = 3.48\text{E}^{-2} \times \delta^{13}\text{C} + 0.893$	0.58 (1, 79)	0.450	0.007
Scup				
Hg-TL	$\log(\text{Hg}) = 3.29\text{E}^{-3} \times \text{TL} - 1.283$	338.6 (1, 125)	< 0.0001	0.732
Butterfish				
Hg-TL	$\log(\text{Hg}) = 3.63\text{E}^{-3} \times \text{TL} - 1.636$	58.38 (1, 86)	< 0.0001	0.407
Squid				
Hg-ML	$\log(\text{Hg}) = 2.86\text{E}^{-3} \times \text{ML} - 1.246$	57.18 (1, 83)	< 0.0001	0.411
Cancer crab				
Hg-CW	$\log(\text{Hg}) = 7.36\text{E}^{-3} \times \text{CW} - 1.399$	86.35 (1, 40)	< 0.0001	0.689

Table 5

Summary statistics for small-sample, bias-corrected Akaike's Information Criterion (AIC_c) and Akaike's weights (w_i) used to select optimal multivariate regression models. Regression analyses were used to predict dogfish and skate total mercury (Hg) concentrations (ppm dry weight) and stable nitrogen ($\delta^{15}N$) and carbon ($\delta^{13}C$) isotope signatures as function of several explanatory variables, including body size [mm; total length (TL) and disk width (DW)], time of catch (year and day of collection), and location of catch [decimal degrees; latitude (Lat) and longitude (Long)]. Positive (+) and negative (-) symbols in parentheses after each explanatory variable denote their level of effect on each response variable, and "NS" is not significant.

Species/Response variable	Optimal model	F (df)	p	R ²	AIC _c	w _i
Smooth dogfish						
Hg	TL (+)	19.56 (1, 53)	< 0.0001	0.273	-153.3	0.56
$\delta^{15}N$	Lat (+)	5.74 (2, 40)	< 0.01	0.232	-33.6	0.51
$\delta^{13}C$	Lat (+) / Day (+)	2.95 (2, 40)	< 0.01	0.243	-85.3	0.87
Spiny dogfish						
Hg	TL (+) / Day (-) / Year (-)	19.56 (3, 123)	< 0.0001	0.328	-432.5	0.74
$\delta^{15}N$	TL (+) / Long (-) / Day (+)	21.56 (3, 105)	< 0.0001	0.388	-122.1	0.67
$\delta^{13}C$	Lat (NS)	0.72 (1, 105)	0.398	0.007	-63.3	0.40
Little skate						
Hg	DW (+)	18.51 (1, 172)	< 0.0001	0.098	-504.8	0.97
$\delta^{15}N$	Day (+) / Lat (+) / Long (-) / DW (+)	11.09 (4, 87)	< 0.0001	0.348	-100.2	0.70
$\delta^{13}C$	DW (+) / Long (-)	5.55 (2, 87)	< 0.01	0.116	-190.5	0.70
Winter skate						
Hg	DW (+) / Year (+)	9.10 (2, 147)	< 0.0005	0.112	-498.1	0.81
$\delta^{15}N$	DW (+) / Long (-) / Day (+)	68.95 (3, 79)	< 0.0001	0.731	-96.4	0.51
$\delta^{13}C$	DW (-) / Year (+) / Day (+)	15.97 (1, 79)	< 0.0001	0.387	-149.8	0.82

Table 6

Summary statistics for analysis of covariance (ANCOVA) models used to examine the effect of sex and species on intra- and inter-specific total mercury bioaccumulation rates (ppm dry weight as function of age).

Factor	Age × Sex		Age		Sex	
	<i>F</i> (df)	<i>p</i>	<i>F</i> (df)	<i>p</i>	<i>F</i> (df)	<i>p</i>
Smooth dogfish	1.63 (1)	0.208	17.25 (1)	< 0.0001	0.08 (1)	0.778
Spiny dogfish	1.17 (1)	0.282	53.44 (1)	< 0.0001	1.65 (1)	0.202
Little skate	0.38 (1)	0.539	17.75 (1)	< 0.0001	7.82 (1)	< 0.01
Winter skate	0.59 (1)	0.443	10.43 (1)	< 0.005	0.52 (1)	0.473

Factor	Age × Species		Age		Species	
	<i>F</i> (df)	<i>p</i>	<i>F</i> (df)	<i>p</i>	<i>F</i> (df)	<i>p</i>
Dogfishes	2.01 (1)	0.158	67.68 (1)	< 0.0001	168.02 (1)	< 0.0001
Skates	3.53 (1)	0.061	17.09 (1)	< 0.0001	35.55 (1)	< 0.0001

Table 7

Stomach contents of smooth dogfish and spiny dogfish. Values represent the percent frequency of occurrence (%F), percent weight (%W), alimentary index (%IA; eqn. 1), and the mean (± 1 SD) stomach content weight relative to whole-body wet weight and number of unique prey per stomach. The mean (± 1 SD) of intact prey body size is also reported, and include total length (fish), carapace length (crab), carapace width (lobster) and mantle length (squid). Squared brackets indicate major taxon sub-totals for %W and %IA.

Prey Taxon	Smooth dogfish				Spiny dogfish			
	%F	%W	%IA	Size	%F	%W	%IA	Size
Fish		[32.3]	[22.5]			[64.7]	[64.5]	
Butterfish (<i>Peprilus triacanthus</i>)	2.9	1.5	0.1	6.0 \pm 0	42.3	42.0	48.4	10.8 \pm 1.9
Scup (<i>Stenotomus chrysops</i>)	5.9	6.0	0.6	-	17.3	3.9	1.9	9.4 \pm 2.3
Silver hake (<i>Merluccius bilinearis</i>)	0	0	0	-	5.8	3.7	0.6	22.0 \pm 0
American eel (<i>Anguilla rostrata</i>)	0	0	0	-	1.9	1.8	0.1	40.6 \pm 0
Searobin (<i>Prionotus</i> sp.)	0	0	0	-	1.9	0.7	0.03	-
Smallmouth flounder (<i>Etropus microstomus</i>)	0	0	0	-	1.9	0.2	0.01	-
Sand lance (<i>Ammodytes americanus</i>)	0	0	0	-	1.9	0.1	0.01	10.2 \pm 0
Herring (<i>Alosa</i> spp.)	0	0	0	-	1.9	0.04	<0.01	-
Skate (Rajidae)	5.9	4.1	0.4	-	0	0	0	-
Spiny dogfish (<i>Squalus acanthias</i>)	2.9	0.8	0.04	-	0	0	0	-
Unidentified fish	64.7	19.9	21.4	-	40.4	12.3	13.5	-
Crustaceans	-	[64.8]	[76.5]	-	-	[5.3]	[1.9]	-
Cancer crab (<i>Cancer</i> spp.)	82.4	54.7	74.6	5.5 \pm 2.6	13.5	5.2	1.9	7.6 \pm 3.6
Lobster (<i>Homarus americanus</i>)	8.8	3.0	0.4	8.3 \pm 3.9	0	0	0	-
Hermit crab (<i>Pagurus</i> spp.)	8.8	0.8	0.1	2.3 \pm 0	0	0	0	-
Mud crab (<i>Panopeus herbstii</i>)	2.9	2.2	0.1	6.7 \pm 0	0	0	0	-
Spider crab (Majidae)	2.9	0.8	0.4	4.6 \pm 0	0	0	0	-
Isopod (Isopoda)	5.9	0.2	0.02	4.5 \pm 0	0	0	0	-
Amphipod (Amphipoda)	2.9	<0.01	<0.01	-	3.9	<0.01	<0.01	-
Mud shrimp (Thalassinidea)	5.9	0.2	0.02	-	0	0	0	-
Unidentified crabs	29.4	2.5	1.2	-	0	0	0	-
Unidentified crustaceans	5.9	0.5	0.05	-	3.9	0.01	<0.01	-
Molluscs	-	[0.4]	[0.03]	-	-	[27.3]	[31.3]	-

Prey Taxon	Smooth dogfish				Spiny dogfish			
	%F	%W	%IA	Size	%F	%W	%IA	Size
Longfin squid (<i>Doryteuthis pealeii</i>)	5.9	0.3	0.03	9.3 ± 5.7	42.3	27.2	31.3	16.8 ± 4.6
Shortfin squid (<i>Illex</i> sp.)	0	0	0	-	1.9	0.1	<0.01	5.5 ± 0.0
Moon snail (Naticidae)	2.9	0.1	<0.01	-	0	0	0	-
Worms	-	[0.5]	[0.2]	-	-	[0]	[0]	-
Polychaete (Polychaeta)	17.7	0.5	0.2	-	0	0	0	-
Ribbon worm (Nemertea)	2.9	<0.01	<0.01	-	0	0	0	-
Cnidarians – Hydrozoa	2.9	0.02	<0.01	-	0	0	0	-
Animal remains	26.5	1.9	0.8	-	30.8	2.7	2.3	-
Eelgrass/Algae/Detritus	11.8	0.05	0.01	-	1.9	<0.01	<0.01	-
Sediment	8.8	0.04	<0.01	-	5.8	0.1	<0.01	-
Stomachs examined (n)	34				57			
Stomach content weight/whole-body weight (%)	1.70 ± 1.00				2.76 ± 2.78			
Number of unique prey/stomach	3.47 ± 1.83				2.00 ± 1.12			

Table 8

Stomach contents of little skates and winter skates. Values represent the percent frequency of occurrence (%F), percent weight (%W), alimentary index (%IA; eqn. 1), and the mean (± 1 SD) stomach content weight relative to whole-body wet weight and number of unique prey per stomach. The mean (± 1 SD) of intact prey body size is also reported, and include total length (fish), carapace width (crab), carapace length (lobster) and mantle length (squid). Squared brackets indicate major taxon sub-totals for %W and %IA.

Prey Taxon	Little skate				Winter skate			
	%F	%W	%IA	Size	%F	%W	%IA	Size
Fish	-	[3.8]	[0.2]	-	-	[35.1]	[4.0]	-
Butterfish (<i>Peprilus triacanthus</i>)	1.7	3.1	0.1	6.8 \pm 0	2.2	4.6	0.4	12.1 \pm 4.0
Sand lance (<i>Ammodytes americanus</i>)	0	0	0	-	4.4	15.4	2.4	11.0 \pm 1.4
Scup (<i>Stenotomus chrysops</i>)	0	0	0	-	2.2	13.6	1.0	12.1 \pm 4.7
Herring (<i>Alosa</i> spp.)	0	0	0	-	1.1	0.5	0.02	-
Unidentified fish	9.2	0.7	0.1	-	5.6	1.1	0.2	-
Crustaceans	-	[64.4]	[68.9]	-	-	[35.5]	[51.0]	-
Amphipod (Amphipoda)	83.3	29.5	50.6	-	74.4	15.6	40.4	-
Isopod (Isopoda)	10.8	0.4	0.1	-	16.7	2.3	1.3	-
Sand shrimp (<i>Crangon septemspinosa</i>)	55.8	7.4	8.5	-	34.4	3.6	4.3	-
Pandalid shrimp (Pandalidae)	5.0	1.7	0.2	-	3.3	0.1	0.01	-
Hooded shrimp (Cumacea)	4.2	0.04	< 0.01	-	6.7	0.03	0.01	-
Mud shrimp (Thalassinoida)	3.3	2.3	0.2	-	2.2	1.7	0.1	-
Grass shrimp (Hippolytidae/Palaemonidae)	4.2	1.1	0.1	-	2.2	0.1	0.01	-
Cancer crab (<i>Cancer</i> spp.)	25.0	7.7	4.0	0.8 \pm 0.4	13.3	6.3	2.9	1.8 \pm 0.8
Hermit crab (<i>Pagurus</i> spp.)	0.8	0.01	< 0.01	-	0	0	0	-
Lobster (<i>Homarus americanus</i>)	1.7	0.9	0.03	2.7 \pm 0	2.2	0.9	0.1	2.0 \pm 0
Unidentified shrimp	10.0	1.9	0.4	-	12.2	0.4	0.2	-
Unidentified crabs	22.5	7.0	3.2	-	10.0	3.1	1.1	-
Unidentified decapods	5.0	1.5	0.2	-	1.1	0.5	0.02	-
Unidentified crustaceans	25.0	3.2	1.6	-	20.0	1.1	0.7	-
Molluscs	-	[1.7]	[0.05]	-	-	[6.2]	[1.0]	-
Bivalves (Bivalvia)	7.5	0.1	0.02	-	11.1	0.2	0.1	-
Longfin squid (<i>Doryteuthis pealeii</i>)	0.8	1.2	0.02	12.8 \pm 0	4.4	6.0	0.9	10.0 \pm 4.4

Prey Taxon	Little skate			Winter skate					
	%F	%W	%IA	Size	%F	%W	%IA	Size	
Shortfin squid (<i>Illex</i> sp.)	0.8	0.4	0.01	-	0	0	0	-	
Worms	-	[16.8]	[7.6]	-	-	[10.8]	[21.3]	-	
Polychaete (Polychaeta)	50.8	16.8	17.6	-	57.8	10.6	21.3	-	
Ribbon worm (Nemertea)	0	0	0	-	1.1	0.1	<0.01	-	
Peanut worm (Sipunculoidea)	0	0	0	-	1.1	0.1	<0.01	-	
Round worm (Nematoda)	0.8	<0.01	<0.01	-	0	0	0	-	
Porifera – Sponge	0.8	<0.01	<0.01	-	1.1	<0.01	<0.01	-	
Cnidarians – Hydrozoa	0	0	0	-	1.1	<0.01	<0.01	-	
Animal remains	50.0	12.7	13.1	-	53.3	12.2	22.6	-	
Eelgrass/Algae/Detritus	3.3	0.3	0.02	-	3.3	0.02	<0.01	-	
Sediment	15.0	0.2	0.1	-	22.2	0.2	0.1	-	
Stomachs examined (n)	125			92					
Stomach content weight/whole-body weight (%)	0.35 ± 0.30			0.37 ± 0.33					
Number of unique prey/stomach	4.58 ± 2.65			4.24 ± 2.80					