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"METHYLMERCURY LEVELS IN COMMERCIALLY HARVESTED SPINY DOGFISH (*SQUALUS ACANTHIUS*) FROM OFF THE COAST OF MASSACHUSETTS"

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DATA ACCESSIBILITY STATEMENT

The data that support the findings of this study are openly available in the Harvard Dataverse at https://doi.org/10.7910/DVN/OEQ9PB.

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1. ABSTRACT

Spiny Dogfish are small sharks that are harvested for seafood, primarily off the Eastern Seaboard of the United States. For the purposes of establishing seafood consumption advisories with regards to methylmercury (MeHg) content, the US Food and Drug Administration (FDA) classifies Spiny Dogfish as "sharks", an overly-broad taxonomic grouping with a mean MeHg content of 0.98 μg/g (wet weight). Because the FDA mean includes fishes of different species, collected internationally over the last four decades, there is reason to believe that the FDA mean value is not reflective of the fish harvested in the Spiny Dogfish fishery. To evaluate how closely Spiny Dogfish match the values in the FDA's generic "shark" category, we collected muscle samples from 102 commercially harvested Spiny Dogfish caught off Cape Cod, Massachusetts in the 2018 fishing season. Among the fish we sampled, 100% had MeHg concentrations lower than the FDA "shark" mean, and 77.5% had MeHg concentrations that the FDA considers safe for consumption at a frequency of once per week. The mean MeHg concentration in our samples was 0.378 μg/g (wet weight), less than half of the value for the generic "shark" category. Moreover, through a comparison with data from the 1970s, we document a 30% decline in MeHg concentrations for select size categories of Spiny Dogfish off the coast of New England. Exposure simulations indicate that, to minimize risk, Spiny Dogfish should be consumed no more than twice per month by most people.

2. INTRODUCTION

Spiny Dogfish *Squalus acanthias* are small sharks, which are widely distributed along the continental shelves of Europe, Africa, Australia, South America, southern Asia, and eastern North America (Carpenter 2018). These fish are taxonomically distinct from the Pacific Spiny Dogfish *Squalus suckleyi*, which is found in the northern Pacific Ocean; however, that distinction has only been made recently (Ebert et al. 2010). A large fishery for Spiny Dogfish exists in the United States, where landings of the species have increased over the last decade (ACCSP 2019a). While Spiny Dogfish landed in the United States is used for both human consumption and the reduction industry (e.g., pet food and fertilizer), food fish is predominantly landed by the gillnet fleet out of Southern New England, and fish destined for the reduction industry is predominantly landed by the bottom trawl fleet fishing out of North Carolina (Maurer 2017). The Commonwealth of Massachusetts, in Southern New England, has accounted for 46% of landings of this species on the Atlantic Coast from 2007-2017 (between 32% and 55% annually; ACCSP

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2019b). The majority of those landings (> 80%) occur in Chatham, MA (S. Reed, Massachusetts Division of Marine Fisheries, *pers. comm.*). In Massachusetts, Spiny Dogfish make up ~7% of finfish landings by weight (ACCSP 2019b). The Spiny Dogfish fishery is primarily prosecuted by day-boat operators using longlines and gillnets and thus, provides a source of revenue and employment for smaller ports, particularly during the summer (Hall-Arber et al. 2001).

Spiny Dogfish have a low market value in New England relative to groundfish such as Atlantic Cod *Gadus morhua* and Haddock *Melanogrammus aeglefinus* (Didden 2018). Despite this, Spiny Dogfish presents an alternative to traditional multispecies fisheries in the region because of substantial shifts in biomass from an ecosystem dominated by gadids and flounders to one dominated by small elasmobranchs since the 1980s (Fogarty and Murawski 1998; Frisk et al. 2008). Additionally, climate change is projected to reinforce these biomass shifts as the range of Spiny Dogfish expands and the range of gadids contracts (Nye et al. 2009). For all of these reasons, the Spiny Dogfish fishery will likely remain important for coastal New England in the coming decades.

Spiny Dogfish meat is particularly popular for human consumption in the European Union (EU), and over 90% of the fried fish sold by chip shops in the United Kingdom is Spiny Dogfish (Hobbs et al. 2019). Because the Northeast Atlantic (European) stock of Spiny Dogfish is considered to be endangered, almost no landings of these fish are allowed in European fisheries (ICES 2018). Thus, nearly all EU demand must be met through imports, which included over 5 kilo metric tons of Spiny Dogfish meat in 2018, and over 128 kilo metric tons in the last decade (EUMOFA 2019), predominantly from the Northwest Atlantic Ocean (Dell'Apa et al. 2013). Spiny Dogfish meat is not as popular in the United States, which may be partially due to public concern with the elevated mercury (Hg) levels documented in sharks (FDA 2012). Additionally, the grouping of all shark species in both consumption advisories (EPA and FDA 2019) and large summary studies of Hg intake through fish consumption (Sunderland et al. 2018) can create a perception of uncertainty around Hg levels in a particular species.

Mercury is a naturally occurring element that is introduced to the environment through natural and human activities (Mason et al. 2012). Average environmental levels of Hg are thought to have tripled as a result of industrialization since the middle of the 19th century (Mason et al. 2012; Lamborg et al. 2014). While Hg emitted into the atmosphere can be distributed globally, Hg deposition hotspots can also develop due to local sources such as industries that use

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Hg (e.g. battery production and gold mining) or industries where Hg is a byproduct (e.g. coal combustion; Evers et al. 2014). While Hg emissions have been declining in North America and Western Europe, they have been increasing in rapidly developing East Asian nations, especially in China (Streets et al. 2019). Wet deposition of Hg is the primary source by which Hg enters the ocean (Driscoll et al. 2013). A fraction of Hg occurs in seawater as monomethylmercury (MeHg), a form that is efficiently entrained in pelagic food webs, resulting in a buildup of MeHg in marine biota (Chen et al. 2014).

Levels of MeHg in apex predators can be up to eight orders of magnitude higher than in seawater. Because MeHg biomagnifies with each trophic level, upper-level piscivores such as tunas, sharks, seals, and some seabirds are at risk of elevated exposure to this toxic compound. Moreover, assimilated MeHg is lost from animals at a very low rate, resulting in high concentrations in long-lived fish. Therefore, the magnitude of MeHg accumulation in fish can be influenced by age, diet, physiology, ecology, and geographic location, among other factors (Storelli et al. 2001; Teffer et al. 2014; Baumann et al. 2017).

Concerns over MeHg concentrations in Spiny Dogfish occur because this species fits into several of the categories of concern identified above. First, they are thought to be long-lived. A variety of methods have been used to estimate the ages of Spiny Dogfish in the past, including visual evaluation of dorsal spines (Orlov et al. 2011), sectioning dorsal spines (Soldat 1982), bomb radiocarbon aging of dorsal spines (Campana et al. 2006), and histologically stained vertebrae (Bubley et al. 2012). Although producing accurate age estimates from these fish is challenging (Natanson et al. 2018a, 2018b; Harry 2018), many published estimates suggest these fish can live for several decades (Stehlik 2007; Orlov et al. 2011).

Second, Spiny Dogfish are predatory. The content of their diet varies seasonally with the availability of prey, but off the coast of New England a large proportion of it in any given year is made up of small pelagic species such as Atlantic Mackerel *Scomber scombris*, Atlantic Herring *Clupea harengus*, squid (*Illex illecebrosus* or *Loligo pealeii*), and ctenophores (Smith and Link 2010). Finally, the physiology of elasmobranchs lends itself to longer MeHg retention because of the low rate of bile production, and thus, slow excretion of contaminants (Ballatori and Boyer 1986). This combination of slow excretion and high trophic position can lead to an accumulation of Hg in the flesh of Spiny Dogfish (Adams and McMichael 1999; Taylor et al. 2014; St. Gelais and Costa-Pierce 2016).

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From a human health perspective, MeHg exposure is of concern when intake exceeds the reference dose of 0.1 μg MeHg per kg of human body weight per day, or 0.7 μg MeHg per kg of human body weight per week (EPA 2001). Typically, MeHg exposure is thought to be a concern for people who consume large quantities of predatory fishes (Hightower and Moore 2003). A recent analysis of seafood consumption in the United States shows that although tuna accounts for 18.8% of American seafood consumption by weight, it accounts for 37.8% of MeHg intake nationally, while shrimp accounts for only 9.7% of MeHg intake despite higher (19% of total seafood) consumption by weight (Sunderland et al. 2018). Because of the well-known health problems associated with MeHg exposure (Grandjean et al. 1997; ATSDR 1999), it is important to define the exposure levels that could exist given the expected range of MeHg concentrations in fishes available on the market, consumption frequency (i.e. number and size of meals) and consumer body mass, as guided by the EPA reference dose of 0.1 μg per kg of human body weight per day (EPA 2001).

Qualitative evaluations of purchaser preference indicates that concern over MeHg contamination is one of several impediments to developing large-scale markets for Spiny Dogfish in the United States (J. Francolini, independent marketing consultant, pers. comm.). The United States Food and Drug Administration (FDA) and the Environmental Protection Agency (EPA) joint standards for MeHg exposure recommend consumers of all ages avoid eating fish with a mean concentration of Hg > 0.46 μg/g in wet tissue (EPA and FDA 2019; Hg as MeHg). These standards are meant to provide guidance across a wide range of fishes and locales, so they often do not separate out individual species and rely on broad taxonomic categories such as "crab", "clam", "shark", and "skate". Additionally, geographic origin is not considered but may impact contamination levels (Baumann et al. 2017). The most recent information available through the FDA databases places Spiny Dogfish in the "shark" category (Cladis et al. 2014) with an estimated average Hg concentration of 0.98 μg/g in wet tissue (FDA 2012). Such a broad grouping is likely uninformative, given the diversity of shark species whose longevity, growth rates, habitats, diets, ingestion rates migrations, reproduction, and metabolism can be highly variable, and thus influence the levels at which MeHg concentrates in the muscle tissue.

We have therefore undertaken this study to provide information on the current concentrations of MeHg in Spiny Dogfish that are landed for human consumption by the commercial fishing industry in eastern Massachusetts (which represents a substantial portion of

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the global catch), investigate how patterns of MeHg concentration relate to fish size, condition, and sex, and relate those concentrations to human exposure considering the variability in meal sizes, frequency of consumption, and human body masses as well as MeHg concentrations. Given the concerns around aging methods described above, we dispensed with age estimation for this study. Additionally, we compare our results with previous research conducted on MeHg in Spiny Dogfish off the coast of New England.

3. METHODS

3.1. *Sample Collection*

Spiny Dogfish sampled during this study were captured as part of normal, commercial fishing operations by the day boat fleet operating out of the Chatham Fish Pier (Chatham, MA), and sampling was timed to match the regular commercial fishing season. The proportion of samples matched the proportion of landings ($X^2 < 2.36$, $p > 0.12$) in all months except for October ($X^2 = 6.42$, $p = 0.01$), during which time, a higher proportion of fish were landed than sampled. All captures took place in NOAA Statistical Area 521 (West Side South Channel) or Statistical Area 537 (Off No Man's Land). Participating vessels fished using fixed gear (either longlines or gillnets). Researchers met commercial harvesters dockside to collect whole fish and record fishing location (when available, Figure 1). All fish were processed on the same day they were captured. Each fish was sexed by observing the presence or absence of claspers as the diagnostic characteristic, and measured for total length (TL; cm) and weight (kg). Pre-caudal length (PCL) was back-calculated from total length using the equation $PCL = TL \times 0.807$ (Dell'Apa et al. 2013). Fulton's K was calculated for each fish as $K = \frac{L}{W^3} \times (1 \times 10^5)$ where K is the condition factor of the fish, L is the pre-caudal length of the fish (in mm), and W is the weight of the fish (in g; Pope and Kruse 2007). Following external assessment, a tissue sample was excised from the dorsal-axial musculature. We removed the skin from each tissue sample and placed the samples into individually labeled borosilicate vials for storage at -18 – -20°C. At the conclusion of the sample collection period, all samples were shipped frozen to the University of Connecticut for further processing and Hg analyses.

3.2. Mercury Analysis

Concentrations of total mercury (THg) were determined as previously described by Taylor et al. (2014) with slight modifications regarding quality check and assurance (QC/QA),

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which employed typical practices of the University of Connecticut chemical ecology laboratory. Wet tissues were weighed before and after freeze-drying to determine a wet weight / dry weight conversion factor for each individual. Freeze-dried fish muscle tissues were homogenized to a consistency as close to a powder as possible. Elasmobranch muscle tissue has a high lipid content (Reum 2011), making the muscle pasty when homogenizing with a BeadBug (Benchmark Scientific, Sayreville, NJ). Homogenized muscle samples were used for analysis of total mercury (THg) on a MA-3000 Hg analyzer (Nippon Instruments Corporation, Tokyo, Japan).

About 11% (n=11) of samples were analyzed in triplicate to provide a more accurate estimate of the error structure associated with the analysis. Moreover, analysis of each triplicate sample was followed with a blank consisting of deionized water and a certified reference material (CRM). In addition to THg analysis, a subset of samples (n = 16) was analyzed for MeHg. Analysis of MeHg was conducted to ensure that THg was an appropriate proxy for MeHg in this species (Kim et al. 2016). Small sample masses (10-20 mg) were digested in 4.5 N nitric acid aqueous solution overnight at 60°C in trace metal cleaned, 30 mL Teflon digestion vessels (Mason et al. 2019). Aliquots equal to 30 µL of digest were resuspended in deionized water. A pH between 4.0 and 5.0 is required for sodium tetraethylborate (NaBET) to ethylate MeHg for the analysis, so the pH values of dilute tissue digests were adjusted by addition of potassium hydroxide solution (8N) and buffered with acetate buffer (pH = 4.2) to reach a pH of 4.7. The total volume of solution in borosilicate analysis vials was 30 ± 0.1mL. Detection of MeHg relied on cold vapor atomic fluorescence (CVAFS) using a Tekran 2700 detection system coupled with an autosampler (Tekran Instruments Corporation, Toronto, Canada). Quality check and assurance were conducted by analyzing blanks, certified reference material (DORM-2) and analytical replicates. Standards were also checked throughout the run to assure the constancy of calibration, which was conducted at the beginning of the analysis using Alfa Aesar MeHg aqueous standards (Alfa Aesar, Tewksbury, MA).

Analysis of DORM-2 standards for THg (n=9) resulted in mean concentration of 4.39 ± 0.05 µg/g and 4.08 ± 0.26 µg/g for MeHg (n=3), within the certified average ± standard deviation values for THg (4.470 ± 0.032 µg/g) and MeHg (4.06 ± 0.09 µg/g). A total of 102 individual muscle tissues were analyzed for THg with 16 samples in triplicate producing mean relative standard deviations of < 5.6%. For the MeHg analysis, two samples were analyzed in triplicate

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producing mean relative standard deviations of ±14%. One muscle tissue sample was weighed and digested in duplicate to account for a compound error due to weighing, analysis etc. and resulted in an error of ±7%.

We determined percent water content for each individual and used the formula $Hg_{wet} = Hg_{dry} \times \frac{\%\,dry}{100\%}$ to back-calculate wet-weight ppm MeHg, the metric commonly used for consumer-facing food safety advisories.

3.3. Data analysis

We used Wilcoxon-Mann-Whitney tests to compare size, Fulton's K, and THg content of male and female Spiny Dogfish. THg concentration (µg/g) in wet tissue was log-transformed and modeled as a function of fish total length (cm), weight (kg), and capture date using generalized linear models (Fabris et al. 1992). Model fit was evaluated using corrected Akaike Information Criterion (AICc) scores. To make our results comparable to the FDA and EPA advisories, we followed FDA and EPA protocols (U.S. EPA 2017) and computed a bootstrapped 95% confidence interval for the mean THg content of Spiny Dogfish. Following the methods of Taylor et al. (2014), we also modeled THg concentration as a function of length using least-squares regression. We separated fish into three length categories (Table) following the categories of Greig et al. (1977) to compare THg concentrations off Southern New England today to those from Georges Bank in the early 1970s using t-tests. To test the representativeness of our sample, we used the sample size mean calculation function from the samplingbook package in R (Manitz et al. 2017).

3.4. Consumption Model

We simulated THg exposure from Spiny Dogfish consumption for a combination of different serving sizes and body weights (Table) eating between one and five meals per month. Serving sizes follow the EPA / FDA guidelines for seafood portions, such that children up to 5 years old should have 1 oz portions, children between 6 and 8 years old should have 2 oz portions, children from 9 to 11 years old should have 3 oz portions, and anyone over 11 should have 4 oz portions (U.S. EPA 2017). For each combination of variables in Table , we calculated exposure at one meal by randomly selecting a THg concentration (µg/g wet) from our dataset and multiplying it by the serving size of meat consumed (g) to get total mass of ingested THg (µg) for each meal. Hg consumption was summed across the meals consumed in a given month to get total mass of THg (µg) consumed over the course of the month. This process was repeated

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100,000 times for each combination of meals consumed, body weight, and serving size and compared with the monthly exposure limit for each body weight as calculated using the EPA / FDA technical information page (U.S. EPA 2017) and the equation $\text{Total exposure limit } (\mu\text{g}) = 0.1 \left(\frac{\mu\text{g/kg}}{\text{day}}\right) \times \text{W (kg)} \times 30.41 \frac{\text{days}}{\text{month}}$. All analyses were conducted using in R v3.4.2 (R Core Team 2017) using $\alpha = 0.05$ as the threshold for statistical significance. The following R packages were used; qcpR (Spiess 2018), lubridate (Grolemund 2014), boot (Canty and Ripley 2017), and glm2 (Marschner and Donoghoe 2018).

4. RESULTS

4.1. Fish sampling

We sampled 102 Spiny Dogfish from commercial fishing vessels landing at the Chatham Fish Pier between June 13 and October 4, 2018. Of those, 80.6% were female and 19.4% were male. Female fish had a larger average total length (864.7 ± 53.2 mm) than male fish (755.0 ± 37.5 mm; $W = 92.5$, $p < 0.001$; Figure 2). Condition factor ranged from 0.642 to 2.030 ($\mu = 1.010$, $\sigma = 0.170$) and there was no difference between sexes ($W = 839$, $p = 0.277$). At least 83% of the females were mature and had candles or pups.

4.2. Mercury measurement

THg and MeHg concentrations were tightly related ($p < 0.001$, $R^2 = 0.891$, Figure). MeHg accounted for between 94.4% and 131% of THg with an average of 111%. Numbers higher than 100% are likely the result of compounded measurement error in the instruments used. The analytical error for Hg recovery from Spiny Dogfish tissue was 10.4%. Additionally, the analytical error on MeHg measurements was 13.94%, and the analytical error on THg measurements 1.1%. Therefore, the compounded error for THg measurements could be as substantial as -11.38% or +11.61%, and the compounded error for MeHg measurements could be as substantial as -14.89% or +25.79%. Effectively, all Hg present was in the form of MeHg.

Percent water content in the tissue ranged from 62 to 80% (median: 72%). Spiny Dogfish sampled in this study had an average THg concentration of 0.378 μg/g (± 0.164 μg/g) in wet tissue and 1.373 μg/g (± 0.591 μg/g) in dry tissue. Wet concentrations ranged from 0.13 μg/g to 0.86 μg/g and bootstrapped 95% confidence bounds (1 million replicates) of the mean were 0.348 μg/g and 0.411 μg/g. There were no significant differences in THg concentrations between female and male Spiny Dogfish ($W = 592.5$, $p = 0.172$). Generalized linear models (GLMs)

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indicated that the most consistent and significant predictor of THg was not length, but weight (Table). In the best-fitting model, weight was positively associated with THg concentrations, such that heavier fish had higher concentrations of Hg. No other predictor variables were significant in the best-fitting model, and the best-fitting model had a relative likelihood of 0.582. An additional model with ΔAICc = 0.927 indicated that male fish may have higher THg concentrations at a given size than females, and had a relative likelihood of 0.366. No other models had a low enough ΔAICc value ($\Delta AICc \leq 2$) to qualify as candidates, and most had relative likelihoods of 0 (Table).

4.3. Comparison with earlier research

When comparing the results of this study with the results of Greig et al. (1977; Table 2), we found no difference in THg between the two studies for fish between 755 mm and 854 mm (t = 0.819, df = 33.829, p < 0.209). However, smaller fish (< 755 mm) had a 34.3% lower average THg concentration in 2018 (t = 2.631, df = 22.787, p = 0.007), and larger fish (> 854 mm) had a 31.7% lower average THg concentration in 2018 (t = 2.957, df = 29.444, p = 0.003).

We also compared the findings of our study with the findings of Taylor et al. (2014). Generally, fish from our study had higher condition factors (W = 2310, p < 0.001), although the distributions of fish lengths sampled in both studies were similar (W = 7099, p = 0.142). Following the methods of Taylor et al. (2014), we used least squares exponential-regression to estimate THg concentrations (µg/g dry weight) as a function of pre-caudal length (mm; L) at THg = $e^{-0.00199 \times L - 1.3322}$. Our research found THg = $e^{-0.00190 \times L - 1.05968}$ (Figure 4), indicating similar relationships between the two studies.

4.4. Representativeness of sample

The “samplingbook” package in R provides a simple interface for implementing many of the statistical tests described in Kauermann and Küchenhoff (2011) that can be used for experimental design or post-hoc testing. We used the sample.size.mean function post-hoc to calculate how many samples would be necessary to estimate the mean Hg concentration of our target population (commercially harvested Spiny Dogfish landed in Chatham, MA). Using a desired precision of 0.1724 µg/g dry weight (~0.05 µg/g wet weight), and the standard deviation from our dataset (0.591 µg/g dry weight), we calculated that a sample size of 46 fish would be necessary to characterize the mean Hg concentration found in our target population. Using the same function restructured to calculate precision, our sample size (n=102) is able to estimate the

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population mean concentration of Hg with a precision of 0.033 μg/g wet weight. We also used the same sample.size.mean function to calculate the precision of our mean length estimates for both female fish (11.5 mm) and male fish (16.5 mm) landed in the fishery.

4.5. Mercury consumption simulation

Overall, 22.5% of the fish sampled in our study had MeHg concentrations that would put *them in the EPA-FDA "Choices to Avoid" category (> 0.46 μg/g), 58.8% fell in the "Good* eChoices, 1x per week" category (0.23 – 0.46 μg/g), 14.7% fell in the "Good Choices, 2x per week" category (0.15 – 0.23 μg/g), and 4% fell in the "Best Choices, 3x per week" category. Simulations of MeHg consumption indicated that if no other MeHg was consumed during a given month all age group and serving size combinations could consume up to two meals of Spiny Dogfish per month with a < 5% chance of exceeding the FDA's recommended maximum exposure level (Figure 5, Table 3). Larger individuals within each age group could consume at least three meals per month (up to as many as seven meals per month for the largest adults) with a < 5% chance of exceeding the recommended maximum exposure level (Table 3).

5. DISCUSSION

Our objectives were to characterize MeHg contamination in Spiny Dogfish harvested for human consumption, compare that level against EPA/FDA recommendations and against the results of previous research undertaken in the region. We characterized MeHg contamination in Spiny Dogfish harvested for human consumption by sampling directly from the commercial landings in Chatham, MA throughout the 2018 Spiny Dogfish fishing season. Our sample size (n = 102) exceeded the sample size of 87% of the studies currently used to make MeHg recommendations by the FDA and EPA, which have an average sample size of n = 80 and a median sample size of n = 41. Additionally, our results showed substantially lower MeHg concentrations in Spiny Dogfish than are currently in the FDA / EPA's generic "shark" category, reinforcing the need for more consumer-appropriate taxonomic breakdowns of consumption advisories. We found similar levels of MeHg as were documented by Taylor et al. (2014) and St. Gelais and Costa-Pierce (2016), and lower values than were documented by Greig et al. (1977).

The fish sampled in this study were sampled randomly from the commercial landings in Chatham, Massachusetts, a port that is responsible for a large portion of the Spiny Dogfish fishery destined for human consumption (ACCSP 2019a, S. Reed MADMF, *pers. comm.*). Thus, results should be thought of not as representative of the Spiny Dogfish population overall, but

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portion of the Spiny Dogfish population that is targeted by the fishery. Because of our sampling focus on the commercial food fishery, the sex ratio in this study was skewed heavily towards the larger, more desirable female fish, which are preferentially targeted by the fishery.

Methylmercury (MeHg) accounted for 100% of THg in nearly all samples; therefore, our discussion refers to MeHg, as that is the form of concern for human health. The average wet tissue MeHg concentration measured in Spiny Dogfish during this study (0.378 µg/g, n=102) was substantially lower than that measured in Spiny Dogfish in Italy (6.5 µg/g, n=15; Storelli et al. 2001), Crete (2.07 µg/g, n=47; Kousteni et al. 2006), Southeastern Australia (1.75 µg/g, n=2; Pethybridge et al. 2012), and Korea (0.98 µg/g, n=17; Kim et al. 2016), but similar to values found in Ireland (0.25 µg/g, n=6, small fish only; Domi et al. 2005), Japan (0.354 µg/g, n=75; Endo et al. 2009), and Rhode Island, USA, (~0.3 µg/g depending on the dry to wet conversion factor applied, n=124; Taylor et al. 2014). Global models indicate that Hg deposition hotspots exist in Eastern Europe, Southeast Asia, and Southeastern Australia (Selin et al. 2008), corresponding to regions of highest MeHg in Spiny Dogfish based on the available published data.

In contrast to the majority of findings on sex-based differences in MeHg concentrations in Spiny Dogfish, there was no significant difference in MeHg concentrations between male and female Spiny Dogfish in this and the Taylor et al. (2014) studies where the size range of fish were similar. Age estimation for Spiny Dogfish is not reliable (Natanson et al. 2018a), we therefore could not evaluate whether the absence of sex based MeHg differences was due to potential truncation of the age structure of our sampled population. Additionally, because the fishery is primarily prosecuted with gillnets, our sample size distribution may not be representative of the full range of sizes present in the population at large. However, given the tight precision estimates for both fish length and Hg concentrations, for samples randomly collected across a period of months from eight different commercial fishing vessels, we believe the data are representative of the commercially fished Spiny Dogfish population off Cape Cod. Since landings in Chatham represent nearly half of all U.S. East Coast Spiny Dogfish landings (and the majority of food fish landings, i.e., fish not caught and sold for bait or fertilizer), we believe these data are broadly representative of Spiny Dogfish caught for human consumption on the East Coast of the United States.

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We observed substantial declines in MeHg concentrations from similar research conducted four decades ago. In just over 67% of our Spiny Dogfish, MeHg concentrations in muscle tissue were ~31% lower than measurements taken from similarly sized fish in the early 1970s. Various factors could contribute to changes in MeHg concentrations observed in predators (such as in this study) including ecosystem shifts, food abundance, changes in feeding rates and growth rates, or a change in the regional pool of bioavailable MeHg (Stenhouse et al. 2018; Schartup et al. 2019). It is difficult to draw a direct connection between any of these factors and the resulting MeHg concentrations in fish tissues. For example, at present we are unable to link the atmospheric deposition of Hg into the surface ocean and its relationship to the pool of bioavailable MeHg, the form that is mobile in the marine food web. However, a recent study by Schartup et al. (2019) suggests that an increase in the availability of Atlantic Herring *Clupea harengus* as prey may have resulted in decreased MeHg in the tissue of Spiny Dogfish off the coast of New England from the 1970s to 2000s.

Total global emissions of inorganic Hg have been declining over the last several decades after reaching a global high in the 1970s (Streets et al. 2019). Although the direct connection between atmospheric Hg emissions and bioaccumulation of MeHg in marine food webs remains poorly understood, the decline of MeHg in the meat of Spiny Dogfish and other species from the North Atlantic (e.g. Bluefish *Pomatomus saltatrix*; Cross et al. 2015) lowers the risk of negative impacts in people who consume these and other fish. However, changes in water temperature, prey availability, and Hg emissions under different climate change regimes may lead to increases in MeHg contamination in Spiny Dogfish and other large predators in the coming decades (Schartup et al. 2019) as squids and other warmwater forage species (with higher MeHg content) replace Atlantic Herring as the primary forage base for the ecosystem.

Exposure to MeHg in humans is driven by three factors: body mass, contamination level, and amount of fish consumption. Individuals with smaller body mass (i.e., children) are at high risk for exposure, and face the highest consequences for exposure (Bose-O'Reilly et al. 2010). Health risks also exist for people who frequently consume fish with moderate or high levels of MeHg (Taylor and Williamson 2017; von Stackelberg et al. 2017). Concentrations of MeHg in Spiny Dogfish caught off Cape Cod are in the same FDA category as a variety of other fish species such as Bluefish and Striped Bass *Morone saxatilis* (Sunderland 2007; Cladis et al. 2014)

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that are commonly eaten in the United States and are more widely available in regional fish markets (Masury and Schumann 2019).

The bootstrapped average range of Spiny Dogfish MeHg concentrations documented in this study was also substantially lower than the bootstrapped average range documented in the generic 'shark' classification currently used in the EPA-FDA guidelines (0.35 – 0.41 µg/g vs. 0.91 – 1.05 µg/g) . Data supporting the Federal guidelines includes information from a variety of shark species (e.g., Spiny Dogfish, Smooth Hammerhead Sharks *Sphyrna zygaena*, and Mako Sharks *Isurus oxyrhinchus*) captured around the world between the early 1970s and the present (Karimi et al. 2012). In the FDA databases, shark data is not separated by species (FDA 2012, 2017). Even in the studies that separate sharks by species, most species are represented in the data by fewer than twenty individual animals (Karimi et al. 2012; Cladis et al. 2014). The authors of the most recent study used in the EPA-FDA guidelines (Cladis et al. 2014) recommend that species specific advice should be developed for taxonomic groups where within-group MeHg variability is high, such as sharks, cods, trouts, and sea basses, to better inform consumers.

Our simulations suggest that consumption of Spiny Dogfish meat should be limited to no more than once or twice per month (1 oz – 4 oz portions, as appropriate for an individual's age bracket), especially when other sources of seafood are incorporated into the diet. Additionally, our simulations of MeHg intake indicate that although Spiny Dogfish are not in the "Best Choices 3x per Week" category, they can be a safe source of protein when consumed in moderation and in the appropriate serving size as defined by the FDA (USOFR 2019). Furthermore, the simulation results fall within the same EPA-FDA categorical recommendations as the bootstrapped mean, such that both indicate that Spiny Dogfish can be consumed several times per month (at the appropriate portion size defined by the EPA / FDA; U.S. EPA 2017). Most importantly, our findings illustrate the issues inherent in classifying fish by overly broad taxonomic group (e.g., "shark") under the EPA-FDA standards (Sunderland 2007; Cladis et al. 2014; U.S. EPA 2017; Sunderland et al. 2018). Concentrations of MeHg display large differences between species within these groups due to differences in life histories, feeding ecology, and other factors. Seafood consumers would benefit from information that mirrors the detail found in seafood labeling (e.g., at the species level for Spiny Dogfish, or at the family level for fishes such as anchovies, family Engraulidae).

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6. ACKNOWLEDGEMENTS

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Author Manuscript DRAFT

WORKS CITED

ACCSP. 2019a. Annual summary for 2007-2017 Spiny Dogfish commercial landings by state. Atlantic Coastal Cooperative Statistics Program.

ACCSP. 2019b. Massachusetts commercial landings, 2017. Atlantic Coastal Cooperative Statistics Program.

Adams, D. J., and R. H. McMichael. 1999. Mercury levels in four species of sharks from the Atlantic coast of Florida. Fishery Bulletin 97:372–379.

ATSDR. 1999. Mercury. Page 20. Center for Disease Control, Agency for Toxic Substances and Disease Registry, Public Health Statement 7439-97–6, Washington, D.C.

Ballatori, N., and J. L. Boyer. 1986. Slow biliary elimination of methyl mercury in the marine elasmobranchs, *Raja erinacea* and *Squalus acanthias*. Toxicology and Applied Pharmacology 85(3):407–415.

Baumann, Z., R. P. Mason, D. O. Conover, P. Balcom, C. Y. Chen, K. L. Buckman, N. S. Fisher, and H. Baumann. 2017. Mercury bioaccumulation increases with latitude in a coastal marine fish (Atlantic silverside, *Menidia menidia*). Canadian Journal of Fisheries and Aquatic Sciences 74(7):1009–1015.

Bose-O'Reilly, S., K. M. McCarty, N. Steckling, and B. Lettmeier. 2010. Mercury exposure and children's health. Current Problems in Pediatric and Adolescent Health Care 40(8):186–215.

Canty, A., and B. Ripley. 2017. boot. R, Oxford, UK. https://cran.r-project.org/web/packages/boot/boot.pdf Accessed 04/14/2020.

This article is protected by copyright. All rights reserved

Carpenter, K. E. 2018. *Squalus acanthias*, Picked Dogfish. https://www.fishbase.se/summary/Squalus-acanthias.html. Accessed 04/14/2020.

Chen, C. Y., M. E. Borsuk, D. M. Bugge, T. Hollweg, P. H. Balcom, D. M. Ward, J. Williams, and R. P. Mason. 2014. Benthic and pelagic pathways of methylmercury bioaccumulation in estuarine food webs of the Northeast United States. PLoS ONE 9(2):e89305.

Cladis, D. P., A. C. Kleiner, and C. R. Santerre. 2014. Mercury content in commercially available finfish in the United States. Journal of Food Protection 77(8):1361–1366.

Cross, F. A., D. W. Evans, and R. T. Barber. 2015. Decadal declines of mercury in adult Bluefish (1972–2011) from the Mid-Atlantic Coast of the U.S.A. Environmental Science & Technology 49(15):9064–9072.

Dell'Apa, A., J. C. Johnson, D. G. Kimmel, and R. A. Rulifson. 2013. The international trade and fishery management of Spiny Dogfish: A social network approach. Ocean & Coastal Management 80:65–72.

Didden, J. 2018. Spiny Dogfish advisory panel informational document -- August 2018. Mid-Atlantic Fishery Management Council.

Domi, N., J. M. Bouquegneau, and K. Das. 2005. Feeding ecology of five commercial shark species of the Celtic Sea through stable isotope and trace metal analysis. Marine Environmental Research 60(5):551–569.

Driscoll, C. T., R. P. Mason, H. M. Chan, D. J. Jacob, and N. Pirrone. 2013. Mercury as a global pollutant: sources, pathways, and effects. Environmental Science & Technology 47(10):4967–4983.

Ebert, D. A., W. T. White, K. J. Goldman, L. J. V. Compagno, T. S. Daly–Engel, and R. D. Ward. 2010. Resurrection and redescription of *Squalus suckleyi* (Girard, 1854) from the North Pacific, with comments on the *Squalus acanthias* subgroup (Squaliformes: Squalidae). Zootaxa 2612(1):22.

Endo, T., Y. Hisamichi, O. Kimura, Y. Kotaki, Y. Kato, C. Ohta, N. Koga, and K. Haraguchi. 2009. Contamination levels of mercury in the muscle of female and male Spiny Dogfishes (*Squalus acanthias*) caught off the coast of Japan. Chemosphere 77(10):1333–1337.

EPA. 2001. Methylmercury (MeHg). United States Environmental Protection Agency, Assessment CASRN 22967-92-6, Washington, D.C.

Author Manuscript DRAFT

This article is protected by copyright. All rights reserved

EPA, and FDA. 2019. Screening values for fish categories. United States Environmental Protection Agency.

EUMOFA. 2019. EU imports of Dogfish, 2000-2018. European Market Observatory for Fisheries and Aquaculture Products, Yearly Ad-Hoc Query 1547220131296, Brussels, Belgium.

Evers, D. C., J. DiGangi, J. Petrlik, D. G. Buck, J. Šamánek, B. Beeler, M. A. Turnquist, S. K. Hatch, and K. Regan. 2014. Global mercury hotspots: new evidence reveals mercury contamination regularly exceeds health advisory levels in humans and fish worldwide. Page 20. Biodiversity Research Institute, 2014–34, Portland, ME.

Fabris, G. J., C. Monahan, G. Nicholson, and T. I. Walker. 1992. Total mercury concentrations in Sand Flathead, *Platycephalus bassensis* Cuvier & Valenciennes from Port Phillip Bay, Victoria. Australian Journal of Marine and Freshwater Research 43:1393–1402.

FDA. 2012. Mercury concentrations in fish: FDA monitoring program (1990-2010). United States Food and Drug Administration.

FDA. 2017. Mercury levels in commercial fish and shellfish (1990-2012). U.S. Food and Drug Administration.

Fogarty, M. J., and S. A. Murawski. 1998. Large-scale disturbance and the structure of marine systems: fishery impacts on Georges Bank. Ecological Applications 8(sp1):S6–S22.

Frisk, M. G., T. J. Miller, S. J. D. Martell, and K. Sosebee. 2008. New hypothesis helps explain elasmobranch "outburst" on Georges Bank in the 1980s. Ecological Applications 18(1):234–245.

Grandjean, P., P. Weihe, R. F. White, F. Debes, S. Araki, K. Yokoyama, K. Murata, N. Sørensen, R. Dahl, and P. J. Jørgensen. 1997. Cognitive deficit in 7-year-old children with prenatal exposure to methylmercury. Neurotoxicology and Teratology 19(6):417–428.

Greig, R. A., D. Wenzloff, C. Shelpuk, and A. Adams. 1977. Mercury concentrations in three species of fish from North Atlantic offshore waters. Archives of Environmental Contamination and Toxicology 5(1):315–323.

Grolemund, G. 2014. lubridate. https://cran.r-project.org/web/packages/lubridate/lubridate.pdf. Accessed 04/14/2020.

This article is protected by copyright. All rights reserved

Hall-Arber, M., C. Dyer, J. Poggie, J. McNally, and R. Gagne. 2001. New England's fishing communities. Page 431. Massachusetts Sea Grant, Technical Report MITSG 01-15, Cambridge, MA.

Harry, A. V. 2018. Evidence for systemic age underestimation in shark and ray aging studies. Fish and Fisheries 19(2):185–200.

Hightower, J. M., and D. Moore. 2003. Mercury levels in high-end consumers of fish. Environmental Health Perspectives 111(4):604–608.

Hobbs, C. A. D., R. W. A. Potts, M. Bjerregaard Walsh, J. Usher, and A. M. Griffiths. 2019. Using DNA barcoding to investigate patterns of species utilisation in UK shark products reveals threatened species on sale. Scientific Reports 9(1).

ICES. 2018. EU request for ICES to provide advice on a revision of the contribution of TACs to fisheries management and stock conservation. ICES.

Karimi, R., T. P. Fitzgerald, and N. S. Fisher. 2012. A quantitative synthesis of mercury in commercial seafood and implications for exposure in the United States. Environmental Health Perspectives 120(11):1512–1519.

Kauermann, G., and H. Küchenhoff. 2011. Stichproben. Springer Berlin Heidelberg, Berlin, Heidelberg.

Kim, S.-J., H.-K. Lee, A. C. Badejo, W.-C. Lee, and H.-B. Moon. 2016. Species-specific accumulation of methyl and total mercury in sharks from offshore and coastal waters of Korea. Marine Pollution Bulletin 102(1):210–215.

Kousteni, V., P. Megalofonou, M. Dassenakis, and E. Stathopoulou. 2006. Two mercury concentrations in edible tissues of two elasmobranch species from Crete (eastern Mediterranean Sea). Cybium 30(4):119–123.

Lamborg, C. H., C. R. Hammerschmidt, K. L. Bowman, G. J. Swarr, K. M. Munson, D. C. Ohnemus, P. J. Lam, L.-E. Heimbürger, M. J. A. Rijkenberg, and M. A. Saito. 2014. A global ocean inventory of anthropogenic mercury based on water column measurements. Nature 512(7512):65–68.

Manitz, J., M. Hempelmann, G. Kauermann, H. Kuechenhoof, S. Shao, C. Oberhauser, N. Westerheide, and M. Wiesenfarth. 2017. samplingbook. R. https://cran.r-project.org/web/packages/samplingbook/samplingbook.pdf. Accessed 04/14/2020.

Author Manuscript DRAFT

This article is protected by copyright. All rights reserved

Marschner, I., and M. Donoghoe. 2018. glm2. Sydney, Australia. https://cran.r-project.org/web/packages/glm2/glm2.pdf. Accessed 04/14/2020.

Mason, R. P., Z. Baumann, G. Hansen, K. M. Yao, M. Coulibaly, and S. Coulibaly. 2019. An assessment of the impact of artisanal and commercial gold mining on mercury and methylmercury levels in the environment and fish in Cote d'Ivoire. Science of the Total Environment 665:1158–1167.

Mason, R. P., A. L. Choi, W. F. Fitzgerald, C. R. Hammerschmidt, C. H. Lamborg, A. L. Soerensen, and E. M. Sunderland. 2012. Mercury biogeochemical cycling in the ocean and policy implications. Environmental Research 119:101–117.

Masury, K., and S. Schumann. 2019. Eat like a fish: diversifying New England's seafood marketplace. Eating with the Ecosystem.

Maurer, J. 2017, November 15. Cucalorus Connect panel discusses accelerating state's seafood economy. Greater Wilmington Business Journal. Wilmington, NC.

Natanson, L. J., A. H. Andrews, M. S. Passerotti, and S. P. Wintner. 2018a. History and mystery of age and growth studies in elasmobranchs. Page Shark Research: Emerging Technologies and Applications for the Field and Laboratory. CRC Press, Boca Raton, Florida, USA.

Natanson, L. J., G. B. Skomal, S. L. Hoffmann, M. E. Porter, K. J. Goldman, and D. Serra. 2018b. Age and growth of sharks: do vertebral band pairs record age? Marine and Freshwater Research 69(9):1440.

Nye, J., J. Link, J. Hare, and W. Overholtz. 2009. Changing spatial distribution of fish stocks in relation to climate and population size on the Northeast United States continental shelf. Marine Ecology Progress Series 393:111–129.

Orlov, A. M., E. F. Kulish, I. N. Mukhametov, and O. A. Shubin. 2011. Age and growth of spiny dogfish *Squalus acanthias* (Squalidae, Chondrichthyes) in pacific waters off the Kuril Islands. Journal of Ichthyology 51(1):42–55.

Pethybridge, H., E. Butler, D. Cossa, R. Daley, and A. Boudou. 2012. Trophic structure and biomagnification of mercury in an assemblage of deepwater chondrichthyans from southeastern Australia. Marine Ecology Progress Series 451:163–174.

Pope, K. L., and C. G. Kruse. 2007. Condition. Pages 423–472 Analysis and interpretation of freshwater fisheries data. American Fisheries Society, Bethesda, Maryland.

Author Manuscript DRAFT

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R Core Team. 2017. R: A language and environment for statistical computing. The R Foundation for Statistical Computing, Vienna, Austria.

Reum, J. C. P. 2011. Lipid correction model of carbon stable isotopes for a cosmopolitan predator, Spiny Dogfish *Squalus acanthias*. Journal of Fish Biology 79(7):2060–2066.

Schartup, A. T., C. P. Thackray, A. Qureshi, C. Dassuncao, K. Gillespie, A. Hanke, and E. M. Sunderland. 2019. Climate change and overfishing increase neurotoxicant in marine predators. Nature 572(7771):648–650.

Selin, N. E., D. J. Jacob, R. M. Yantosca, S. Strode, L. Jaeglé, and E. M. Sunderland. 2008. Global 3-D land-ocean-atmosphere model for mercury: present-day versus preindustrial cycles and anthropogenic enrichment factors for deposition: global 3-D land-ocean-atmosphere model for mercury. Global Biogeochemical Cycles 22(2)

Smith, B. E., and J. S. Link. 2010. The trophic dynamics of 50 finfish and 2 squid species on the Northeast US continental shelf. Page 646. Northeast Fisheries Science Center, Technical Memorandum NMFS-NE-216, Woods Hole, MA.

Spiess, A.-N. 2018. qpcR. R, Hamburg, Germany. https://cran.r-project.org/web/packages/qpcR/qpcR.pdf 04/14/2020

St. Gelais, A. T., and B. A. Costa-Pierce. 2016. Mercury concentrations in Northwest Atlantic winter-caught, male Spiny Dogfish (*Squalus acanthias*): A geographic mercury comparison and risk-reward framework for human consumption. Marine Pollution Bulletin 102(1):199–205.

von Stackelberg, K., M. Li, and E. Sunderland. 2017. Results of a national survey of high-frequency fish consumers in the United States. Environmental Research 158:126–136.

Stehlik, L. L. 2007. Spiny dogfish, *Squalus acanthias*, life history and habitat characteristics. National Marine Fisheries Service, NOAA Technical Memorandum NMFS-NE-203, Highlands, NJ.

Stenhouse, I. J., E. M. Adams, J. L. Goyette, K. J. Regan, M. W. Goodale, and D. C. Evers. 2018. Changes in mercury exposure of marine birds breeding in the Gulf of Maine, 2008–2013. Marine Pollution Bulletin 128:156–161.

Storelli, M. M., S. R. Giacominelli, and G. O. Marcotrigiano. 2001. Total mercury and methylmercury in tuna fish and sharks from the South Adriatic Sea. Italian Journal of Food Science 1(13):101–106.

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DRAFT

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Streets, D. G., H. M. Horowitz, Z. Lu, L. Levin, C. P. Thackray, and E. M. Sunderland. 2019. Global and regional trends in mercury emissions and concentrations, 2010–2015. Atmospheric Environment 201:417–427.

Sunderland, E. M. 2007. Mercury exposure from domestic and imported estuarine and marine fish in the U.S. seafood market. Environmental Health Perspectives 115(2):235–242.

Sunderland, E. M., M. Li, and K. Bullard. 2018. Decadal changes in the edible supply of seafood and methylmercury exposure in the United States. Environmental Health Perspectives 126(1):017006.

Taylor, D. L., N. J. Kutil, A. J. Malek, and J. S. Collie. 2014. Mercury bioaccumulation in cartilaginous fishes from Southern New England coastal waters: contamination from a trophic ecology and human health perspective. Marine Environmental Research 99:20–33.

Taylor, D. L., and P. R. Williamson. 2017. Mercury contamination in Southern New England coastal fisheries and dietary habits of recreational anglers and their families: implications to human health and issuance of consumption advisories. Marine Pollution Bulletin 114(1):144–156.

Teffer, A. K., M. D. Staudinger, D. L. Taylor, and F. Juanes. 2014. Trophic influences on mercury accumulation in top pelagic predators from offshore New England waters of the northwest Atlantic Ocean. Marine Environmental Research 101:124–134.

U.S. EPA. 2017. EPA-FDA Fish Advice: Technical Information. U.S. Environmental Proection Agency. https://www.epa.gov/fish-tech/epa-fda-fish-advice-technical-information. Accessed 04/14/2020.

USOFR. 2019. Reference amounts customarily consumed per eating occasion. Code of Federal Regulations Title 21(Part 101):Subpart A Section 12.

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TABLES

Table 1– AICc scores and model weights for glms of $\ln(\mathrm{THg}_{wet})$ predicted by total length (L), mass (W), condition factor (K), capture date (D), sex (S), and whether or not the females were carrying live young (P). β values are only shown if they were determined to be significant (non-zero). A (+) sign next to the β value indicates that the effect was positive, and a (-) sign indicates the effect was negative.

Model	β	**ΔAICc**	**Weight**
$L+W+D+S+L*S$	β_{W}(+), p=0.025	0	0.582
$L+W+D+S$	β_{males}(+), p=0.013	0.92756	0.366
W		5.14161	0.045
$W+P+W*P$		8.84868	0.007
$K+D$		16.4583	0
L		16.88886	0
$L+S+L*S$		17.12158	0
K		18.06937	0
$L+P+L*P$		20.63608	0
D		25.5084	0

Table 2 – Comparison of mean THg concentration (µg/g, wet tissue) in muscle tissue of Spiny Dogfish caught off the coast of Southern New England in the early 1970s (Greig et al. 1977) and 2018 (this study) using size categories defined by Greig et al. (1977).

Study	Size (mm)	n	$\bar{x}$ THg (µg/g)	s THg (µg/g)	t	df	p
1977	< 755	16	0.350	0.160	2.631	22.787	0.007
2018		13	0.230	0.079			
1977	755 – 854	17	0.420	0.165	0.819	33.829	0.209
2018		33	0.379	0.173			
1977	> 855	25	0.600	0.300	2.957	29.444	0.003
2018		55	0.413	0.148			

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Table 3 – Chance of exceeding FDA/EPA Hg exposure limits based on body weight (lbs) and portion sizes (taken from FDA recommendations). In each box, the integer outside the parentheses indicates the maximum number of meals of Spiny Dogfish that can be consumed per month before having a > 0.05 chance of exceeding the FDA/EPA recommended Hg exposure level for the month. The number inside the parentheses indicates the chance of exceeding the recommended Hg exposure level for the month. All values assume no other sources of mercury are consumed within a given month.

Weight	≤ 5 years	6 – 8 years	9 – 11 years	≥ 12 years
(lbs)	1 oz (28.3 g)	2 oz (56.6 g)	3 oz (84.9 g)	4 oz (113.2 g)
25	2 (0.037)			
50	4 (0.005)	2 (0.037)		
75		3 (0.014)	2 (0.037)	
100		4 (0.006)	2 (0.001)	2 (0.037)
125			3 (0.003)	2 (0.003)
150				3 (0.014)
175				4 (0.037)
200				4 (0.006)
225				5 (0.016)
250				6 (0.033)
275				6 (0.007)
300				7 (0.014)

FIGURES

Figure 1 – Capture locations of Spiny Dogfish sampled in this study (white circles). All fish were landed at the Chatham Fish Pier on the southeast corner of Cape Cod, MA (gray star).

Figure 2 – Length (mm) frequency histograms for female (F, dark gray) and male (M, hatched) Spiny Dogfish sampled from the Chatham Fish Pier in summer 2018.

Figure 3 – Methylmercury (MeHg) and total mercury (THg) measurements for the subset of Spiny Dogfish (n = 16) that were analyzed for both. The dashed line is the 1:1 line. The dotted line is the modeled relationship between MeHg and THg ($p = 0.002$, $R^2 = 0.522$). The gray bars

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indicate confidence bounds associated with measurement error that were calculated from samples run in triplicate.

Figure 4 -- Total mercury (THg) concentrations (μg/g wet weight) of spiny dogfish plotted as a function of pre-caudal length (mm) along with lines of best fit for our study (dark circles, solid line) and Taylor et al. 2014 (open circles, dashed line). The FDA and EPA thresholds (1.0 μg/g and 0.3 μg/g respectively) are marked with light gray dashed lines.

Figure 5 -- Simulated monthly Hg intake (μg; left y-axis) based on servings of Spiny Dogfish consumed (x-axis) for children ages 5 and under eating 28.3 g (1 oz) servings (a), ages 6 – 8 eating 56.6 g (2 oz) servings (b), 9 – 11 eating 84.9 g (3 oz) servings (c), and adults ages > 11 eating 113.2 g (4 oz) servings (d). Dashed lines represent maximum recommended exposure values for different body weights (kg; right y-axis).

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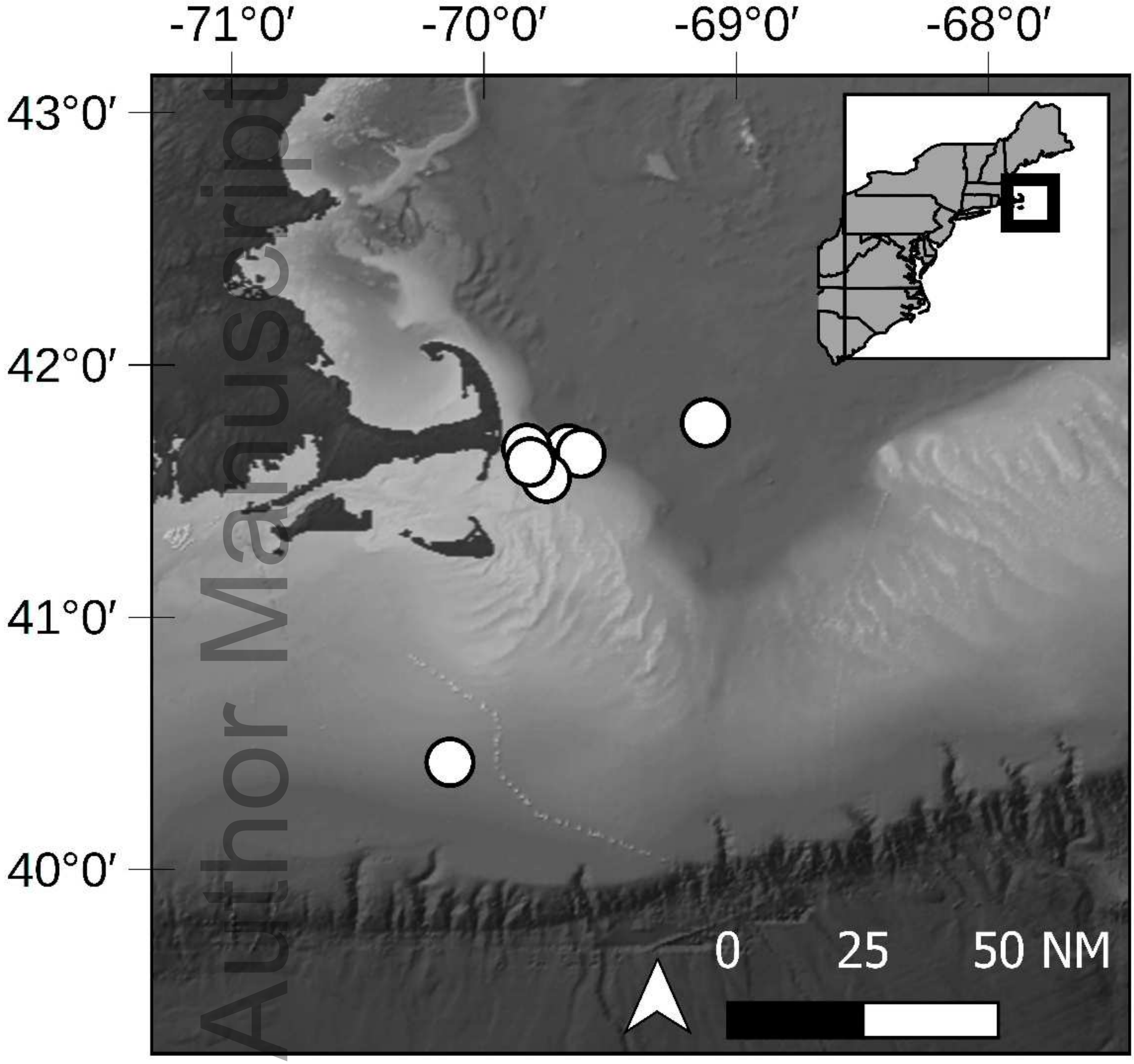

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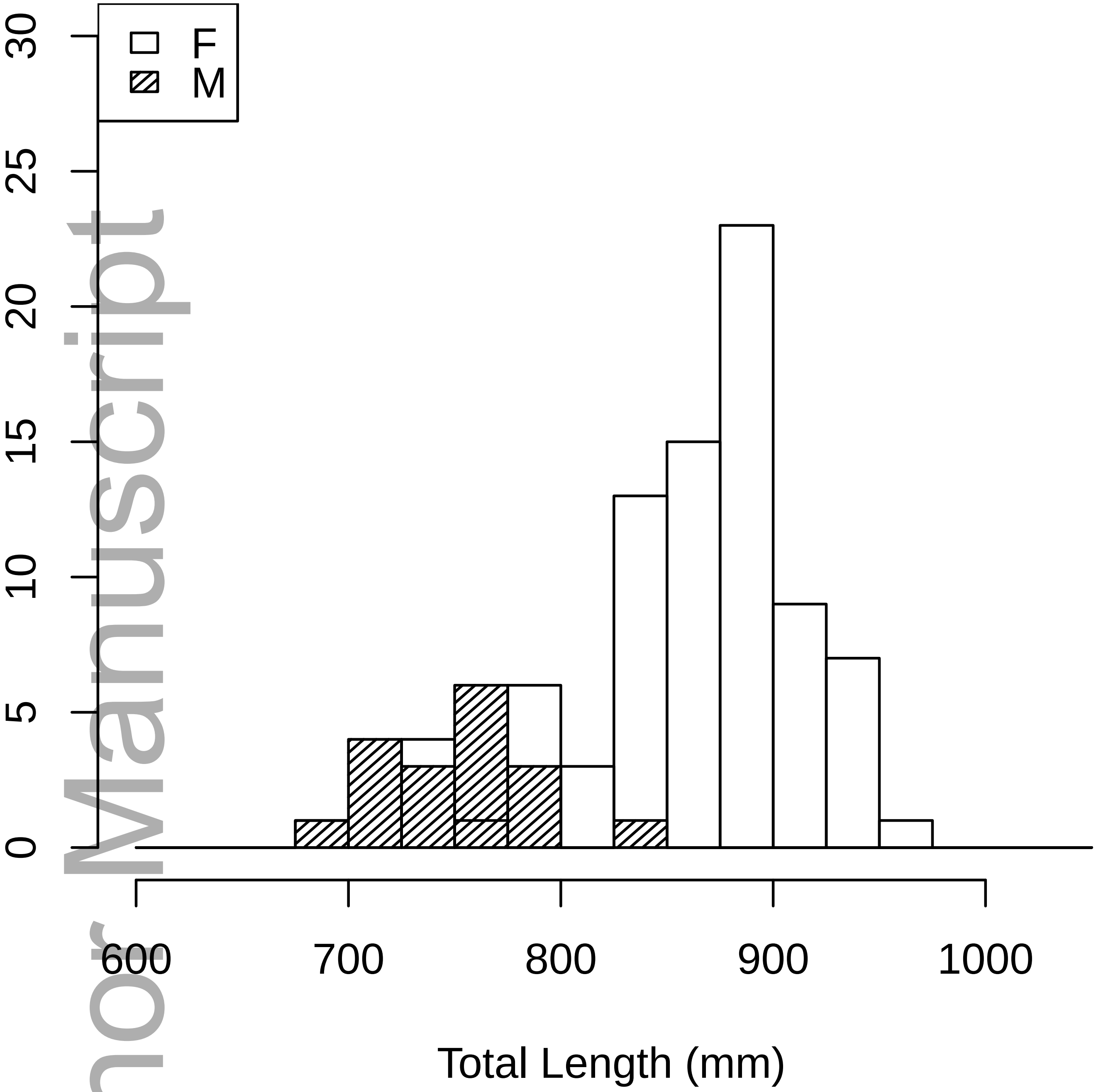

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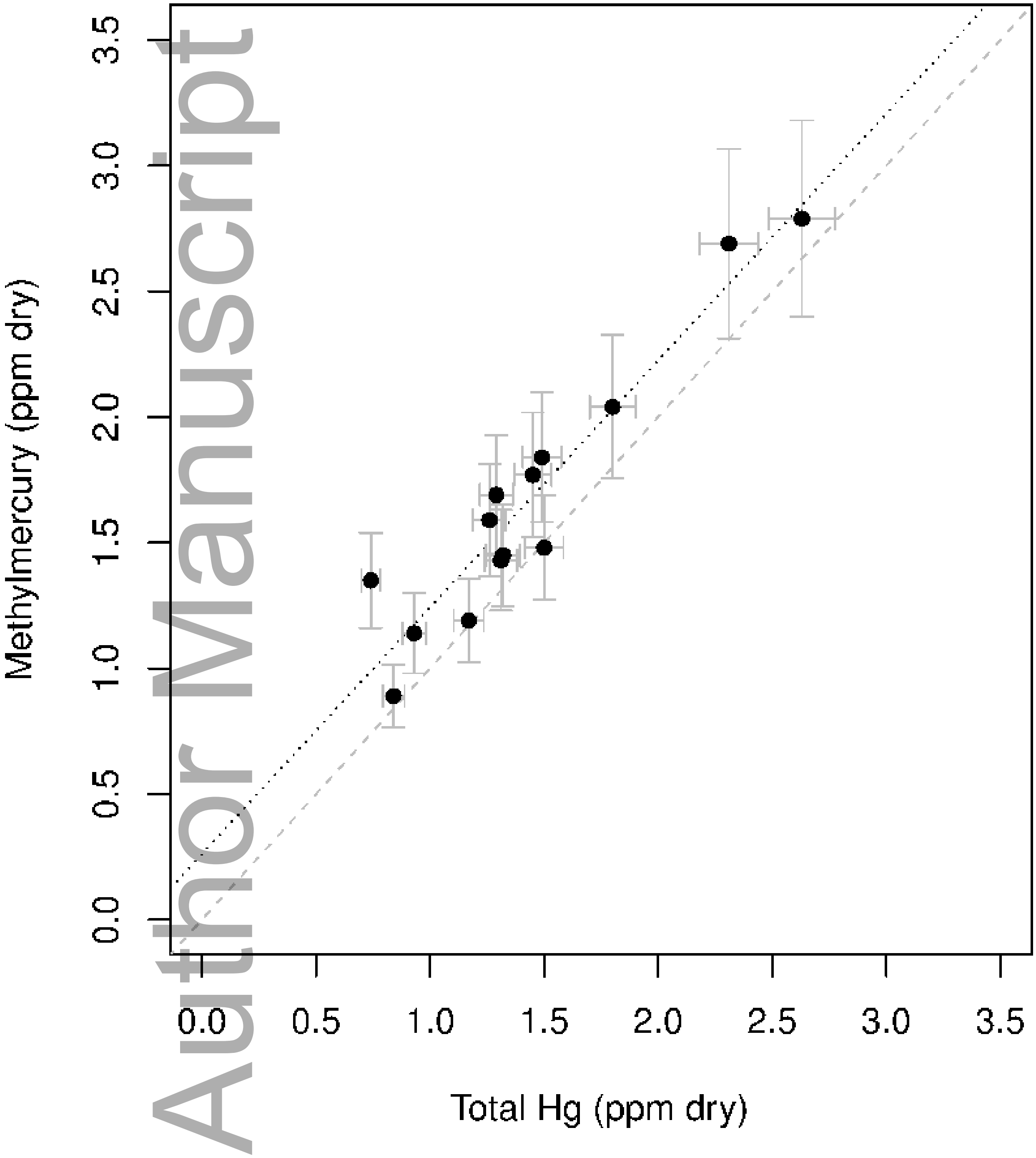

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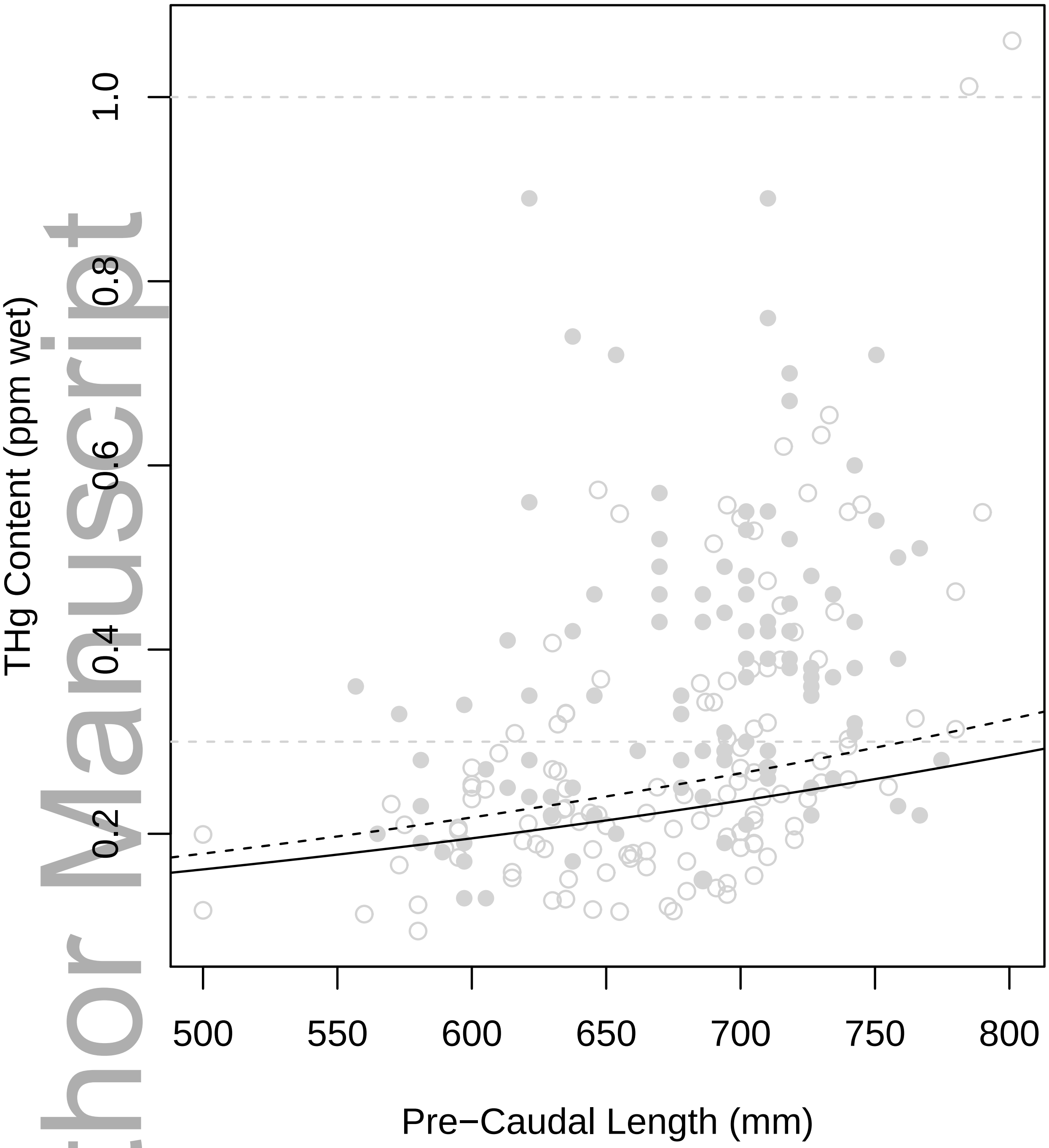

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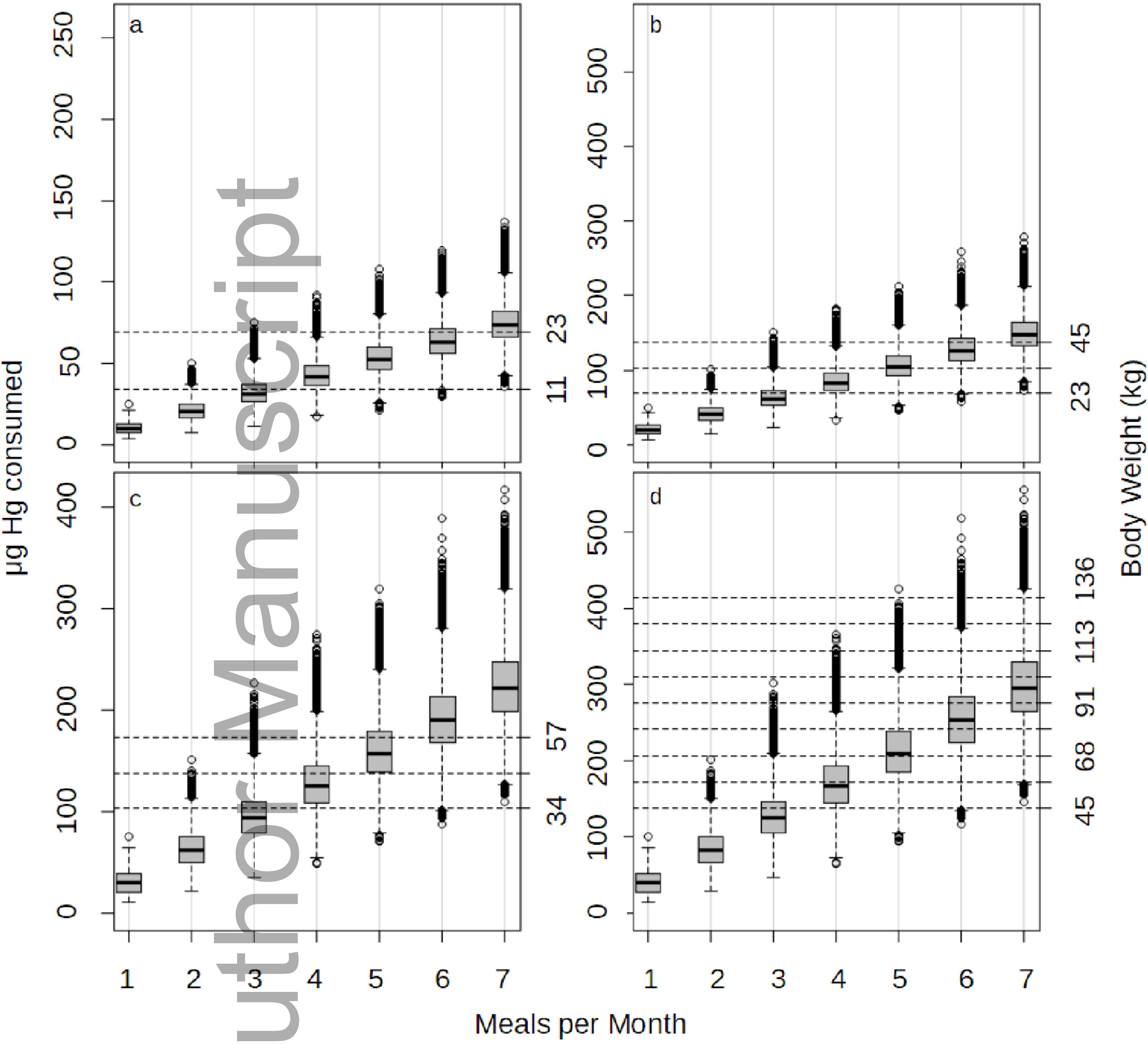

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