MERCURY BIOACCUMULATION IN TILEFISH (*Lopholatilus chamaeleonticeps*) FROM OFFSHORE WATERS OF SOUTH CAROLINA, USA.

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ABSTRACT

Mercury (Hg) concentrations in Tilefish (*Lopholatilus chamaeleonticeps*) have been reported to be one of the highest of all fish species, resulting in advisories that, historically, have recommended zero consumption. The current study assesses Hg bioaccumulation in Tilefish targeted by the commercial fisheries operating off the coast of South Carolina, USA. We provide results for an under-sampled region and explore how life history potentially impacts Hg uptake in Tilefish. Mercury concentration in Tilefish muscle tissue ranged from 0.10 to 0.99 ppm, with a mean of 0.23 ppm (n=63). The majority of Tilefish samples (95%) were within the "Good Choices" range for consuming at least one serving per week, with 62% being within the range considered best for eating two meals a week", per suggestion by the US EPA and US FDA (2017). The present study of Tilefish from the western Atlantic further substantiates the importance of monitoring Hg in commercial fish species regionally.

Keywords: Toxicology, Environmental Health, Ecology

Capsule: Tilefish from offshore South Carolina have considerably lower concentrations of Hg, substantiating the importance of monitoring Hg in commercial fish species regionally.

INTRODUCTION

Tilefish (*Lopholatilus chamaeleonticeps*), or 'Golden Tilefish', is a long-lived, demersal species found from Nova Scotia to Suriname and throughout the Gulf of Mexico and continental Caribbean (Carpenter 2002). They are deep-dwelling (81 to 540 m), but generally found in a relatively narrow zone along the continental slope; its habitat is generally restricted to mud bottom (Dooley 1978).

Tilefish support valuable commercial fisheries along the Atlantic coast of the USA including the Gulf of Mexico. While the annual landings of Tilefish have overall declined since their peak in 1979 of 4,073 metric tons, the annual landings of Tilefish since 2001 have remained steady at 1100+ metric tons (NMFS Fish Statistics and Economic Division, pers. comm.; Fig. 1). The importance of this deep-water species as a protein source to fish-consuming citizens stresses the need for a complete understanding of possible health concerns that could be linked to its consumption, throughout its geographic range.

Eating fish is an important source of low-fat protein including Omega-3 fatty acids that have been found to benefit human cardiovascular health (Albert *et al.* 2002; Patterson 2002). Unfortunately, fish consumption is also the primary source of mercury (Hg) for humans (WHO 1976; IOM 2007; Chen *et al.* 2012). With the potential to bioaccumulate to high levels in some organisms and penetrate blood-brain barriers, Hg can become a health concern for pregnant women and children when seafood is consumed frequently (Yang *et al.* 1997; Burger & Gochfeld 2006; Bank *et al.* 2007). Mercury occurs in the environment naturally; however, anthropogenic emissions of mercury have been larger than natural emissions since the start of the industrial age about 200 years ago (UNEP 2013).

High concentrations of Hg were reported for Tilefish from the Gulf of Mexico (Hall *et al.* 1978), and Tilefish were listed by the United States Food & Drug Administration (FDA) as having levels of Hg too high for consumption by pregnant women and children (US EPA & US FDA 2004). While public awareness of Hg contamination in seafood is very important, evidence of regional differences in Hg levels exists within a species (Adams and McMichael 2007, Harris *et al*. 2012; Sinkus *et al.* 2017). Therefore, improved understanding of the geographic patterns of Hg levels is necessary for the angler and/or consumer. Driven by updated Tilefish mercury data from the Atlantic, the EPA and the FDA altered their advice for consumption in 2014, listing Tilefish from the Atlantic and Gulf of Mexico separately. The aim of the current study is to summarize Hg concentration in muscle tissue of Tilefish off the coast of South Carolina, United States.

METHODS

Tissue samples of Tilefish were collected from fish caught off the coast of South Carolina (Figure 2) by commercial fishermen and the South Carolina Department of Natural Resources from 2009 to 2010. Gear used for sampling was bottom longlines and all specimens were caught off the continental shelf slope in depths greater than 200 m.

For each fish, the sagittal otoliths were removed, standard length (SL), fork length (FL), total length (TL), were measured to the nearest mm and whole weight (when available) recorded to the nearest g. A 300–400 g scaled, skin-on piece of axial muscle tissue sample was excised from each fish and stored as outlined by Sinkus *et al*. (2017). Fish age was determined by otoliths increment analysis, utilizing standard methods outlined in Sinkus *et al*. (2017). To determine fish sex standard procedures for histological sample collection, processing, and interpretation were used, as described by White and Palmer (2004).

Muscle tissue was analyzed for Hg concentration utilizing practices outlined in Sinkus *et al*. (2017). Briefly, carefully prepared 0.2–0.3 g subsamples in tared sterile nickel boats were analyzed for total mercury (THg) in a direct Hg analyzer, DMA–80 (Milestone Inc., Monroe, CT). Standard reference materials (SRMs), DORM-2 dogfish liver tissue and TORT-2 lobster hepatopancreas (National Research Council, Canada) were used to create a matrix–matched calibration curve that was then used to determine the amount of THg in the sample. Method blanks, duplicates and 2 different SRMs, Dolt–4 dogfish liver tissue and 1566b oyster tissue (National Research Council, Canada and NIST, Gaithersberg, MD), were analyzed consistently to ensure quality control. Calibration curves for the sample runs had r^2 values exceeding 0.99. Recovery of the SRMs, mean method detection limits based on three times the standard deviation of method blanks (0.39 ng of Hg, less than 10% of the lowest sample), and differences between duplicate measurements of the same samples was within standard acceptable limits.

Because Hg accumulates in tissues of fishes mainly as methylmercury (MeHg), making up more than 90% of the THg in fish muscle of some carnivorous and omnivorous species (Bloom 1992, Bank *et al.* 2007), THg was measured as a proxy for MeHg. For simplicity, the term Hg from this point on represents THg wet weight concentration in muscle tissue reported in ppm (mg Hg kg^{-1} wet wt.).

Prior to 2017, the EPA and FDA fish consumption advice concerning mercury was centered around a screening level of 0.3 ppm. A fish species that had a mean Hg concentration below that screening level was deemed as a healthy option for consuming two meals of 12 ounces per week. If found above that level, advice was given to only consume one meal of 6 ounces per week, except for four types of fishes, King mackerel (*Scomberomorus cavalla*), Shark, Swordfish (*Xiphias gladius*), and 'Golden' Tilefish, due to their higher Hg concentrations (US EPA & US FDA 2004). An update to the consumption guides in 2017, set new categories, with three tiers, fishes that were "Best Choices", "Good Choices", and "Choices to Avoid", each having new screening values. "Best Choice" fishes have Hg concentrations below 0.15 ppm, of which it is recommended to eat three servings per week. "Good Choice" fishes have two tiers; it is recommended to have two servings per week of fishes that have Hg concentrations between 0.16 ppm and 0.23 ppm and to have one serving per week of fishes that have Hg concentrations between 0.24 ppm and 0.46 ppm. It is recommended to avoid eating fish that have Hg concentrations greater than 0.46 ppm (US EPA & US FDA 2017).

A series of bivariate linear regressions and Spearman's correlation analyses were used to assess the relationships between Hg concentration and fish age, length, and weight to determine if significant correlations existed. Mercury data were ln-transformed to address issues with heteroscedasticity prior to evaluating the relationship between Hg concentration and fish characteristics using simple linear regression analysis. Comparisons of Hg between sexes were carried out using ANOVA.

Statistical analyses were performed using R statistical program (R Development Core team 2016) and the α value was set at 0.05 for all statistical tests. Error is represented as standard error unless otherwise stated.

RESULTS

Samples of muscle tissue from 63 specimens of Tilefish were collected during 2009 - 2010 and analyzed for Hg concentrations. Fish TL ranged from 499 mm to 967 mm with a mean of 691 ± 12.8 mm, fish whole weight ranged from 1290 to 10,900 g with a mean of 4144 ± 256.9 g, and fish age ranged from 6 to 19 years with a mean of 9.8 ± 0.34 years. Mercury concentrations ranged from 0.10 to 0.99 ppm with a 0.23 ± 0.017 ppm. Of the 63 samples analyzed, 95% were within the "Good Choices" range for consuming at least one serving per week, with 62% being within the range considered best for eating two meals a week. Regression and correlation analyses revealed no significant relationships between Hg level and total length ($F(1,61) = 0.08$, $P = 0.78$, *adj* $R^2 = -0.015$; Spearman's correlation: $\rho = 0.05$, $P = 0.68$; Figure 3), Hg level and whole weight ($F(1,61) = 0.18$, $P = 0.68$, *adj* $R^2 = -0.013$; Spearman's correlation: $\rho = 0.08$, $P = 0.54$; Figure 4), and Hg level and age ($F(1,58) = 2.37$, $P = 0.13$, *adj* $R^2 = 0.023$; Spearman's correlation: $\rho = -0.20$, P = 0.12; Figure 5). There was no significant difference in mean Hg concentration between male and female Tilefish (ANOVA: $F_{1, 61} = 0.003$, $P < 0.96$; Figure 6).

Significant differences in mean length were observed between males and females (ANOVA: $F_{1, 61} = 20.2$, $P < 0.001$), and weights (ANOVA: $F_{1, 61} = 15.4$, $P<0.001$) with males being larger at similar ages. However, no significant difference in mean age was found between males and females (ANOVA: $F_{1,58} = 0.01$, $P < 0.94$).

DISCUSSION

The present study provides the most recent information on Hg concentrations of Tilefish in western Atlantic waters and is the first to assess Tilefish specimens off the coast of South Carolina, USA. The range of Hg concentrations from the present study (0.10-0.99 ppm) were comparable to values of other similar studies. Most of the samples (95%) were within the range of “Good Choices” fish set by the EPA and FDA. The mean Hg concentration for Tilefish in the present study (0.23 ppm) was 16% of mean Hg concentration (1.45 ppm) in the study conducted by NOAA-NMFS and used by the FDA to set consumption guidelines (Hall *et al.* 1978). Unlike results from previous studies on other species (Bank *et al.* 2007; Sinkus *et al.* 2017), the present study suggested there was no correlation between Hg levels and Tilefish length, weight, or age. Additionally, while depth has been indicated as an influential factor on Hg accumulation in other studies (Choy *et al*. 2009), depth had limited capability to predict Hg concentrations in Tilefish for the current study. One possible reason for this is the limited range of depth from which these samples were selected (210 m – 240 m).

Previous work, an independent study commissioned by the Montauk Tilefish Association in 2002, reported a similarly low range of Hg (0.03-0.27 ppm) in Tilefish captured in New England with mean of 0.09 ppm (*personal communication* Jose Montanez, Mid-Atlantic Fishery Management Council). The FDA also conducted its own study (2002-2003) of Hg levels in various fish species, including Tilefish caught off the northeastern United States, and found a range in concentration of Hg in Tilefish of 0.06-1.12 ppm, with a mean of 0.21 ppm (US FDA, 2014), which is similar to the results of the present study. These studies show that Hg levels in Tilefish vary depending on where and when they were captured, and that Tilefish along the Western Atlantic captured in the 2000s have considerably lower Hg concentration than what has been publicized for Gulf of Mexico fish captured in the 1970's. As a result of the Montauk Tilefish Association study and the FDA's most recent study of Hg in Tilefish, the FDA now reports Hg levels for Gulf of Mexico and the Atlantic regions separately.

Of the fish that the EPA and FDA have listed as having the highest Hg levels [King mackerel (*Scomberomorus cavalla*), Shark, Swordfish (*Xiphias gladius*), and 'Golden' Tilefish], and issued fish consumption advisories for women who are pregnant or of childbearing age, Tilefish is the only one that is not a pelagic species in an upper trophic level (US EPA & US FDA, 2014). Nothing in the literature specifically examines why Tilefish, with a life history completely different from pelagic King mackerel, Shark, or Swordfish would have Hg levels of concern. Tilefish live in deep water (81 to 540 m) habitat over mud bottoms, where they are known to dig and shelter in burrows (Able *et al.* 1982; Grossman *et al.* 1985). However, it is reported that there is a general trend of increasing Hg concentrations in pelagic fishes with increasing water depth (Choy *et al.* 2009). In coastal environments, inorganic Hg is transformed into MeHg primarily in sediments, as well as in the open ocean, with this conversion taking place largely at depths between 200 and 1000 meters in the water column (UNEP 2013). Therefore, Tilefish may access Hg simply due to the depth of their habitat, and possibly from the sediment found at those depths. Mercury transformation at depth could potentially be why there is no correlation to Hg levels and size, age, or sex. Another species from the Malacanthidae family, Blueline Tilefish (*Caulolatilus microps*) was found to exhibit weak correlations between Hg concentrations and fish size, age, or sex (Sinkus *et al.* 2017). Further research should assess Hg levels in species of *Malacanthus*, their prey, and the sediments of their associated burrows in order to better understand the process of Hg uptake in this family of fishes.

Government advisories and associated media coverage have informed the public of the benefits and risks of consuming fish. However, there is a dearth of information regarding the concentrations of Hg in fish from different geographic regions. Assessing mercury levels in commercially harvested fish requires samples from the geographical range of a species. Only then will the consumer have the necessary information to make decisions on which species of fish to eat (Sinkus *et al.* 2017) or what geographic source of fish to avoid (Burger & Gochfeld 2006).

CONCLUSIONS

While the authors acknowledge that there are some risks in consuming Tilefish, the public should know that Tilefish from Atlantic waters do not represent the higher risk that the FDA has historically presented. The differences in Hg levels from different geographic areas justify that the FDA, as well as individual states, should regularly monitor Hg levels of commercial fish, changes in market sources, species composition and size, as well as consumer patterns (Karimi *et al.*, 2012) to inform the public about the geographic distribution of Hg concentration in the fish they eat.

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FIGURES

Figure 1. Commercial landings of Tilefish from the Atlantic and Gulf of Mexico.

Figure 2. Map of study area in the Atlantic Ocean off South Carolina for Tilefish. Grey dots represent sampling locations and the size of the dots correlates to the number of fished sampled from that location. The black line represents the Continental Shelf Break.

Figure 3. Observed (filled circles) and predicted (solid line) Hg concentrations as a function of total length for Tilefish. Short dashed curves = 95% confidence intervals. Dark grey shaded area = "Choices to Avoid", grey shaded area = "Good Choices" for one meal a week, light grey shaded area = "Good Choices" for two meals a week, unshaded area = "Best Choices", per suggestion by the US EPA and US FDA (2017).

Figure 4. Observed (filled circles) and predicted (solid line) Hg concentrations as a function of whole weight for Tilefish. Short dashed curves = 95% confidence intervals; Dark grey shaded area = "Choices to Avoid", grey shaded area = "Good Choices" for one meal a week, light grey shaded area = "Good Choices" for two meals a week, unshaded area = "Best Choices", per suggestion by the US EPA and US FDA (2017).

Figure 5. Observed (filled circles) and predicted (solid line) Hg concentrations as a function of age for Tilefish. Short dashed curves = 95% confidence intervals; Dark grey shaded area = "Choices to Avoid", grey shaded area = "Good Choices" for one meal a week, light grey shaded area = "Good Choices" for two meals a week, unshaded area = "Best Choices", per suggestion by the US EPA and US FDA (2017).

Figure 6. Observed (closed dots= females; open squares = males) Hg concentrations as a function of total length for Tilefish. Ellipses represent 50% and 95% confidence intervals for females (black) and males (grey).

Figure 1

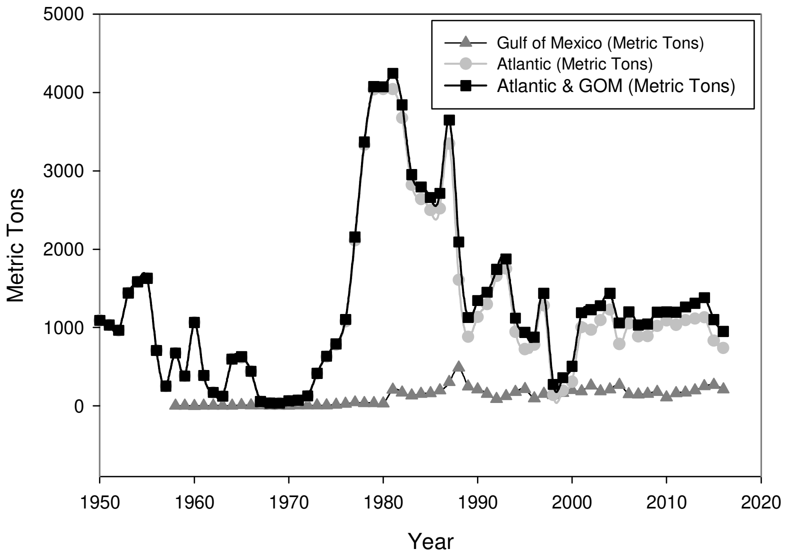
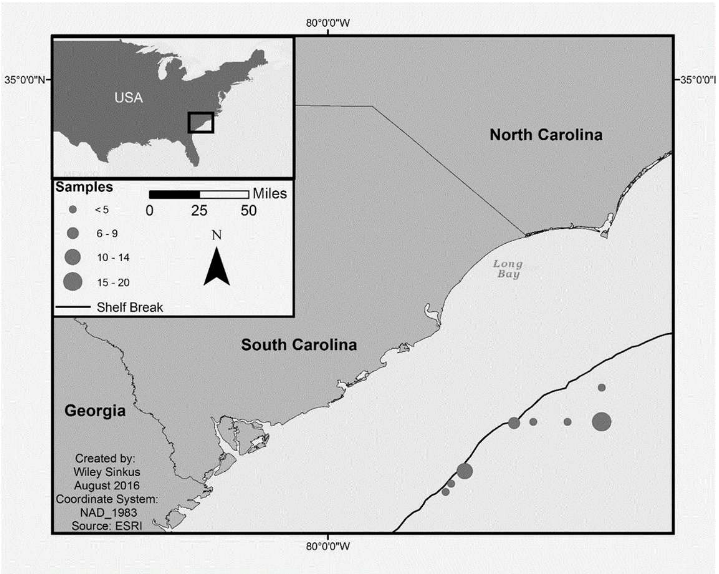


Figure 2


80°0'0"W
35°0'0"N
USA
North Carolina
Samples
< 5
6 - 9
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15 - 20
Shelf Break
Miles
0 25 50
N
Long Bay
South Carolina
Georgia
Created by:
Wiley Sinkus
August 2016
Coordinate System:
NAD_1983
Source: ESRI
80°0'0"W

Figure 3

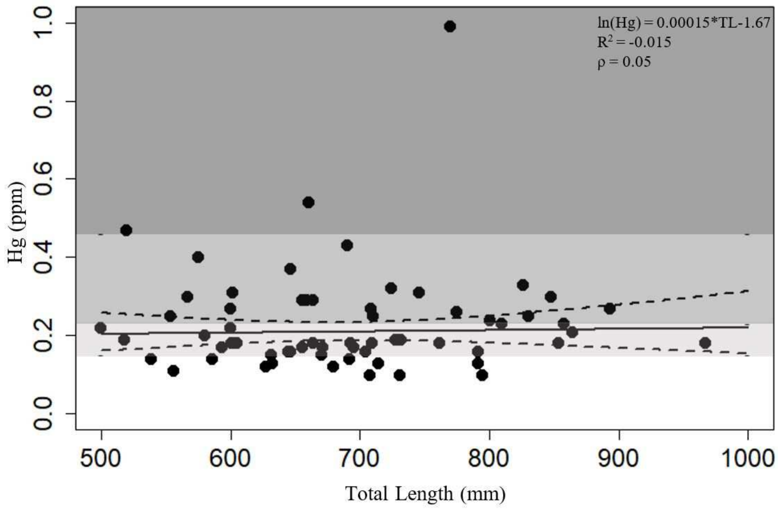

ln(Hg) = 0.00015*TL-1.67
R^2 = -0.015
ρ = 0.05
Hg (ppm)
0.0
0.2
0.4
0.6
0.8
1.0
500
600
700
800
900
1000
Total Length (mm)

Figure 4

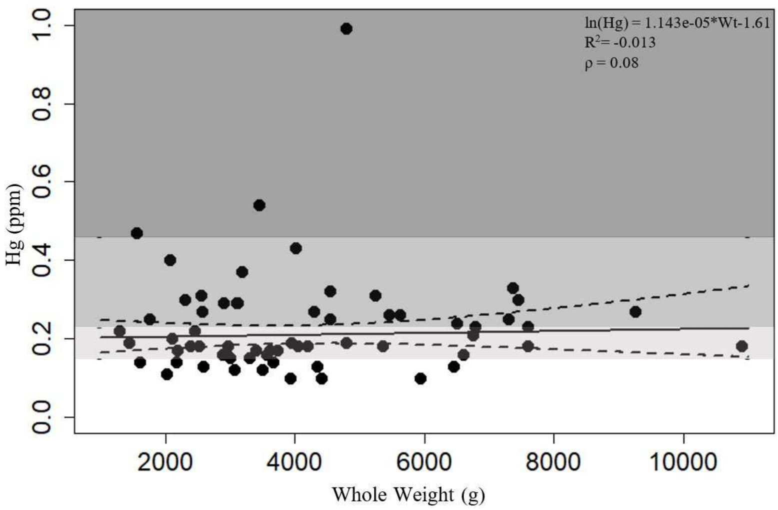


Figure 5

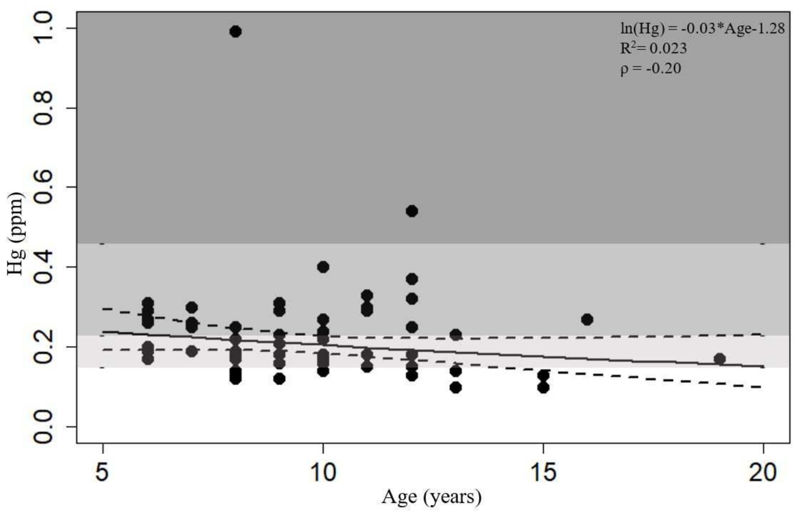
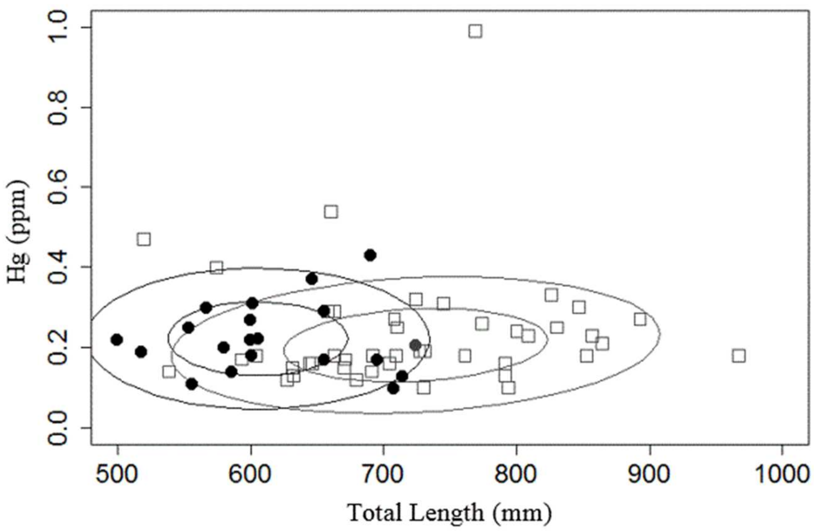


Figure 6



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Graphical Abstract: Percent of Tilefish samples with mercury concentrations within 0.1 ppm bins. Filled circle represents the mean and SE bars of samples from the current study and filled star represents mean mercury concentrations from Hall *et al.* 1978. Dark grey shaded area = "Choices to Avoid", grey shaded area = "Good Choices" for one meal a week, light grey shaded area = "Good Choices" for two meals a week, unshaded area = "Best Choices", per suggestion by the US EPA and US FDA (2017).