



Mercury levels of yellowfin tuna (*Thunnus albacares*) are associated with capture location[☆]



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ABSTRACT

Mercury is a toxic compound to which humans are exposed by consumption of fish. Current fish consumption advisories focus on minimizing the risk posed by the species that are most likely to have high levels of mercury. Less accounted for is the variation within species, and the potential role of the geographic origin of a fish in determining its mercury level. Here we surveyed the mercury levels in 117 yellowfin tuna caught from 12 different locations worldwide. Our results indicated significant variation in yellowfin tuna methylmercury load, with levels that ranged from 0.03 to 0.82 $\mu\text{g/g}$ wet weight across individual fish. Mean mercury levels were only weakly associated with fish size ($R^2 < 0.1461$) or lipid content ($R^2 < 0.00007$) but varied significantly, by a factor of 8, between sites. The results indicate that the geographic origin of fish can govern mercury load, and argue for better traceability of fish to improve the accuracy of exposure risk predictions.

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1. Introduction

Mercury is a widespread environmental toxicant of concern for wildlife and human health. Mercury occurs naturally and is also released into the atmosphere through anthropogenic sources, most notably the combustion of coal (Driscoll et al., 2013; Fitzgerald and Lamborg, 2007; Gustin et al., 2008; Mason et al., 1994; Selin, 2009). Atmospheric mercury is deposited into the oceans, where it is converted into inorganic and/or organic mercury (Mason et al., 1994). The inorganic mercury exhibits limited bioaccumulation, while the organic form (methylmercury) is highly refractory and bioaccumulates in high trophic level animals (Mason et al., 1995; Morel et al., 1998).

Methylmercury is a neurotoxicant in humans and wildlife (Bose-O'Reilly et al., 2010; Grandjean et al., 2010; Rice et al., 2014; Scheuhammer et al., 2007; Wiener and Spry, 1996), and is a reactive molecule that can bind to cellular proteins thereby increasing its half-life in the cell (Bošnjak et al., 2009; Mason et al., 1996). High levels of methylmercury in the human body have been associated with developmental disorders, including neurological effects

(Antunes Dos Santos et al., 2016; Counter and Buchanan, 2004; Karagas et al., 2012). Importantly, it is estimated that in the United States alone each year at least 316,000, and possibly as many as 637,000, children are born with umbilical cord blood levels of mercury sufficient to cause neurodevelopmental defects (Trasande et al., 2005).

Consumption of contaminated seafood is considered the major route of human exposure to mercury (Castro-González and Méndez-Armenta, 2008; Kraepiel et al., 2003; Rice et al., 2014), and fish consumption advisories routinely recommend a reduction of intake of high mercury fish by pregnant and nursing women. In addition, considerable attention has been given to understanding the distribution of mercury in commercial food fish (Colman et al., 2015; Hightower and Moore, 2003; Lowenstein et al., 2010; Yamashita et al., 2005), as high levels of mercury have been reported in large predatory fish species including swordfish, sharks, and tuna (FDA, 2014; García-Hernández et al., 2007; Kaneko and Ralston, 2007; Matthews, 1983).

Among these species, tunas are of primary importance, as they are one of the most widely consumed group of fish species in the world. Three species of tunas, skipjack (*Katsuwonus pelamis*), yellowfin (*Thunnus albacares*), and bigeye (*Thunnus obesus*) account for 93% of all tuna consumed (ISSF, 2015). Among these, yellowfin and bigeye are large predatory fish with high levels of mercury (Boush and Thieleke, 1983; Kaneko and Ralston, 2007; Kojadinovic et al., 2006; Kraepiel et al., 2003; Ordiano-Flores et al., 2011), often

Abbreviations: Hg, mercury; MeHg, methylmercury.

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Table 1
Mean total mercury and methylmercury concentrations in yellowfin tuna (*Thunnus albacares*). Listed are the means (\pm S.D.) of total mercury (Hg) and methylmercury (MeHg) in $\mu\text{g/g}$ wet weight from tuna caught at the 12 locations. In addition, the means of the standard lengths are listed in centimeter (cm) and the means of the lipid content in the tuna muscle in weight%. Significance letters of the pairwise multiple comparison of mean methylmercury levels across sampling sites using Tukey-Kramer HSD at $\alpha = 0.05$ are listed (A-D). For details on the p-values of the respective pairwise comparison, please refer to Table S1.

Sampling sites	Samples [n]	Total Hg [$\mu\text{g/g}$ ww]	MeHg [$\mu\text{g/g}$ ww]	Letters	Standard length [cm]	Lipids [wt%]
NPO	10	0.602 \pm 0.181	0.506 \pm 0.166	A	117.35 \pm 19.06	1.52 \pm 1.16
NEPO	10	0.154 \pm 0.026	0.164 \pm 0.024		55.87 \pm 1.97	3.25 \pm 1.12
GOM	9	0.23 \pm 0.171	0.246 \pm 0.176	B	103.72 \pm 13.36	1.23 \pm 1.04
SEPO	10	0.297 \pm 0.176	0.238 \pm 0.133	B	71.45 \pm 3.99	1.23 \pm 0.85
NWAO	10	0.43 \pm 0.085	0.345 \pm 0.077	B	92.8 \pm 3.49	0.41 \pm 0.16
NEAO	10	0.206 \pm 0.035	0.199 \pm 0.036	B	80.42 \pm 7.13	6.08 \pm 2.56
SEAO	8	0.348 \pm 0.101	0.308 \pm 0.088	B	84.94 \pm 23.22	2.76 \pm 1.38
IO	10	0.245 \pm 0.055	0.236 \pm 0.057	B	97.07 \pm 2.32 ^a	0.6 \pm 0.18
SCS	10	0.181 \pm 0.042	0.16 \pm 0.035	B	109.4 \pm 8.6	1.09 \pm 0.58
NCS	10	0.269 \pm 0.027	0.224 \pm 0.019	B	96.9 \pm 3.67	0.71 \pm 0.6
NWPO	10	0.064 \pm 0.016	0.064 \pm 0.018		100.45 \pm 1.15	0.68 \pm 0.2
SWPO	10	0.239 \pm 0.151	0.234 \pm 0.149	B	108.25 \pm 25.93	0.62 \pm 0.45

^a Fork length.

exceeding the EPA consumption advisory limit of 0.3 $\mu\text{g/g}$ wet weight (USEPA, 2010). For instance, in raw (i.e. sushi) tuna preparations collected from restaurants and supermarkets in New York, New Jersey, and Colorado, bigeye tuna with mercury levels up to 2.3 $\mu\text{g/g}$ wet weight were reported, while yellowfin samples had mercury levels as high as 1.4 $\mu\text{g/g}$ wet weight (Lowenstein et al., 2010).

Numerous factors can contribute to variation in mercury levels within fish, generally including species, size, migratory biology, and origin. While several studies in tuna have examined associations between fish size and mercury levels (Bosch et al., 2016; Drevnick et al., 2015), less is known about the impact of capture location, with most studies focusing on regional versus global differences (Colman et al., 2015; Cumont et al., 1972; Ferriss et al., 2011; Hall et al., 1978; Kojadinovic et al., 2006). In one recent study, juveniles of the highly migratory bluefin tuna (*Thunnus orientalis*) showed elevated levels of mercury upon arrival in the eastern Pacific Ocean, possibly consistent with exposure occurring during their development in the western Pacific Ocean (Colman et al., 2015). Thus, further characterization of geographic variation is essential to better understand how fish origin can govern mercury level.

Yellowfin tuna are distributed worldwide in the tropical and subtropical waters and provide an interesting case study in which to examine differences in mercury levels from different locations. Yellowfin are the second most widely consumed tuna species in the world and account for 27% of the global catch (ISSF, 2015). Like other large predatory fish, yellowfin can magnify mercury through trophic transfer up the food web, but because they do not migrate across oceans (Block et al., 2011), would be expected to reflect geographic differences in mercury levels.

To explore this variability, we measured mercury levels in white dorsal muscle tissue of 117 yellowfins caught at 12 different locations from around the globe. The results indicate that capture location of fish could be a critical factor in determining the mercury level of yellowfin and argue for improved traceability of these fish in the global food supply.

2. Material and methods

2.1. Tuna collection

Mixed-sex yellowfin tuna (*Thunnus albacares*) were collected from across the globe from the four known yellowfin stocks in the Atlantic Ocean, the Eastern Pacific, the Western Pacific, and the

Indian Ocean (ISSF, 2015). Collectors were either from Scripps Institution of Oceanography (SIO) or affiliated with local academic and commercial fisheries. The white dorsal muscle tissue was sampled from 12 locations, including the North East Pacific Ocean (NEPO; $n = 10$), Gulf of Mexico (GOM; $n = 9$), South East Pacific Ocean (SEPO; $n = 10$), Northwest Atlantic (NWAO; $n = 10$), Northeast Atlantic Ocean (NEAO; $n = 10$), South East Atlantic Ocean (SEAO; $n = 8$), Indian Ocean (IO; $n = 10$), South China Sea (SCS; $n = 10$), North China Sea (NCS; $n = 10$), Northwest Pacific Ocean (NWPO; $n = 10$), Southwest Pacific Ocean (SWPO; $n = 10$), and the North Pacific Ocean (NPO; $n = 10$). The target sampling size was 100 cm, translating to roughly 1–2 years old yellowfin (Kikkawa and Cushing, 2002). All tunas were either captured by trolling, purse seine or longline and coordinates for capture were recorded to within 100 km. Captured tuna were filleted and the dorsal muscle immediately frozen on ice. Tuna muscle were stored at -20 °C until shipped on dry ice to SIO. At SIO, all samples were stored at -80 °C at SIO, and subsamples of 56.7 g (2oz) or 113.4 g (4oz) were sent on dry ice to Brooks Rand Labs (BRL, Seattle, WA) for analysis of total mercury (Hg), methylmercury (MeHg).

2.2. Mercury analysis

All samples were received, prepared, analyzed, and stored per BRL standard operating procedures (SOPs) and Environmental Protection Agency (EPA) methodology. The method holding time requirement was met such that all samples were analyzed within one year from collection. Total mercury (Hg) and methylmercury (MeHg) were measured per EPA methods 1631E and 1630. Briefly, for total mercury detection all mercury was oxidized to Hg(II), purged, and trapped using a BRL MERX-T unit, and finally detected using a Brooks Rand Model III cold vapor atomic fluorescence spectrophotometer (CVAFS). For methylmercury detection, samples were purged and trapped using a BRL MERX-M unit, the released Hg species separated using gas chromatography (GC), thermally reduced to elemental mercury, and finally detected using a Brooks Rand Model III cold vapor atomic fluorescence spectrophotometer (CVAFS).

2.3. Data analysis

All samples were reported on a wet-weight basis. For methylmercury, the certified reference material was TORT-2 and DOLT-4 dogfish liver certified for trace metals. For total mercury, the certified reference material was DORM-3 fish protein certified for

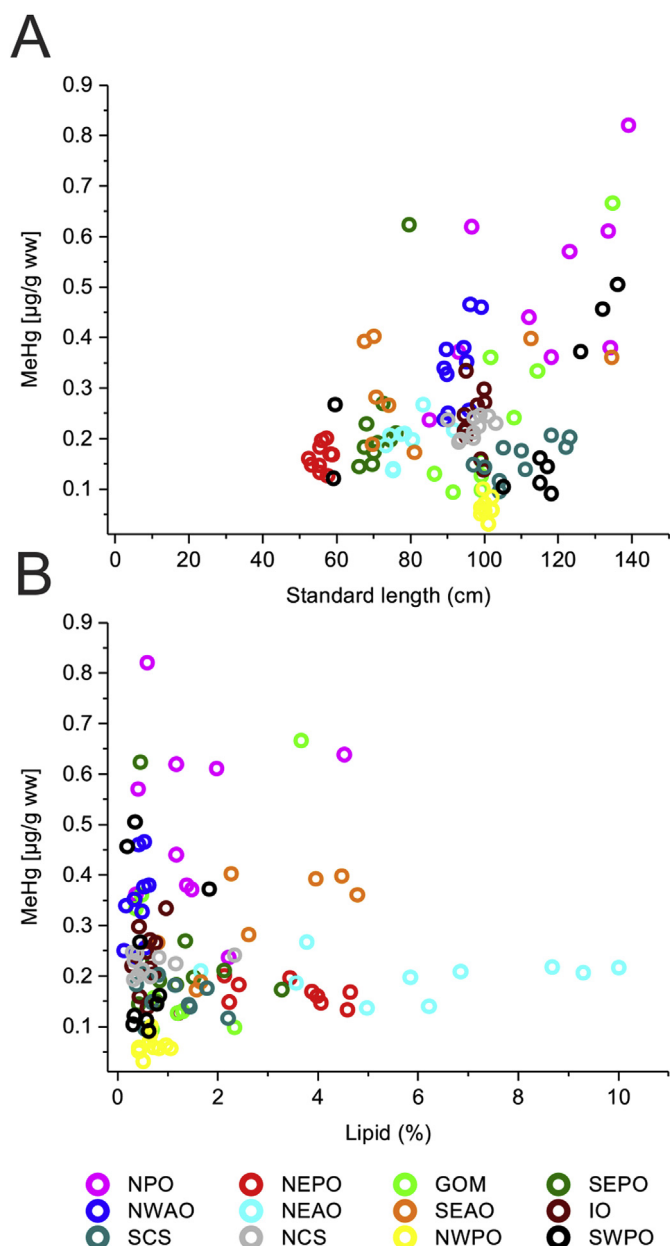


Fig. 1. Relationships between fish length and lipid content, versus methylmercury concentration. (A) Methylmercury concentration of each of the 117-individual fish versus standard length in cm ($R^2 < 0.1461$, Pearson's $r < 0.3822$). The slope of the fitted regression line was significantly different from zero ($p < 0.0001$). (B) Methylmercury concentration versus lipid content from 117 individual fish ($R^2 < 0.00007$, Pearson's $r < -0.00837$). The slope of the fitted regression line was not significantly different from zero ($p < 0.9286$).

trace metals. Duplicates and spike recovery were used to confirm reproducibility and accuracy of measurements. Method detection limits (MDLs) and method reporting limits (MRLs) were sample-specific and adjusted for the mass of sample aliquot digested and the volume of digestate analyzed. For total mercury, MDLs ranged from 0.54 to 4.82 ng/g and MRLs ranged from 1.8 to 16.1 ng/g wet weight. For methylmercury, MDLs ranged from 0.9 to 1.6 ng/g and MRLs ranged from 2.7 to 4.8 ng/g wet weight. The results of the analysis were method blank corrected per BRL standard operating procedure (SOP). For this, four method blanks were analyzed with every batch of samples and if the mean value and standard deviation of those blanks met the acceptance criteria (i.e. the mean was

< 2 ng/g wet weight and the standard deviation was < 0.7 ng/g wet weight), the mean of the method blank results was subtracted from the sample results. Consistent with previous studies, virtually all the mercury detected in tuna was in the form of methylmercury (Table 1).

To measure the correlation between methylmercury concentration and lipid content or length of the fish, we applied linear regression analysis and determined Pearson's correlation coefficient (r) and coefficient of determination (COD or R^2). To test the differences among sampling locations, we used one-way analysis of variance (ANOVA) followed by a Tukey post-hoc test. The null hypothesis of equal means between locations was rejected when the p -value was below a significance level of $\alpha = 0.05$. Detailed information on the ordered p -values for each pairwise comparison of sample locations from the Tukey-Kramer honest significant difference (HSD) test are provided in Table S1. All data were analyzed and plotted using the programs JMP Pro version 12 and OriginPro 2016.

3. Results and discussion

Here we measured mercury concentrations in white dorsal muscle of yellowfin tuna caught from 12 different locations worldwide and investigated the role of size and regional differences in mercury level. We selected for white muscle tuna samples since this tissue type has been shown to have more conservative mercury levels with lower variability as compared to dark muscle (Balshaw et al., 2008; Bosch et al., 2016). In addition, dorsal muscle tissue is considered a representative portion of fish for human consumption (Adams et al., 2003).

Fish ranged in length from 52.32 cm (10.1 kg) to 139.5 cm (186 kg), and methylmercury levels were only weakly associated with size ($R^2 < 0.1461$, Pearson's $r < 0.3822$, $p < 0.0001$) (Fig. 1A). For instance, in the case of the NWPO and the SCS, tuna had among the lowest methylmercury levels of all fish despite being among the largest fish caught (Table 1, Fig. 1A). A similar weak relationship has been observed between white muscle mercury concentrations and curved fork length of wild and penned Bluefin tuna (Colman et al., 2015) as well as body mass in yellowfin and bigeye tuna (Ferriss et al., 2011).

Likewise, across all 117 tuna, methylmercury levels showed no correlation with the lipid content ($R^2 < 0.00007$, Pearson's $r < -0.0084$, $p < 0.9286$) in the muscle tissue (Fig. 1B), consistent with an association of this compound with cellular proteins rather than fat (Amlund et al., 2007). Indeed, the fish with the highest methylmercury concentration of 0.821 $\mu\text{g/g}$ wet weight from the NPO had a low lipid content of less than 0.6 wt%.

In contrast, strong effects were observed when examining the impact of capture location on mean methylmercury levels. Several sites were significantly different from each other [$F = 10.89$, $P = < 0.0001$, with $df = (11, 105)$] (Figs. 2 and 3). Mean methylmercury levels across sites varied by a factor of 8 with the highest mean levels detected in fish from the NPO ($0.51 \pm 0.17 \mu\text{g/g}$ wet weight) and the lowest in fish from the NWPO ($0.06 \pm 0.02 \mu\text{g/g}$ wet weight) (Fig. 3, Table 1).

Among individual fish, methylmercury levels differed by a factor of 26, with the 20 most contaminated fish caught in the NPO, GOM, SEPO, SWPO, NAWAO, and SEAO. Among these, a cluster of eight fish were all caught in the NPO, with one fish having the highest methylmercury level of all fish with 0.821 $\mu\text{g/g}$ wet weight (Fig. 4). In contrast, the 20 least contaminated fish were caught in the NEPO, GOM, SWPO, SCS and NWPO. Notably, 10 out of these 20 least polluted fish were all caught in the NWPO, with the lowest methylmercury level of 0.032 $\mu\text{g/g}$ wet weight (Fig. 4).

Dynamic sources of both natural and anthropogenic sources of mercury contribute to the mercury in the environment. Two of the

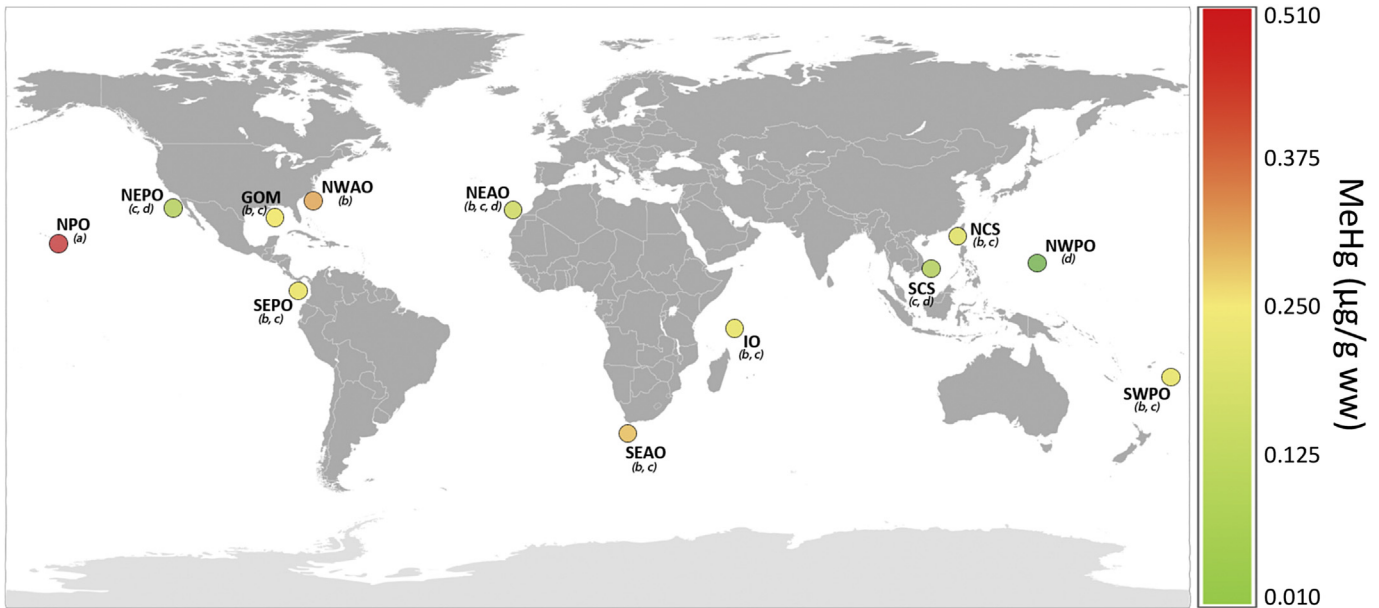


Fig. 2. The geographic differences in methylmercury levels of yellowfin tuna (*Thunnus albacares*). The heat map shows the mean levels of methylmercury for tuna caught from 12 different locations (µg/g wet weight). Groups with means that were significantly different from each other are indicated with letters ($\alpha = 0.05$).

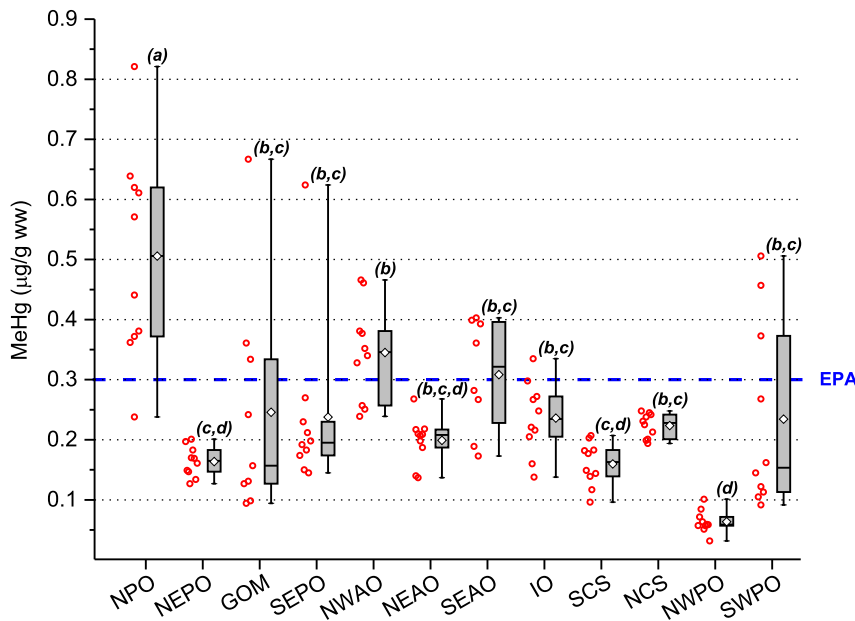


Fig. 3. Mean levels of methylmercury in yellowfin tuna vary significantly by location of capture. (A) Box and whisker plots represent the mass-based ranges of methylmercury at the 12 capture locations. The horizontal line represents the 50th percentile, and the box represents the 25th and 75th percentiles. The white diamonds show the mean values and the bars at the end of the whiskers represent the minimum and maximum values. The red hollow spheres to the left of each boxplot display the values for each individual fish. EPA's maximum advisable methylmercury fish tissue concentration is marked as a blue-dashed line. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

more contaminated sites were relatively distant from human terrestrial inputs, including the NPO and SWPO. However, these sites could be downwind of atmospheric sources of mercury originating in Asia (Strode et al., 2008; Sunderland et al., 2009), and it is estimated that more than half the current global anthropogenic mercury emissions originate from the Asian continent (Pirrone et al., 2010; Selin, 2009; Streets et al., 2009). In addition, given their proximity to volcanic activity there could be additional natural sources of mercury at these locations (Fitzgerald and Lamborg, 2007; Gustin et al., 2008; Pyle and Mather, 2003; Selin, 2009).

As with previous studies we also detected relatively high levels of methylmercury in the fish caught from the GOM and NAWO, suggesting anthropogenic sources of mercury contamination in these fish. Indeed, this might be consistent with mercury contamination via the Atlantic Ocean Loop Current and/or mercury originating in the Mississippi River and being drained into the GOM (Buck et al., 2015; Harris et al., 2012a, 2012b).

An important factor to consider is that ecological variables, in addition to mercury sources, could also play an important role in governing the differences among sites. For instance, although we

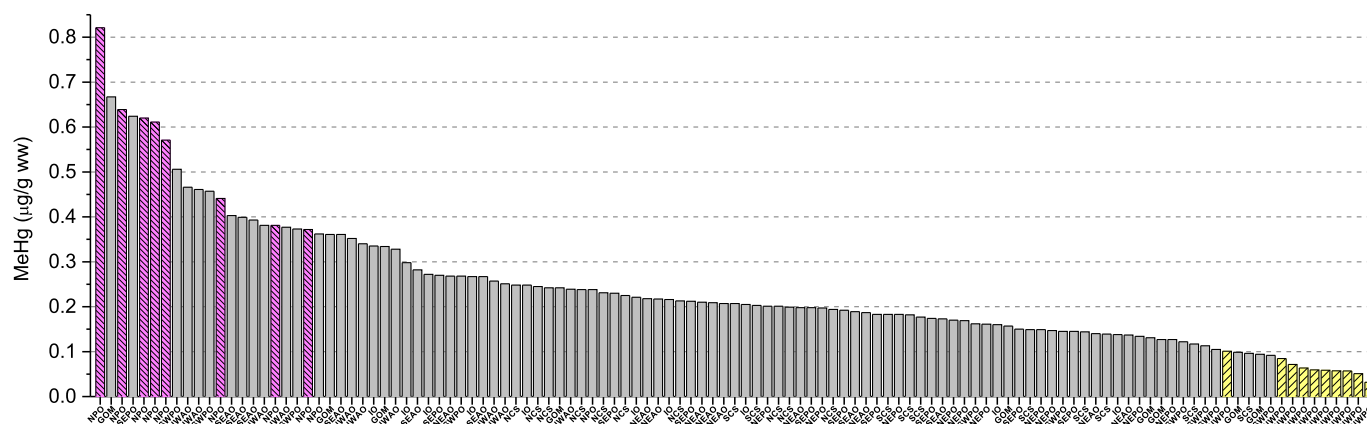


Fig. 4. Individual tuna methylmercury levels indicate clustering by location of catch. Shown are the ranked methylmercury levels across the 117-individual tuna. Eight of the top 20 contaminated fish (pink pattern) were caught in the NPO, while all 10 tuna from the NWPO contributed to the bottom 20 contaminated fish (yellow pattern). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

looked at a single species, it is well appreciated that differences in population structure among the different sites could influence the actual trophic level of fish (Frank et al., 2005; Layman et al., 2007; Post, 2002). This shift in trophic level could lead to variations in the diet of tuna at different sites. While 1–2-year-old tuna typically feed on smaller fish, crustaceans, squid and gelatinous organisms (Bertrand et al., 2002; Fiedler and Bernard, 1987; Ménard et al., 2006; Olson and Boggs, 1986; Perrin et al., 1973), some recent studies on yellowfin tuna show a more opportunistic feeding behavior, depending on the availability of prey (Rohit et al., 2010; Weng et al., 2016). Notably, the pattern of mercury accumulation in our analyzed yellowfin tuna does not follow the spatial pattern of mercury concentration in the respective thermocline water bodies (Lamborg et al., 2014), indicating that some combination of trophic status, i.e. feeding behavior, and geographic location could be driving the mercury concentrations in these fish.

An implication of our study is that geographic origin of wild-caught fish could be an important factor that governs the potential benefits and risks of fish consumption. It is already well known that there can be significant differences in mercury levels across different fish species, and these differences have been a key element in consumption advisories (USEPA, 2010, 2000). Our study highlights how levels can vary within a single species. In seven locations, the NPO, GOM, SEPO, NWAQ, SEAO, IO, and SWPO, the individual fish methylmercury levels often exceeded the EPA's human health criterion (USEPA, 2010) of 0.3 µg/g wet weight (Fig. 3) and overall 24% (28 fish) were above EPA's methylmercury criterion to protect consumers of fish and shellfish (Figs. 3 and 4). None of the fish had methylmercury levels above current FAO/WHO guidelines and FDA action levels of 1 µg/g wet weight (FDA, 2011; Food and Agriculture Organization (FAO), 2003). However, using this type of geographic information to guide consumption advisories remains a challenge since tracking of fish from capture location to the point of sale is limited (Bailey et al., 2016; Jacquet and Pauly, 2008), particularly for globally distributed species such as yellowfin tuna which are sold in a variety of products, including fresh or frozen steaks and canned.

4. Conclusions

This study provides a snapshot of mercury levels in wild yellowfin tuna from around the globe. The results indicate that the geographic origin of catch is a critical factor in determining the mercury levels of fish within species. Thus, human exposure risk to

dietary mercury from tuna might be better managed if future guidelines and advisory levels for consumption are informed by the capture location and not just the species and means of production. In such a scenario, mercury exposure might be minimized in vulnerable populations, such as pregnant and nursing women, by providing a means to identify clean sources in the food supply.

Contributions

S.C.T.N., S.S. and A.H. conceived and designed the study. S.C.T.N. and A.H. analyzed the data and conducted statistical analyses. S.C.T.N. and A.H. wrote the manuscript. L.T.B. collected fish samples.

Conflict of interest

The authors declare they have no actual or potential competing financial interests.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at <http://dx.doi.org/10.1016/j.envpol.2017.05.070>.

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