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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 μg/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.

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

[a] Fork length.

exceeding the EPA consumption advisory limit of 0.3 μg/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 μg/g wet weight were reported, while yellowfin samples had mercury levels as high as 1.4 μg/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