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Trophic influences on mercury accumulation in top pelagic predators from offshore New England waters of the northwest Atlantic Ocean

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ABSTRACT

Trophic pathways and size-based bioaccumulation rates of total mercury were evaluated among recreationally caught albacore tuna (*Thunnus alalunga*), yellowfin tuna (*Thunnus albacares*), shortfin mako shark (*Isurus oxyrinchus*), thresher shark (*Alopias vulpinus*), and dolphinfish (*Coryphaena hippurus*) from offshore southern New England waters of the northwest Atlantic Ocean between 2008 and 2011. Mercury concentrations were highest in mako (2.65 ± 1.16 ppm) and thresher sharks (0.87 ± 0.71 ppm), and significantly lower in teleosts (albacore, 0.45 ± 0.14 ppm; yellowfin, 0.32 ± 0.09 ppm; dolphinfish, 0.20 ± 0.17 ppm). The relationship between body size and mercury concentration was positive and linear for tunas, and positive and exponential for sharks and dolphinfish. Mercury increased exponentially with $\delta^{15}N$ values, a proxy for trophic position, across all species. Results demonstrate mercury levels are positively related to size, diet and trophic position in sharks, tunas, and dolphinfish, and the majority of fishes exhibited concentrations greater than the US EPA recommended limit.

1. Introduction

In contrast to the wealth of knowledge that exists regarding mercury bioaccumulation in freshwater ecosystems, trophic pathways and rates of accumulation are poorly understood in the marine environment (Doyon et al., 1998; Harris and Bodaly, 1998; Greenfield et al., 2001; Da Silva et al., 2005; Walters et al., 2010). The North Atlantic Ocean has been identified as a region of high dissolved mercury concentrations relative to the Pacific, Southern and Arctic Oceans, which has implications for enhanced bioavailability to marine food-webs in this region (Lamborg et al., 2014).

The burning of fossil fuels is a primary and continued source of mercury in the atmosphere (USGS, 2000; Varekamp et al., 2003). Direct atmospheric deposition as well as runoff along the east coast of the United States further contribute to inorganic mercury loading in coastal and offshore waters of the northwest Atlantic (Van Arsdale et al., 2005; Driscoll et al., 2007; Krabbenhoft and Rickert, 2009; US EPA, 2010; Sackett et al., 2010). Microbial methylation of inorganic mercury occurs in coastal sediments and below the surface mixed layer of open ocean waters (Chen et al., 2008; Blum et al., 2013). Mercury is bioavailable in its methylated form and incorporated into the marine food chain by phytoplankton particulate uptake (Chen et al., 2009; Krabbenhoft and Rickert, 2009). Methylmercury moves between inshore and offshore food-webs through a 'trophic relay' (Kneib, 1997; Chen et al., 2008) and transfer to higher trophic levels is markedly efficient (Driscoll et al., 2007). Diet is the primary pathway for toxic mercury accumulation in both humans and fish; therefore, characterizing mercury sources and accumulation rates in food-webs targeted by fisheries is extremely important for human and ecosystem health assessments (Cabana et al., 1994; Hall et al., 1997; Lepak et al., 2009; Payne and Taylor, 2010). Elevated mercury burdens in humans and animals have been linked to endocrine, neural, and reproductive impairment (Porto et al., 2005; Scheulhammer et al., 2007; Crump and Trudeau, 2009; Tan et al., 2009; Veldhoen et al., 2013).

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Mercury burdens generally increase with body size, age, and trophic position (Piraino and Taylor, 2009; Suk et al., 2009; Taylor et al., in press). However, rates of accumulation vary widely among species and systems due to differences in metabolism, habitat use, life histories and diets (Watras et al., 1998; Burger, 2009; Verdouw et al., 2011). For example, globally-distributed dolphinfish (*Coryphaena hippurus*) have been found to carry lower mercury concentrations at lower latitudes of the North Atlantic Ocean (e.g. Caribbean Sea, Gulf of Mexico; Cai et al., 2007; Adams, 2009) than those measured at higher latitudes of the Mid-Atlantic Bight (Burger and Gochfeld, 2011). Pelagic predator fishes can also exhibit variation in prey composition and size across geographic regions (Olson and Galván-Magaña, 2002) and with growth (Graham et al., 2007). These differences likely contribute to observed spatial and temporal variation in mercury accumulation rates, respectively.

Certain aspects of feeding ecology, such as the degree of piscivory and average prey size in regional diets, are potential indicators of predator mercury concentrations. Piscivorous predators have been shown to have higher concentrations of mercury in their tissues compared to those that feed on invertebrates and other lower trophic level prey (de Pinho et al., 2002; Adams, 2010). Prey size can also affect mercury bioaccumulation of predators, as larger prey are often older and/or feeding at higher trophic levels, thus carrying greater mercury burdens (Piraino and Taylor, 2009; Suk et al., 2009). Additionally, geographic variability in bioavailable mercury can influence mercury accumulation rates and tissue concentrations of migratory fishes across their range (Verdouw et al., 2011). Because tissue mercury concentrations measured at one time point or location represent cumulative uptake across a species' life span and geographic range, regional assessments of fish mercury content and feeding ecology are needed to identify the potential pathways of mercury transfer within marine food-webs. Information from multiple metrics of diet determination representing short- and long-term feeding habits (such as stomach contents and stable isotope analyses, respectively) can be used to more comprehensively characterize these pathways of mercury accumulation in highly migratory predator fishes.

In the northwest Atlantic Ocean, large pelagic fishes migrate seasonally to offshore waters of the southern New England region to feed upon abundant prey resources (Thompson, 1999; Natanson, 2002; MacNeil et al., 2005; ICCAT, 2010). Intense recreational and commercial fishing effort is focused on these pelagic predators during the summer and fall. In particular, dolphinfish, albacore (*Thunnus alalunga*) and yellowfin (*Thunnus albacares*) tunas, shortfin mako (*Isurus oxyrinchus*) and common thresher (*Alopias vulpinus*) sharks are targeted by recreational anglers for sport fishing tournaments where the primary goal is to catch the largest fish possible. Such practices raise human health concerns given the potentially high mercury concentrations of large fish and the high frequency of catch consumption by coastal communities of the northeast United States (Steinback et al., 2009).

As the impacts of climate change continue to grow, increasing ocean temperatures and acidification are impacting mercury cycling and methylation, potentially leading to altered bioaccumulation rates of mercury by marine biota (Deser and Blackmon, 1993; Levitus et al., 2000; Doney et al., 2009; Dijkstra et al., 2013). Climate change is also causing species to shift their geographical ranges, the timing of seasonal migrations, leading to altered trophic interactions (Collie et al., 2008; Nye et al., 2009; Fodrie et al., 2010). Therefore, baseline studies that provide information on trophic pathways and rates of contaminant bioaccumulation are increasingly needed to detect and track shifts as environmental conditions and ecological communities continue to change (Staudinger et al., 2013b). The trophic ecology and mercury accumulation of top predators in offshore waters of southern New England have been poorly studied due in part to logistical difficulties in sampling and to the limited time period these fishes spend in this region. Mercury analysis of tuna and dolphinfish at this northern extent of their migratory range contributes further information relating to the cumulative nature of mercury accumulation by complementing previous studies focused in southern regions of the Atlantic Ocean, including the South Atlantic Bight and Gulf of Mexico (e.g. Adams, 2009; Senn et al., 2010; Petre et al., 2012).

To increase our understanding of the trophic pathways and mercury bioaccumulation in dolphinfish, albacore and yellowfin tunas, and mako and thresher sharks, total mercury was measured in the edible muscle tissues of these fishes. Interspecific differences in the relationships between mercury concentration, body size, diet, and feeding ecology were quantified. Results are discussed in the context of both human and ecosystem health, and are intended to assist fishers and the public in making informed decisions about the fish they catch and consume.

2. Materials and methods

2.1. Sample collection and processing

Dorsal muscle samples were collected from dolphinfish, tunas, and sharks landed dockside at recreational fishing tournaments on Cape Cod and the Islands of Massachusetts during summer and early fall (July, August and September) of 2008–2011. Tissue samples were removed, immediately placed on ice, and frozen until laboratory analyses were conducted. In addition, eight prey species were selected for mercury analysis based on their nutritional importance (percent weight) and population scale feeding habits (percent occurrence) (Teffer, 2012). Atlantic herring (*Clupea harengus*), round herring (*Etrumeus teres*), shortfin squid (*Illex illecebrosus*), longfin squid (*Loligo pealeii*), spiny dogfish (*Squalus acanthias*), Atlantic butterfish (*Peprilus triacanthus*), silver hake (*Merluccius bilinearis*) and bluefish (*Pomatomus saltatrix*) were collected from bottom trawl surveys conducted by the National Marine Fisheries Service (NMFS) during fall of 2008 and 2011. Predator weights were taken dockside (kg). Predator lengths were measured as curved fork length (CFL), prey fish as total length (TL), and squid as mantle length (ML). All body lengths were measured in centimeters (cm).

2.2. Mercury analysis

For all predator and prey specimens, a 1.0 g subsample of dorsal muscle tissue (dorsal mantle from squid) was excised and weighed (±0.0001 g), freeze-dried for 24 h in a Labconco freeze dry system, re-weighed to assess moisture content, and homogenized using mortar and pestle. All samples were analyzed for total mercury (ppm dry weight) using a Milestone DMA-80 Mercury Analyzer (Cizdziel et al., 2002). This method utilizes thermal decomposition, amalgamation, and atomic absorption spectrophotometry (EPA method 7473; US EPA, 1998), with an instrument detection limit of 0.01 ng mercury. Certified reference materials (CRMs) prepared by the National Research Council Canada, Institute of Environmental Chemistry (Ottawa, Canada) were used to calibrate the DMA-80 and included TORT-1 (lobster hepatopancreas) and DORM-2 (dogfish muscle) (US EPA, 1998). Calibration curves were highly linear (mean $r^2 = 1.00$; range $r^2 = 0.99–1.00$; $P < 0.0001$), and the recovery of independently analyzed samples of DORM-2, DOLT-3 (dogfish liver), and NIST 2702 (marine sediment) CRMs ranged from 85.2% to 99.4% (mean = 93.7%). All samples were analyzed as duplicates, and an acceptance criterion of 10% was implemented.