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Methylmercury in dried shark fins and shark fin soup from American restaurants

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HIGHLIGHTS

- Concentrations of monomethylmercury (MMHg) in fins and soup varied among shark species.
- MMHg was highest in fins and soup from large, high trophic level sharks.
- Estimated human exposures to MMHg from shark fin soup are relatively low.

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ABSTRACT

Consumption of meat from large predatory sharks exposes human consumers to high levels of toxic monomethylmercury (MMHg). There also have been claims that shark fins, and hence the Asian delicacy shark fin soup, contain harmful levels of neurotoxic chemicals in combination with MMHg, although concentrations of MMHg in shark fins are unknown. We measured MMHg in dried, unprocessed fins ($n = 50$) of 13 shark species that occur in the international trade of dried shark fins as well as 50 samples of shark fin soup prepared by restaurants from around the United States. Concentrations of MMHg in fins ranged from 9 to 1720 ng/g dry wt. MMHg in shark fin soup ranged from <0.01 to 34 ng/mL, with MMHg averaging 62 ± 7% of total Hg. The highest concentrations of MMHg and total Hg were observed in both fins and soup from large, high trophic level sharks such as hammerheads (*Sphyrna* spp.). Consumption of a 240 mL bowl of shark fin soup containing the average concentration of MMHg (4.6 ng/mL) would result in a dose of 1.1 μg MMHg, which is 16% of the U.S. EPA's reference dose (0.1 μg MMHg per 1 kg per day in adults) of 7.4 μg per day for a 74 kg person. If consumed, the soup containing the highest measured MMHg concentration would exceed the reference dose by 17%. While shark fin soup represents a potentially important source of MMHg to human consumers, other seafood products, particularly the flesh of apex marine predators, contain much higher MMHg concentrations and can result in substantially greater exposures of this contaminant for people.

1. Introduction

Many marine apex predators contain high concentrations of mercury (Hg) and are potential sources of this contaminant for humans (Fitzgerald and Clarkson, 1991). Monomethylmercury (MMHg), as opposed to complexes of inorganic Hg, is the toxic form of Hg bioaccumulated by fish (Bloom, 1992). Shark muscle, for example, can contain concentrations of MMHg (~0.5–50 μg/g wet weight; Hornung et al., 1993; Storelli et al., 2003) that are far greater than the recommended human consumption advisory limit for MMHg in fish (0.3 μg/g; U.S. EPA, 2001). In addition to neurotoxicity, effects of MMHg may include increased risk of cardiovascular disease in adults who eat fish (Salonen et al., 1995; Karagas et al., 2012). Moreover, maternal transfer of MMHg to prenatal life stages can inhibit the neurological and cardiovascular development and growth of some children (Grandjean et al., 1997; Murata et al., 1999; Sorenson et al., 1999; Steuerwald et al., 2000; Karagas et al., 2012).

Although it is well established that shark meat contains high values of Hg, less is known about concentrations in other shark body parts that are consumed by humans. Dried shark fins are among the most highly traded and consumed shark parts in the world. They are used to make the Asian delicacy - shark fin soup, a luxury dish that is often served as an appetizer. This expensive delicacy consists mostly of ceratotrichia (or "fin needles"; elastin and collagen fibers that resemble rice noodles when cooked) served with either chicken broth or another stock for flavor (Rose, 1996). Recent claims have been made that shark fin soup can

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contain elevated levels of contaminants, potentially putting consumers at risk (Holtcamp, 2012; Mondo et al., 2012). To date, however, there are no published studies documenting Hg concentrations in either shark fins or shark fin soup. Our objective was to examine concentrations of total Hg and MMHg in dried unprocessed shark fins from wild captured animals, and shark fin soup, offered for retail sale in the United States, and evaluate potential health risk to human consumers.

2. Materials and methods

2.1. Samples

All samples were collected using standard clean procedures for Hg analysis. Whole, dried unprocessed shark fins examined in this study were collected from wild-captured specimens taken from fisheries of the United States, Sri Lanka, Fiji, and South Africa as part of a separate study. All fins were sun-dried. Fins were stored at Stony Brook University in separate plastic bags at room temperature for 12 to 18 months prior to sampling. A small piece (~10 mm × 10 mm) of the distal portion of each fin was analyzed for Hg content. Fins originated from one of 13 shark species, all of which are known to occur in the international dried fin trade (Table 1).

Bowls of shark fin soup were purchased for Hg analysis from 50 restaurants located throughout the United States by volunteers in March and April 2012 (Table S1). Either species or genus of origin of the fin used to make each bowl of soup was assessed using a mini-DNA barcode protocol on a single, randomly selected fin needle taken from each bowl (Fields et al., in review). Subsamples of soup for Hg speciation analysis were collected in 50-mL polypropylene centrifuge tubes that were cleaned with a rigorous technique suitable for trace-level Hg analysis (Hammerschmidt et al., 2011).

2.2. MMHg analysis

Dried, unprocessed fin samples (0.05–0.3 g dry wt.) were freeze dried and broken into pieces by hand while inside zip-type plastic bags and weighed into 15-mL centrifuge tubes containing 7 mL of 4.57 M HNO_3 (Hammerschmidt and Fitzgerald, 2006). Soup samples were shipped frozen to Wright State University, where soup was thawed and homogenized in a stainless steel blender cleaned with detergent and rinsed with reagent-grade water (resistivity ≥ 18 MΩ·cm). Separate aliquots of the homogenate were either freeze-dried for the analysis of carbon and nitrogen contents or digested with HNO_3 (4.57 M final concentration) for MMHg determination. MMHg was measured by flow-injection gas-chromatographic cold vapor atomic fluorescence spectrometry (CVAFS) after derivatization with sodium tetraethylborate (Bloom, 1989; Tseng et al., 2004).

Quality assurance of MMHg determinations included analyses of procedural blanks and standards, procedural replicates, and certified reference materials from the National Research Council of Canada, lobster hepatopancreas (TORT-2) and fish protein (DORM-3). Standard solutions of MMHg were calibrated using a Hg(II) solution traceable to the U.S. National Institute of Standards and Technology (NIST). All measurements of MMHg in DORM-3 ($n = 27$ samples) and all but one measurement of TORT-2 ($n = 30$) were within the certified ranges. The mean (±1SD) measured concentration of MMHg in DORM-3 was 329 ± 21 ng/g dry wt. (certified range = 299–411 ng/g) and that in TORT-2 was 150 ± 7 ng/g (certified range = 139–165 ng/g). Imprecision of MMHg analyses between procedural duplicates of soup averaged 7.7 relative percent difference ($n = 7$). The estimated detection limit for MMHg was ~0.01 ng/mL in soup and 0.02 ng/g for a 0.1-g sample of dried fin.

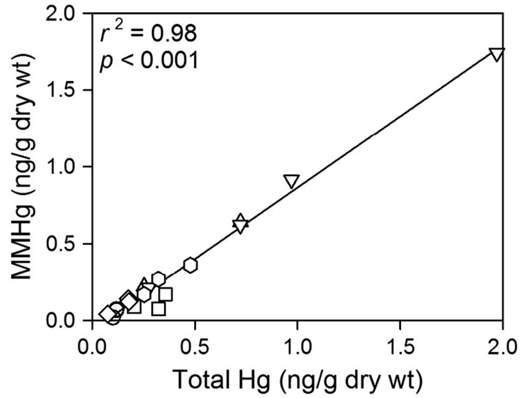

Fig. 1. MMHg is strongly related to total Hg in distal clips of dried shark fins: circle = blue shark (*Prionace glauca*), square = bull shark (*Carcharhinus leucas*), triangle up = copper shark (*Carcharhinus brachyurus*), triangle down = scalloped hammerhead (*Sphyrna lewini*), diamond = smooth hammerhead (*Sphyrna zygaena*), hexagon = shortfin mako (*Isurus oxyrinchus*).

Table 1
Mean ± 1SD concentration of monomethylmercury (MMHg) in distal fin clips of shark genera examined in this study. Ranges are given in parentheses.

Scientific name	Common name	*n*	MMHg (ng/g dry wt.)
Carcharhinus plumbeus	Sandbar shark	5	38 ± 16 (13–55)
Alopias vulpinus	Common thresher	6	87 ± 82 (9–239)
Sphyrna zygaena	Smooth hammerhead	3	101 ± 52 (42–140)
Carcharhinus leucas	Bull shark	3	111 ± 51 (74–170)
Carcharhinus brevipinna	Spinner shark	4	112 ± 72 (11–173)
Carcharhinus obscurus	Dusky shark	6	158 ± 66 (39–230)
Carcharodon carcharias	Great white	4	179 ± 146 (83–396)
Prionace glauca	Blue shark	6	247 ± 277 (19–763)
Isurus oxyrinchus	Shortfin mako shark	3	267 ± 97 (169–362)
Carcharhinus longimanus	Oceanic whitetip	2	299 (67–530)
Sphyrna mokarran	Great hammerhead	2	337 (301–372)
Carcharhinus brachyurus	Copper shark	2	432 (222–642)
Sphyrna lewini	Scalloped hammerhead	4	869 ± 639 (214–1720)

2.3. Total Hg analysis

The 4.57 M HNO_3 digestates of all soup samples and 18 of the fins also were used to quantify total Hg. Aliquots of the digestates were oxidized with BrCl for 12 h prior to the addition of NH_2OH (Hammerschmidt and Fitzgerald, 2006). Sample Hg was reduced with $SnCl_2$ and quantified by dual-Au amalgamation CVAFS (Fitzgerald and Gill, 1979; Bloom and Fitzgerald, 1988).

Total Hg determinations were calibrated with aqueous Hg(II) solutions traceable to the U. S. NIST. All measurements of DORM-3 ($n = 22$) and TORT-2 ($n = 28$) were within the certified ranges: Total Hg in DORM-3 averaged 362 ± 12 ng/g (certified range = 322–442 ng/g) and that in TORT-2 averaged 271 ± 12 ng/g (certified range = 210–330 ng/g). Recovery of known Hg(II) additions averaged 101% ($n = 7$), and imprecision among procedural replicates

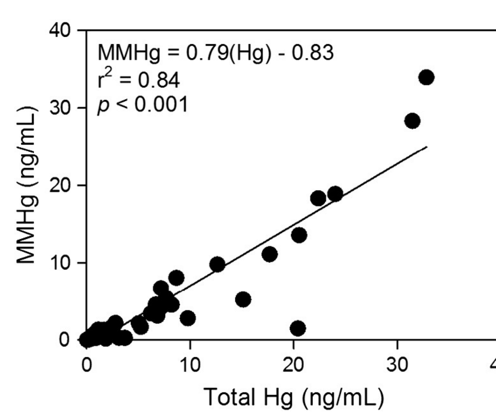

Fig. 2. Relationship between MMHg and total Hg in shark fin soup.

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