# BIOACCUMULATION OF MERCURY IN PELAGIC SHARKS FROM THE NORTHEAST PACIFIC OCEAN

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## ABSTRACT

The common thresher, Alopias vulpinus, and shortfin mako, Isurus oxyrinchus, are large pelagic shark species frequently targeted by sport and commercial fisheries. Being top marine predators, the common thresher and shortfin mako are susceptible to bioaccumulation of heavy elements in their tissues. We investigated the levels of mercury (Hg) in the tissues of these sharks from the central and eastern North Pacific Ocean and how those levels reflect their feeding ecology. For both species we found detectable levels of Hg in the white muscle but not in the liver, and no differences in Hg levels between the sexes, which suggests similar feeding patterns. There was, however, a significant interspecific difference with the shortfin mako having considerably higher Hg levels than the common thresher. This likely reflects the shortfin mako's opportunistic feeding on higher trophic level prey, such as jumbo squid (*Dosidicus gigas*), relative to the common thresher which primarily targets smaller schooling fish. We found strong linear relationships between body size and Hg level for both species with a significantly greater rate of increase for the shortfin mako which also may suggest a higher daily ration. In all common thresher samples, Hg levels were well below the U.S. Food and Drug Administration's established action level of 1.0 µg/g for commercial fish. Nearly all shortfin mako muscle samples from sharks  $\leq 150$  cm fork length (FL) had Hg levels below 1.0  $\mu$ g/g, but all shortfin mako >150 cm FL had muscle Hg levels exceeding this level, with the largest sharks having nearly three times this level.

## INTRODUCTION

The common thresher, *Alopias vulpinus*, and the shortfin mako, *Isurus oxyrinchus*, are active, strong-swimming endothermic pelagic sharks within the order Lamniformes, the mackerel sharks (Compagno 1984; Smith et al. 2008). The common thresher occurs throughout the temperate northeast Pacific Ocean with all sizes occurring typically within 72–135 km of land (Smith et al. 2008). The shortfin mako also occurs throughout the temperate and tropical northeast Pacific Ocean with juvenile and sub-adult sharks occurring inshore while larger sharks occur further offshore (Compagno 2001). Off the California coast, the common thresher feeds mostly on small schooling fish and cephalopods (Preti et al. 2001) while the shortfin mako is thought to be more opportunistic and mainly preys on a large variety of cephalopods and fish, with some large adults preying on marine mammals (PFMC 2003). Thus, both species represent large predators in the marine food web but with slightly different foraging ecologies, which may affect mercury (Hg) bioaccumulation.

Having high market values, both the common thresher and shortfin mako are harvested by commercial fisheries along the coast of California (Holts et al. 1998: Compagno 2001). Highly regarded as sport fish, they are also targeted by anglers in southern California. A seasonal California drift gillnet fishery for broadbill swordfish, *Xiphias gladius*, lands large numbers of these sharks, which are considered secondary targets (Smith and Aseltine-Neilson 2001; Taylor and Bedford 2001). In 2006, U.S. West Coast commercial fisheries landed 159 mt of common thresher and 46 mt of shortfin mako (PFMC 2007). No reliable landings data are available for the recreational catch.

The high trophic positions and market value of these sharks suggest that it would be worthwhile to determine their mercury levels. Environmental levels of Hg, a toxic metal with no known essential function in vertebrates, have increased dramatically since the onset of the Industrial Revolution. Current anthropogenic activities are estimated to account for about 66% of all the mercurv released into the environment annually (Wiener and Spry 1996; Jackson 1997; Downs and Lester 1998). In the marine environment, methylation processes by microorganisms convert inorganic Hg into the more toxic methylmercury (MeHg) (Beckvar et al. 1996; Wiener and Spry 1996). MeHg bioaccumulates in fish, marine mammals and birds due to its rapid uptake and slow rate of elimination (Mason et al. 1995). Studies have shown that high trophic-level fish, such as sharks (Marcovecchio et al. 1991; Hueter et al. 1995), billfishes (Monteiro and Lopes 1990), tunas (Storelli and Marcotrigiano 2004) and large mackerels (Meaburn 1978<sup>1</sup>) are particularly prone to bioaccumulate relatively

<sup>&</sup>lt;sup>1</sup>G. M. Meaburn. 1978. Heavy metal contamination of Spanish and King mackerel. *In* Proceedings of the mackerel colloquium, March 16, 1978, Charleston, Lab., SEFSC, Charleston, SC. 61–66 pp.



Figure 1. Shortfin mako (*Isurus oxyrinchus*), common thresher (*Alopias vulpinus*), bigeye thresher (*Alopias superciliosus*), and sevengill (*Notorynchus cepedianus*) sample collection locations from the coast of California; expanded image is the Southern California Bight.

high levels of MeHg in their tissues. Thus, the common thresher and shortfin mako may also present a dietary source of Hg to humans.

Despite this, data on Hg levels in these two species are limited or nonexistent and no data are available for the northeast Pacific Ocean. We found no published studies on the Hg levels in the common thresher. Kaneko and Ralston 2007 examined Hg levels in 10 thresher sharks from the Honolulu fish market but the samples were most likely a combination of the two thresher species that predominantly occur in this region, the pelagic thresher, A. pelagicus, and the bigeye thresher, A. superciliosus (W. Walsh In prep.<sup>2</sup>). Watling et al. 1981 examined the Hg levels in 19 shortfin mako from South Africa and Kaneko and Ralston 2007 examined 10 shortfin mako from Hawaii. Both found high levels of Hg in the shark muscle (0.59–5.58  $\mu$ g/g in South Africa and  $0.40-3.10 \,\mu\text{g/g}$  in Hawaii). How these Hg levels compare to those of shortfin mako from California is unknown. The objectives of this study are to determine

the levels of Hg in the common thresher and the shortfin mako from the northeast Pacific Ocean, and investigate their relationships with sex, size and feeding ecology.

#### **METHODS**

# California sample collection and mercury analysis

Common thresher and shortfin mako tissue samples were collected from the following three sources: (1) NOAA National Marine Fisheries Service (NMFS) fishery observers aboard commercial drift gillnet vessels operating off central to southern California during the August 2004–January 2005 fishing season, hereafter referred to as observer surveys; (2) the NMFS juvenile shark survey in the Southern California Bight during July 2004–August 2004, hereafter referred to as NMFS surveys; and (3) the southern California shark fishing tournaments from July 2005–August 2005, hereafter referred to as tournament surveys (fig. 1).

A total of 38 common thresher and 33 shortfin mako sharks were sampled. Observer survey samples (33 common thresher and 19 shortfin mako) and NMFS survey

<sup>&</sup>lt;sup>2</sup>W. Walsh. In prep. Pacific Islands Fisheries Science Center 2570 Dole Street Honolulu, Hawaii 96822.

samples (five common thresher and two shortfin mako) were taken at sea while tournament survey samples (12 shortfin mako) were collected at the docks within six hours of capture. From each shark, white muscle and liver were taken using clean stainless steel instruments. Approximately 100 g of white muscle was collected from a region anterior to the origin of the first dorsal fin. For the liver sample, the posterior tip of the liver was taken. All samples were immediately placed in separate polyethylene bags and were either frozen (observer and NMFS surveys) or kept in an ice cooler (tournament surveys) until transport to the NMFS Southwest Fisheries Science Center in La Jolla, California. Once at NMFS, all samples were stored at  $-20^{\circ}$ C. In the laboratory, subsamples (about 20 g) of each tissue were excised using stainless steel instruments and transferred into pre-cleaned glass vials and kept frozen at  $-20^{\circ}$ C until analysis. For all sharks, body size (fork length, FL) and sex were recorded. Observer and NMFS surveys recorded GPS coordinates for capture locations.

To address whether Hg levels in muscle from different parts of the shark differed, one shortfin mako from the NMFS surveys was sampled from multiple regions. Approximately 100 g of muscle was excised from six different regions as follows: axial muscle as described above, lateral muscle from a region directly forward of the origin of the pectoral fin from each side (left and right), ventral muscle from a region approximately 6 cm posterior from the mouth, caudal muscle from the dorsal precaudal region and vertebral muscle from a region ventral to the vertebrae in the stomach cavity. In addition, two bigeye thresher, A. superciliosus were sampled from the NMFS surveys and one sevengill shark, Notorynchus *cepedianus*, was collected from an area <5 nm offshore of San Diego Bay in April 2003. Data from these opportunistically sampled species are not conclusive but are presented here for reference because data for the species are lacking. The bigeye thresher samples were collected in the same manner as described for the NMFS surveys. The sevengill shark was frozen whole and the samples excised at the laboratory in May 2005 as described above for tournament surveys.

Studies have shown that virtually all of the total mercury, >95%, in teleost fish muscle (Hight and Corcoran 1987; Grieb et al. 1990; Bloom 1992) and elasmobranch fish muscle (Branco et al. 2004) is in the form of methylmercury (MeHg). Consequently, the measurement of total Hg has become widely accepted as the standard for regulatory monitoring programs as an accurate approximation of MeHg (Hight and Corcoran 1987; Bloom 1992). Total Hg analyses were conducted using cold-vapor atomic absorption spectrophotometry according to EPA protocol 7471A at Enviromatrix Analytical Inc. in San Diego, California, using a Leeman Labs PS200 II automated mercury analyzer. Quality control measures included standard reference materials, laboratory blanks, duplicate tissue samples and matrix spikes. All data met the data quality objectives specified in the quality control section of EPA SW-846. The method detection limit was 0.005  $\mu$ g/g. Results are reported as ppm  $\mu$ g/g, by wet weight.

#### Hawaii sample collection and Hg analysis

Muscle tissue samples from 27 shortfin mako were collected by the State of Hawaii's Department of Health from commercially landed sharks from the United Fishing Agency, Ltd at the Honolulu fish market between May 1991 and October 1992. Muscle samples of about 0.4 kg were collected from an area immediately posterior to the head region and subsequently analyzed for MeHg using standard AOAC methods at the Hawaii State Environmental Laboratory (B. Brooks, pers. comm.<sup>3</sup>). The exact quality control methods used are unknown, thus interpretations of these data were limited to general comparisons with the California samples. As these sharks were fully dressed at sea (i.e. gutted, de-headed and de-finned) sexes and accurate body-size measurements were not available, but each carcass was weighed using certified trade scales to the nearest pound. Estimated total mass of each shark was calculated with a formula used by the Pacific Fisheries Information Network based on observer records and port sampling for this species: TM = 1.45 \*DM; where TM = total mass in kg, and DM = dressedmass in kg. Subsequently, TM was converted into body size using the length-weight parameters for the shortfin mako shark presented by Kohler et al. 1996.

## Statistical analysis

All analyses were performed using Systat vers. 11.0 (Systat Soft. Inc. Port Richmond, California). Statistical significance was set a priori at  $\alpha = 0.05$ . We used analysis of covariance (ANCOVA) to test for differences in Hg levels between sexes for the common thresher and shortfin mako. No differences were found for either species (p = 0.15, common thresher; p = 0.79, shortfin mako). Thus, sexes were combined for further analyses. We used ANCOVA to examine the relationships between Hg levels and body size for the common thresher versus shortfin mako from California. Parametric assumptions were evaluated with probability plots and Bartlett's test for homogeneity of variances (Zar 1998).

## RESULTS

A summary of the results from all white muscle samples is provided in Table 1 for all species. The results

<sup>&</sup>lt;sup>3</sup>B. Brooks. Pers. commun. Hawaii Dept. of Health 1250 Punchbowl St. Honolulu, HI 96813.

 TABLE 1

 Hg levels in the white muscle from pelagic sharks from California (CA) and Hawaii (HI).

Species	Common name		Hg (µg/g wet weight)			Fork length (cm)	
		п	Range	Mean	Std dev.	Range	Mean
Isurus oxyrinchus (CA)	Shortfin mako	33	0.15-2.90	1.13	0.89	75-330	164
Alopias vulpinus (CA)	Common thresher	38	0.00-0.70	0.13	0.15	63-241	116
Alopias superciliosus (CA)	Bigeye thresher	2	0.46-0.47	0.46	0	178-182	180
Notorynchus cepedianus (CA)	Sevengill shark	1	0.48			118	_
Isurus oxyrinchus (HI)	Shortfin mako	27	0.40-3.10	1.32	0.68	105-240	185

 TABLE 2

 Results of the ANCOVA between common thresher

 (Alopias vulpinus) and shortfin mako (Isurus oxyrinchus)

 from California.

Source	SS	df	MS	F-ratio	р
Species	0.365	1	0.365	6.734	0.01
Fork Length (FL)	8.115	1	8.115	149.789	0.00
Species * FL	3.009	1	3.009	55.549	0.00
Error	3.63	67	0.054		

from the analyses of 15 liver samples (11 common thresher, two shortfin make and two bigeve thresher) showed no detectable levels of Hg; thus no further liver samples were analyzed. From California, 38 common thresher and 33 shortfin mako were analyzed for Hg in their muscle tissue. Common thresher sizes ranged from 63 to 241 cm FL (tab. 1). Common thresher muscle Hg levels ranged from 0.00 to 0.70  $\mu$ g/g (tab. 1). California shortfin mako sizes ranged from 75 to 330 cm FL. Shortfin mako muscle Hg levels ranged from 0.15 to 2.90 µg/g. The ANCOVA found a significant difference in the relationship between muscle Hg and FL for the common thresher and shortfin mako (tab. 2). Muscle Hg levels increased with size significantly faster in the shortfin mako relative to the common thresher. Pearson's linear regression was used to describe the relationship between muscle Hg levels and body size for both species  $(r^2 = 0.47, p = 0.00)$ , for the common thresher;  $r^2 = 0.87$ , p = 0.00, for the shortfin mako; fig. 2). Multiple muscle samples taken from different regions of a single shortfin mako revealed small differences in Hg levels, mean =  $1.23 \pm 0.15$ , range =  $1.06-1.48 \,\mu$ g/g, with the axial muscle representative of the other samples and being very close to the mean at 1.16  $\mu$ g/g.

The two California bigeye threshers measured 178 and 182 cm FL and their muscle Hg levels were 0.46 to 0.47  $\mu$ g/g, respectively (tab. 1). The one sevengill shark sampled was 118 cm FL and had muscle Hg level of 0.48  $\mu$ g/g. The Hawaii shortfin mako body sizes calculated from mass ranged from 105–240 cm FL. The muscle Hg levels ranged from 0.40–3.10  $\mu$ g/g. Due to uncertainties with quality control, regression analysis was not conducted on this dataset.



Figure 2. Pelagic shark muscle Hg levels for California (CA) shortfin mako (*lsurus oxyrinchus*), common thresher (*Alopias vulpinus*), Hawaii (HI) shortfin mako, bigeye thresher (*Alopias superciliosus*), and sevengill (*Notorynchus cepedianus*) plotted versus fork length (FL) from California (CA) and Hawaii (HI). Pearson's linear regression for the CA common thresher (dotted line; Hg = 0.0025\*FL - 0.1629) and CA shortfin mako shark (solid line; Hg =  $0.012^{*}$ FL - 0.5376).

#### DISCUSSION

This represents the first comprehensive study detailing mercury levels in mako and thresher sharks in the eastern North Pacific Ocean. The data provide insight into the influence of sex, size and foraging ecology on mercury levels in commercially important pelagic sharks. The difference in Hg levels between the common thresher and shortfin mako found in this study is largely explained by differences in their feeding ecology and trophic position. The common thresher from California has been shown to feed most heavily on small schooling fish such as the northern anchovy, *Engraulis mordax*, and Pacific sardine, Sardinops sagax (Preti et al. 2001). The shortfin mako from California feeds on schooling fish like the Pacific saury, Cololabis saira, and cephalopods such as the jumbo squid, Dosidicus gigas, revealing opportunistic foraging habits (Preti et al. 2006<sup>4</sup>; Suk 2008). Even at the largest sizes, the common thresher has been

<sup>&</sup>lt;sup>4</sup>A. Preti, S. E. Smith, D. A. Ramon, and S. Kohin. 2006. Diet differences among 3 species of pelagic sharks inhabiting the California Current. 57th Tuna Conference, May 22–25, 2006, Lake Arrowhead, CA.

shown to have a considerably lower trophic position relative to the shortfin mako; When comparing animals of a similar size, stable isotope analysis of nitrogen revealed that the common thresher is one-fourth of a trophic position lower than the shortfin mako (Suk 2008). This trophic difference likely explains the generally higher Hg levels found in the shortfin mako. In addition, the shortfin mako, being an extremely active predator, might be expected to have a higher daily ration in comparison to common thresher of the same size. A higher rate of food intake could help explain the much faster increase in Hg levels with increasing body size in the shortfin mako compared to the common thresher from the same region (Stillwell 1990).

The shortfin mako sampled from Hawaii between 1991–92 had a mean Hg level of 1.32  $\pm$  0.68 µg/g in the muscle. These findings are comparable to those of Kaneko and Ralston 2007 who found a mean Hg level of  $1.81 \pm 0.42 \,\mu\text{g/g}$  in 10 shortfin mako from Hawaii in 2006. The observed difference, although not significant, may be attributed to the wider size range of shortfin mako sampled in this study (8.03–107.7 kg DM versus 40.8-80.3 kg DM), though temporal and spatial distinctions cannot be ruled out. The shortfin mako sampled from California and Hawaii exhibited roughly similar patterns of linear increase in muscle Hg levels with increasing body size. Unfortunately, the uncertainty in the quality control methods for the Hawaii samples constrains further interpretations. As shark tagging efforts have documented migratory movement of shortfin mako between Hawaiian and Californian waters (Wraith et al. In prep.<sup>5</sup>) and the Hg elimination rate of fish species is known to be very slow (Mason et al. 1995), this result is not surprising. Since food habit studies of shortfin mako from the Hawaii region are lacking, we can only speculate that there might not be large differences in the feeding habits of the shortfin make between the two regions or that any differences are overshadowed by the heavy influence of trophic position on the bioaccumulation of Hg in the pelagic environment. Interestingly, muscle Hg levels for the shortfin mako found in this study are also similar to those for shortfin mako from South Africa (Watling et al. 1981), a geographically separate population, where they have been shown to feed heavily on small elasmobranch prey as opposed to teleosts and cephalopods (Cliff et al. 1990). This further suggests minimal spatial differences in Hg levels in highly migratory pelagic fishes in contrast to more resident fishes, where a single point source of Hg could be more influential (Uryu et al. 2001).

The two bigeye thresher collected had essentially identical levels of Hg in their muscle, about 0.46  $\mu$ g/g. When compared to the other species, the bigeye thresher had similar Hg levels to the adult common thresher but lower Hg levels than the shortfin mako regardless of size. This is likely explained by foraging ecology as well. The bigeye thresher feeds on some of the same prey species as the adult common thresher although its diet also consists largely of barracudinas, (family Paralepididae) and Pacific hake, *Merluccius productus*, rather than small pelagic fish (Preti et al. 2001; Preti et al. 2008; Suk 2008). However, the small sample size prevents drawing further conclusions.

In contrast to Marcovecchio et al. 1991 who found nearly equal Hg levels from both the liver and muscle tissues of the narrownose smooth-hound shark, Mustelus schmitti, none of the liver tissue analyzed in this study had detectable levels of Hg. Studies have shown that in fish, Hg binds to the sulfhydryl group (-SH) in protein, which is most prevalent in muscle tissue (Mason et al. 1995). Thus, predictably, Hg was found to be preferentially stored in the muscle of these pelagic sharks. We chose white muscle for this study because it has been shown to be a good indicator of the exposure level of the whole fish and is also what humans typically consume (Prosi 1979). Storelli et al. 1998 found that muscle tissues sampled from different parts of the blackmouth cat shark, Galeus melastomus, showed little differences in Hg levels. Likewise, we found little differences in Hg levels in white muscle sampled from different parts of the shortfin make, although this was from a single specimen. We conclude that the axial muscle tissue sampled in this study accurately represents the potential source of Hg exposure to humans.

All of the common thresher muscle samples in this study had total Hg levels below the U.S. Food and Drug Administration's established action level of  $1.00 \,\mu\text{g/g}$  for commercial fish. Likewise, all but one shortfin mako ≤150 cm FL had total Hg levels below this level. Since shortfin mako landed by the U.S. West Coast drift gillnet fishery are primarily juveniles (mean ~130 cm FL; PFMC 2003), these fish represent a minimal human health concern. In contrast, all shortfin mako >150 cm FL had muscle Hg levels exceeding  $1.00 \,\mu\text{g/g}$ , with some of the largest individuals having nearly three times this level  $(2.90 \ \mu g/g)$ . Kaneko and Ralston 2007 found that the shortfin mako is one of the few pelagic fish species to have an elemental excess of Hg relative to selenium, (Se). Selenium has been shown to negate the toxicity of MeHg by binding to it (Raymond and Ralston 2004). Because Se is not likely to be adequate to counter the MeHg in the shortfin makos sampled, this suggests that some larger shortfin mako may indeed pose a human health risk.

In this study, we have shown that levels of Hg in the muscle tissue of pelagic sharks reflect differences in the feeding ecology and demonstrated a strong linear rela-

<sup>&</sup>lt;sup>5</sup>J. Wraith. In prep. Southwest Fisheries Science Center 8604 La Jolla Shores Dr. La Jolla, CA 92037.

tionship between body size and Hg levels for both the shortfin mako and common thresher. The bioaccumulation rate of Hg was considerably faster (i.e. at smaller body sizes) in the shortfin mako versus the common thresher and reflects the higher trophic position of the shortfin mako. In addition, these results which indicate levels of Hg relative to body sizes can be potentially used in the assessment of potential human health risk posed by consuming these sharks.

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