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Global versus local causes and health implications of high mercury concentrations in sharks from the east coast of South Africa

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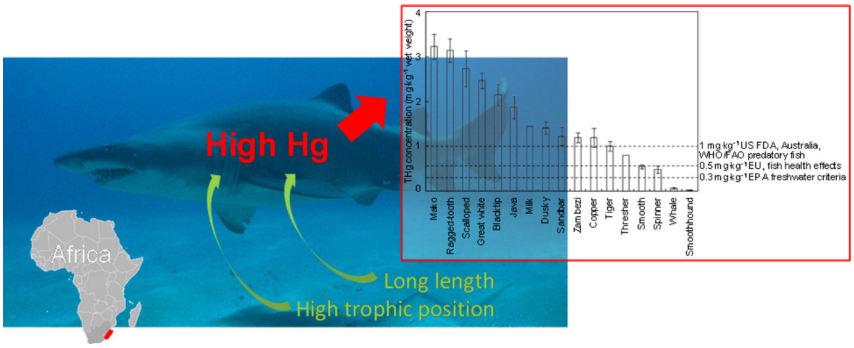
HIGHLIGHTS

- Hg concentrations in 17 shark species from South Africa's east coast were measured.
- Higher values relative to other regions suggested the importance of local emissions.
- Length and trophic position explained most of the mercury variation among species.
- Hg concentrations were above regulatory guidelines for the majority of species.
- Muscle concentrations are of concern for shark and human health.

GRAPHICAL ABSTRACT



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ABSTRACT

Conservation concern regarding the overharvest of global shark populations for meat and fin consumption largely surrounds documented deleterious ecosystem effects, but may be further supported by improved knowledge of possibly high levels in their edible tissues (particularly meat) of the neurotoxin, methylmercury (CH_3Hg). For many regions, however, little data exist on shark tissue Hg concentrations, and reasons for Hg variation within and among species or across regions are poorly understood. We quantified total Hg (THg) in 17 shark species (total $n = 283$) from the east coast of South Africa, a top Hg emitter globally. Concentrations varied from means of around 0.1 mg kg^{-1} dry weight (dw) THg in hardnose smoothhound (*Mustelus mosis*) and whale (*Rhincodon typus*) sharks to means of over 10 mg kg^{-1} dw in shortfin mako (*Isurus oxyrinchus*), scalloped hammerhead (*Sphyrna lewini*), white (*Carcharodon carcharias*) and ragged-tooth (*Carcharias taurus*) sharks. These sharks had higher THg levels than conspecifics sampled from coastal waters of the North Atlantic and North, mid-, and South Pacific, and although sampling year and shark size may play a confounding role, this result suggests the potential importance of elevated local emissions. Values of THg showed strong, species-specific correlations with length, and nearly half the remaining variation was explained by trophic position (using nitrogen stable isotopes, $\delta^{15}N$), whereas measures of foraging habitat (using carbon stable isotopes, $\delta^{13}C$) were not significant. Mercury concentrations were above the regulatory guidelines for fish health effects and safe human

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consumption for 88% and 70% of species, respectively, suggesting on-going cause for concern for shark health, and human consumers of shark meat.

1. Introduction

Global shark populations are declining due to overfishing via bycatch as well as targeted fisheries (Dulvy et al. 2014). Specific fisheries harvest sharks for their meat and for their economically-valuable fins (Clarke et al. 2006). Yet, in addition to deleterious ecosystem effects of eliminating these top predators (Myers et al. 2007), such harvested shark tissues can have substantial levels of the developmental neurotoxin, methylmercury (CH_3Hg) (Cai et al. 2007; Hornung et al. 1993; Kaneko and Ralston 2007; Pethybridge et al. 2010). Regulatory agencies routinely publish fish consumption advisories recommending that people limit or avoid eating sharks and other high trophic level fish with high Hg levels (EPA 2014; http://www.chem.unep.ch/). Nevertheless, shark Hg tissue concentrations are not well-characterized in many regions, and an understanding of the causes of Hg variation within and among shark species, and across regions remains incomplete.

Human activities are currently the main source of primary Hg emissions to the environment, and global emissions and water Hg concentrations in oceans are on the rise (Driscoll et al. 2013; Lamborg et al. 2014). Once released, Hg results not only in local contamination, but also widespread contamination due to long-distance atmospheric transport (Fitzgerald et al. 1998). Upon deposition, microbial activity can result in the formation of CH_3Hg (Blum et al. 2013; Hsu-Kim et al. 2013), which is the form of concern with respect to human and wildlife exposures and toxic effects. Given that oceans are the predominant long-term sink for Hg emissions (Driscoll et al. 2013); significant rates of CH_3Hg production occur in both coastal and open-ocean zones (Blum et al. 2013); and CH_3Hg strongly biomagnifies within ecosystems (Lavoie et al. 2013), it is not surprising that some of the highest tissue levels of $(CH_3)Hg$ reported worldwide have been in marine predators (Driscoll et al. 2013).

Sharks are long-lived, apex predators found throughout the world's oceans (Cortés 1999). It is expected that organisms with such life history traits accumulate high levels of CH_3Hg, given the high uptake and slow elimination rates of CH_3Hg (Boudou and Ribeyre 1997). Indeed, consistent positive correlations of CH_3Hg concentrations with estimated age, or with length or mass as a proxy for age, have been reported for sharks and other aquatic predators (Aubail et al. 2011; Choy et al. 2009; Pethybridge et al. 2010). Given CH_3Hg biomagnification through aquatic food webs, it is expected that sharks feeding at a higher trophic position will have higher CH_3Hg concentrations than those feeding lower in the food web. To date, a small number of studies have reported positive correlations between CH_3Hg concentrations and trophic position, or nitrogen stable isotope ratios ($\delta^{15}N$) as a proxy for trophic position, in sharks (Cai et al. 2007; Cresson et al. 2014; McMeans et al. 2010; Newman et al. 2011; Pethybridge et al. 2012). Additionally, recent studies have found correlations between shark CH_3Hg tissue values and foraging depth or other habitat-use metrics, including carbon stable isotope ratios ($\delta^{13}C$) as a proxy for resource use, e.g., inshore benthic versus offshore pelagic (Choy et al. 2009; Cossa et al. 2012; McMeans et al. 2010).

In addition to biological and ecological factors, local and long-distance Hg inputs likely play a role in Hg variation across regions. Although China by far leads global Hg emissions, South Africa is considered one of the top ten contributors to global emissions, largely due to coal-fired power stations and to a lesser extent to illegal artisanal gold mining (Pacyna et al. 2010; Walters et al. 2011). Concentrations of Hg in precipitation suggest that levels in South Africa are influenced not only by global, but also by regional sources (Gichuki and Mason 2013). However, knowledge of Hg concentrations in sharks (or other species within coastal marine habitats of South Africa) inhabiting waters off southern Africa is poor, with just a single study on shortfin mako sharks (*Isurus oxyrinchus*) from thirty years ago (Watling et al. 1981) and one recent study of smoothhound sharks (*Mustelus mustelus*) (Bosch et al. 2013). As well, little is known regarding Hg concentrations in other marine fish in the area; yet, preliminary work has pointed to high Hg vales in some species from coastal waters (Matooane et al. 2009). If Hg tissue values in South African sharks are of concern with respect to human health, it may be most relevant for countries importing shark products from South Africa. South African shark landings are in fact sent to Australia for the fish and chips trade (although permissible Hg limits apply), and shark fins are exported for the Asian market (da Silva and Bürgener 2007). Although some species in the current study are protected, others are considered major species in the demersal shark trade in South Africa, including copper (*Carcharhinus brachyurus*) and dusky (*Carcharhinus obscurus*) sharks.

In this study, we assess muscle (meat) Hg levels in the largest assemblage of shark species ($n = 17$ species) to date, sampled off the east coast of South Africa. Concentrations of total Hg were analyzed to determine the main factors driving intra-specific and inter-specific Hg level variation, which we hypothesized would include sex, age (using length as a proxy), trophic position (using $\delta^{15}N$ as a proxy), and foraging habitat (using $\delta^{13}C$ as a proxy). Given elevated local Hg emissions, we also compare values measured to that reported in the same species worldwide to test the hypothesis that sharks from South African show elevated muscle Hg levels. We finally discuss implications of our results for both shark and human health.

2. Materials and methods

2.1. Sampling

Full sampling details are provided elsewhere (Hussey et al. 2014). Briefly, sharks ($n = 283$) of 17 different species (Table 1) were sampled from captures in beach protection nets along the east coast of South Africa (KwaZulu-Natal) between 2005 and 2010 (Davidson et al. 2011), except for beach stranded whale sharks (*Rhincodon typus*) ($n = 3$) and a fishery by-catch collection of hardnose smoothhound sharks (*Mustelus mosis*) ($n = 5$). Sex and precaudal length (PCL; hereafter referred to as length) were recorded. White muscle tissue (5 g) was collected anterior to the first dorsal fin in the center of the muscle block and stored at −20 °C.

2.2. Total mercury (THg) analysis

We used THg as a proxy for CH_3Hg, since CH_3Hg comprises more than 90% of THg in fish, including in shark muscle tissue (Pethybridge et al. 2010). We determined dry weight (dw) THg concentrations in freeze-dried, homogenized muscle tissues using a Direct Mercury Analyzer (DMA-80; Milestone Inc., Shelton, CT, USA) at the Canadian Association for Laboratory Accreditation- (CALA-) accredited Great Lakes Institute for Environmental Research (University of Windsor, Windsor, ON, Canada). Quality control procedures included analysis of blanks (20% of runs), in-house biological tissue reference samples, duplicate shark sample analysis, and National Research Council of Canada certified standards (DORM-3, DOLT-4). Concentrations of THg in certified standards ranged from 92 to 102% and 95–113%, respectively. The detection limit, defined as three times the blank standard deviation, was 0.005 mg kg^{-1} dw based on a 0.1 g sample weight.