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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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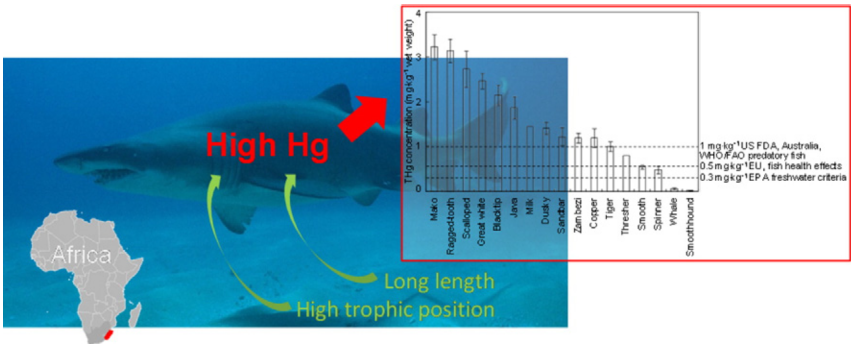
HIGHLIGHTS

- 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 by-catch 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.

Table 1

Mean precaudal length (PCL) (range), δ^{15}N, δ^{13}C, stomach content-based trophic position (TP)[a], forage depth[a,b] and dry weight (dw) total mercury (THg) (±SE) of shark species sampled from the east coast of South Africa from 2005 to 2010.

Species	*n*	PCL (cm)	δ^{15}N (‰)	δ^{13}C (‰)	TP	Depth (m)	THg (mg kg^{-1} dw)
Carcharhiniformes Carcharhinidae							
Carcharhinus amboinensis (Java/Pigeye)	9	131 (106–152)	15.4 ± 0.1	−14.5 ± 0.1	4.3	75	7.79 ± 1.05
Carcharhinus brachyurus (Copper)[c]	5	197 (170–216)	14.1 ± 0.1	−15.5 ± 0.1	4.2	180	4.98 ± 0.88
Carcharhinus brevipinna (Spinner)	19	148 (57–193)	13.5 ± 0.1	−15.5 ± 0.1	4.2	500	2.33 ± 0.41
Carcharhinus leucas (Zambezi/Bull)	11	165 (79–208)	14.6 ± 0.2	−14.9 ± 0.2	4.3	75 (15)	5.32 ± 0.32
Carcharhinus limbatus (Blacktip)	32	153 (113–186)	14.4 ± 0.1	−15.2 ± 0.1	4.2	32 (15)	8.95 ± 0.95
Carcharhinus obscurus (Dusky)	64	164 (70–280)	13.7 ± 0.1	−15.6 ± 0.1	4.2	200 (300)	5.86 ± 0.57
Carcharhinus plumbeus (Sandbar)[c]	6	124 (93–139)	15.0 ± 0.3	−15.2 ± 0.2	4.1	140 (43)	5.10 ± 0.83
Galeocerdo cuvier (Tiger)	18	195 (135–239)	13.0 ± 0.1	−16.1 ± 0.2	4.1	185	4.17 ± 0.46
Rhizoprionodon acutus (Milk/Sharpnose)[c]	2	71–75	13.7–13.9	−16.0 to −15.5	4.1	100	1.89–10.16
Carcharhiniformes Sphyrnidae							
Sphyrna lewini (Scalloped hammerhead)	34	138 (38–223)	14.8 ± 0.1	−15.5 ± 0.1	4.1	256 (13)	12.45 ± 1.84
Sphyrna zygaena (Smooth hammerhead)	15	100 (70–116)	14.7 ± 0.1	−15.8 ± 0.1	4.2	100 (10)	2.27 ± 0.18
Carcharhiniformes Triakidae							
Mustelus mosis (Hardnose smoothhound)[c]	5	100 (99–103)	10.1 ± 0.2	−17.5 ± 0.1	3.8	145	0.09 ± 0.01
Lamniformes Alopiidae							
Alopias vulpinus (Common thresher)[c]	1	200	13.8	−15.9	4.2	225 (100)	3.33
Lamniformes Lamnidae							
Carcharodon carcharias (White)	33	217 (124–363)	15.3 ± 0.2	−14.8 ± 0.1	4.5	640 (125)	10.26 ± 0.72
Isurus oxyrinchus (Shortfin mako)[c]	4	203 (161–220)	15.0 ± 0.3	−15.3 ± 0.3	4.3	370 (125)	13.43 ± 1.18
Lamniformes Odontaspididae							
Carcharias taurus (Ragged-tooth/Sand tiger)	30	191 (149–209)	15.6 ± 0.1	−14.3 ± 0.1	4.4	96 (20)	13.08 ± 1.09
Orectolobiformes Rhincodontidae							
Rhincodon typus (Whale)[c]	3	684 (660–710)	9.9 ± 0.3	−17.3 ± 0.3	3.5	350 (35)	0.26 ± 0.08

[a] Literature-based trophic positions and forage depths.
[b] Midpoint of depth range (parenthetical lists midpoint of usual forage depth range, if available).
[c] Species not included in statistical analyses due to low sample size.

2.3. Stable isotope analysis

Stable isotope analyses and quality control procedures for these shark samples have been described previously (Hussey et al. 2014). In short, we analyzed lipid-removed, freeze dried homogenate samples for δ^{15}N and δ^{13}C ratios using a continuous flow isotope ratio mass spectrometer (Delta V Advantage; Thermo Scientific, Waltham, MA, USA). The analytical precision for δ^{15}N was 0.16‰ and 0.23‰, respectively, and for δ^{13}C was 0.07‰ for both NIST 8414 and internal laboratory fish muscle standards ($n = 111$) Accuracy was indicated by values that were within 0.01‰ and 0.07‰ for δ^{15}N and δ^{13}C, respectively, for the certified NIST 8542 and NIST 8542 ($n = 3$) standards.

2.4. Data analysis

For global comparisons, we performed a literature search using the terms “mercury” and the scientific or common name(s) for each shark species in this study to compile species- and geographic-specific Hg levels. We only considered studies with sample sizes of at least three for a given species to avoid incorrect conclusions regarding global variation as a result of sample size bias. For comparison of shark THg concentrations to other studies reporting wet weight (ww) THg values and to consumption guidelines, we converted dw THg levels to ww THg levels based on an average water content of 76 ± 3% for a subset of $n = 91$ of the shark muscle tissue samples from various species (N. E. Hussey, unpublished data). That is, ww values were estimated as 0.24 × THg (dw).

Statistical analyses of biological and ecological factors influencing South African shark THg levels were done using Statistica version 12 (Statsoft, Tulsa, OK, USA), and *p*-values <0.05 were considered statistically significant. Log-transformed THg concentrations (log[THg]) were used to better approximate normal distribution. Length showed minor deviation from normality, which was not improved by log-transformation, whereas δ^{15}N and δ^{13}C values were normally distributed. We used general linear models (GLMs) to test the influence of biological variables (sex, PCL) on species differences in log[THg]. We did not include species with low sample sizes ($n < 9$) in the statistical analyses to avoid spurious relationships. We removed 12 outliers (studentized residuals >2 SD) and examined probability plots to confirm normality and homoscedasticity of the residuals. We corrected for the effects of significant biological covariates to subsequently investigate inter-species variation in log[THg] using simple linear regressions between species means of log[THg] and δ^{15}N and δ^{13}C. As stable isotopes can vary spatially and with depth in oceans, and the studied sharks can be highly mobile, we similarly investigated relationships of species mean log[THg] with literature-derived depth of occurrence and trophic position. Species midpoint of overall depth range (or “usual” range, if listed) was determined from the depth ranges on FishBase (fishbase.org). Species mean trophic positions were based on diet composition from stomach contents (Cortés 1999). We also examined intra-species variation in log[THg] using simple linear regressions between biological covariate-corrected individual log[THg] and δ^{15}N and δ^{13}C.

3. Results and discussion

3.1. South African shark Hg levels and global comparisons

Detectable concentrations of THg were measured in all shark muscle samples and varied by more than two orders of magnitude from means of around 0.1 mg kg^{-1} dw in smoothhound and whale sharks to more than 10 mg kg^{-1} dw in mako, scalloped hammerhead (*Sphyrna lewini*), white (*Carcharodon carcharias*) and ragged-tooth (*Carcharias taurus*) sharks (Table 1).

Published values for THg in sharks sampled from other regions worldwide were available for 7 of the 17 species examined in this study (Table 2). Incomplete geographic and species coverage, variation in shark lengths among studies, as well as temporal variation in sampling periods, should be noted. Thus, conclusions regarding these global comparisons should be interpreted with caution until more in-depth comparisons can be made. That said, THg concentrations were higher in South African sharks relative to sharks of the same species from coastal waters of the North Atlantic (US), as well as the North (Japan, Mexico, US), mid- (Hawaii) and South Pacific (Australia, Papua New Guinea, Chile), but not relative to sharks from the Mediterranean Sea. The Mediterranean Sea is considered to have anomalously high Hg levels in upper trophic level biota, likely as a consequence of lower growth rates and feeding in zones of greater CH_3Hg production, and thus greater Hg bioavailability (Cossa et al. 2012).

From a global perspective, it may seem surprising that South African sharks showed consistently elevated THg levels relative to sharks sampled in coastal waters of the North Atlantic and North Pacific, given the strong evidence for higher emissions and atmospheric Hg concentrations over the oceans in the Northern Hemisphere versus the Southern Hemisphere (Sprovieri et al. 2010). Sharks in this study were sampled more recently than sharks from some of the other studies, while levels of Hg have steadily increased in the world's oceans since the earliest studies (~1980) (Amos et al. 2013). In agreement, we found 50% higher THg concentrations in mako sharks collected from 2005 to 2010 (this study, sharks of length 161–220 cm) compared to conspecifics of similar size ranges sampled in the same waters in 1980 (Watling et al. 1981, sharks of length 110–260 cm). Some fraction of the geographic differences observed could thus be attributed to temporal variation. Yet, other studies overlapped our sampling period, and South African sharks still showed THg values that were elevated by 28 to 333% relative to conspecifics (tiger, scalloped hammerhead, smooth hammerhead, white and shortfin mako) from other regions. Length could also be a confounding factor in these comparisons, although the South African sharks sampled were not consistently larger than those sampled in other studies. Another explanation may be elevated emissions from local South African sources (Pacyna et al. 2010). As well, high proportions of CH_3Hg relative to THg have been reported in South African freshwater samples (60%) versus US waters (cf. 1 to 5%) (Walters et al. 2011), although their links to marine CH_3Hg concentrations have not been determined. Also, a lack of characterization of oceanic and food web levels of Hg and CH_3Hg within east coast waters of South Africa precludes definitive conclusions regarding the role of local anthropogenic inputs versus other region-specific biogeochemical or ecological factors in contributing to elevated THg levels in these marine predators.

3.2. Biological factors influencing THg levels

Results of the initial GLM ($F_{30,210} = 65.6$, $p < 0.001$) showed that length, and species and their interaction influenced THg concentrations ($p < 0.001$), but sex and other interaction terms were not significant. A lack of influence of sex is consistent with most other studies on Hg in sharks (de Pinho et al. 2002; Endo et al. 2008; Suk et al. 2009) and suggests that maternal offloading of contaminants to young is not a strong driver of muscle Hg levels in female sharks (Le Bourg et al. 2014). Moreover, this would also suggest that males and females of all species analyzed here do not have substantial differences in feeding habits or habitats, at least those that could influence THg values. It should be noted that for many species (white, Java (*Carcharhinus amboinensis*) and smooth hammerheads (*Sphyrna zygaena*)), only juveniles or

Table 2
Wet weight (ww) muscle total mercury (THg) levels (± SE or range) in sharks sampled worldwide.

Species[a]	Time Period	*n*	PCL (cm)	THg (mg kg^{-1} ww)[b]	Reference
C. leucas (Zambezi/Bull)					
Indian Ocean coast, South Africa	2005–10	11	165 ± 6	1.20 ± 0.11	This study
Atlantic Ocean coast, USA	1992–95	53	75.56 (55.2–107.5)	0.77 ± 0.04	Adams and McMichael 1999
C. limbatus (Blacktip)					
Indian Ocean coast, South Africa	2005–10	32	153 ± 4	2.15 ± 0.23	This study
North coast, Australia	1980	234	106 (68–158)	1.05 (0.26–3.20)	Lyle 1984
Atlantic Ocean coast, USA	1992–95	21	93.96 (51.3–162.3)	0.77 ± 0.15	Adams and McMichael 1999
Solomon Sea coast, Papua New Guinea	1977–87	463	11–150	0.22 (0.01–3.27)	Powell and Powell 2001
G. cuvier (Tiger)					
Indian Ocean coast, South Africa	2005–10	18	195 ± 8	1.00 ± 0.11	This study
Pacific Ocean coast, Japan	2007	42	207 ± 50	0.78 ± 0.04	Endo et al. 2008
S. lewini (Scalloped hammerhead)					
Indian Ocean coast, South Africa	2005–10	34	138 ± 9	2.73 ± 0.40	This study
North coast, Australia	1980	82	116 (73–205)	1.15 (0.29–4.92)	Lyle 1984
Pacific Ocean coast, Mexico	2009–10	12	65–83	0.82 ± 0.10	Hurtado-Banda et al. 2012
Solomon Sea coast, Papua New Guinea	1977–87	50	32–300	0.44 (0.02–3.13)	Powell and Powell 2001
S. zygaena (Smooth hammerhead)					
Mediterranean Sea	2001	4	297 ± 64	12.15 ± 4.60	Storelli et al. 2003
Indian Ocean coast, South Africa	2005–10	15	100 ± 3	0.54 ± 0.04	This study
Pacific Ocean coast, Mexico	2006–07	37	~60–184 (total length)	0.16 (0.01–1.93)	Escobar-Sánchez et al. 2010
C. carcharias (White)					
Indian Ocean coast, South Africa	2005–10	33	217 ± 9	2.46 ± 0.17	This study
Pacific Ocean coast, USA	2006–12	30	~100–250	1.21 ± 0.20	Lyons et al. 2013
I. oxyrinchus (Shortfin mako)					
Indian Ocean coast, South Africa	2005–10	4	203 ± 14	3.22 ± 0.28	This study
Indian Ocean coast, South Africa	1980	19	~75–225	2.11 ± 0.28	Watling et al. 1981
Atlantic Ocean coast, USA	2003–08	51	182 ± 3.58	1.83 ± 0.17	Burger and Gochfeld 2011
Pacific Ocean coast, Hawaii	2006	10	NA	1.81 ± 0.13	Kaneko and Ralston 2007
Pacific Ocean coast, Hawaii	1991–92	27	185 (105–240)	1.32 ± 0.13	Suk et al. 2009
Pacific Ocean coast, USA	2004–05	33	164 (75–330)	1.13 ± 0.15	Suk et al. 2009
Pacific Ocean coast, USA	1996–2012	31	~50–350	0.68 ± 0.08	Lyons et al. 2013
Pacific Ocean coast, Mexico	2008	20	89–249 (total length)	0.39 (0.09–0.49)	Veléz-Alavez et al. 2013
Pacific Ocean coast, Chile	2011	69	< and >285	0.006 ± 0.001	Lopez et al. 2013

[a] Only species for which Hg levels have been reported previously are listed here.

[b] Wet weight values in this study were estimated as 0.24 × THg (dry weight).

subadults were analyzed, which could reduce the likelihood of observing sex-based differences. Regardless, since sex had no influence on this dataset, sex terms were removed, and the analysis was re-analyzed with similar results ($F_{19,232} = 109.7$, $p < 0.001$).

The reduced model including length, species and length × species accounted for 90% of the total variation in log[THg]. Concentrations of THg increased with shark length, but the slope of the log[THg]-length relationship was species-specific ($p < 0.001$) (Fig. 1). The average increase was 0.005 log[THg] (mg kg^{-1} dw) per cm increase in length, but slopes for blacktip (*Carcharhinus limbatus*), Java and ragged-tooth sharks were twice the average (0.009–0.013 log[THg] per cm), whereas slopes for white, smooth hammerhead and tiger (*Galeocerdo cuvier*) were relatively lower (<0.003 log[THg] per cm). Blacktip, Java and ragged-tooth sharks are relatively smaller (120–230cm length at maturity; fishbase.org), whereas white, smooth hammerhead and tiger sharks are larger (≥250 cm), suggesting that inter-specific differences in THg-length relationships may be related to species-specific growth and/or food intake rates (Suk et al. 2009; Wetherbee et al. 2012). However, the inclusion of different age ranges among species may play a role; for instance, the smooth hammerheads were all juveniles and thus limited size variation may explain the low (and non-significant) slope for this species. In addition, variation in sample size among species may also have had an influence.

Mercury concentrations are typically correlated with size (weight, length) and age in fish, a pattern attributed to the paradigm of Hg bioaccumulation with age (Driscoll et al. 2013). However, ages were not available for these samples, and accurate and precise shark aging remains an active area of research (Goldman et al. 2012). Regardless, the relationship of either length or age to Hg concentration may be confounded by other factors, as larger (older) sharks tend to feed on prey of a higher trophic position, or have different habitats, habitat ranges or movements, relative to smaller (younger) sharks (Cortés 1999; Speed et al. 2010). In other words, changes in trophic position or foraging habitat with increasing body size, and not only bioaccumulation, could result in an observed pattern of increasing THg concentrations with length. Although we did not find a significant correlation between length and δ^{15}N or δ^{13}C in the overall dataset ($r^2 < 0.003$, $p > 0.4$), length was correlated with both δ^{15}N and δ^{13}C for dusky and ragged-tooth sharks (r^2: 0.12–0.37, p: <0.001–0.007), with δ^{15}N for white and scalloped hammerhead sharks (r^2: 0.28–0.61, p: <0.001–0.01), and with δ^{13}C for spinner (*Carcharhinus brevipinna*) and tiger sharks (r^2: 0.20–0.35, p: 0.01–0.04) (for further discussion of these biological–ecological relationships, see Hussey et al. 2011, 2012). In all instances, however, length explained more of the intra-specific variation in THg concentrations than did either δ^{15}N or δ^{13}C. Length explained more than 80% of the intra-specific variation in THg in blacktip, dusky, scalloped hammerhead, and spinner sharks and more than 50% of the variation in white, Java, ragged-tooth and Zambezi (*Carcharhinus leucas*) sharks, though it accounted for little to none of the variation in tiger and smooth hammerhead sharks. Given that biological and ecological factors were correlated (i.e., size and δ^{15}N or δ^{13}C) for some species, we then determined the ecological factors influencing THg in sharks above and beyond the variation explained by biological factors (i.e. length alone). To do so, we used a separate-slopes model to calculate individual length-corrected logTHg levels at the lower (116.3 cm) and upper quartile (196.7 cm) levels of length and compared inter-specific variation in THg levels in “small” (or young) and “large” (or old) sharks separately.

3.3. Ecological factors influencing THg levels

We were initially interested in using δ^{15}N, δ^{13}C, literature derived-trophic position and/or foraging depth as variables explaining ecological influences on size-corrected species mean THg levels. However, we had to reduce the variables assessed as species-mean δ^{15}N, δ^{13}C, and literature-derived trophic position were correlated. The significant positive correlation of δ^{15}N and stomach content-derived trophic position ($r^2 = 0.46$, $p = 0.03$) is not surprising and justifies the use of δ^{15}N as a reasonable proxy for trophic position in sharks. The significant positive correlation of δ^{15}N and δ^{13}C ($r^2 = 0.67$, $p = 0.004$) is explained by the fact that δ^{13}C also increases with trophic position in a food web, providing strong evidence that the sampled sharks are part of the same food web (N. Hussey, unpublished data). This finding also supports our study of Hg food web dynamics (e.g., biomagnification) using this dataset. Because of correlations among these independent variables, we focused on δ^{15}N, which explained more of the variance in THg than either δ^{13}C or literature-derived trophic position, and also on forage depth. Size-corrected species mean log[THg] increased linearly with δ^{15}N for both large ($r^2 = 0.44$, $p = 0.04$; Fig. 2) and small sharks ($r^2 = 0.43$, $p = 0.04$; plot not shown), indicating that close to half of the residual variance in THg among species was accounted for by differences in δ^{15}N or trophic position. These results show that although size is the dominant factor driving THg levels in these sharks, feeding habits

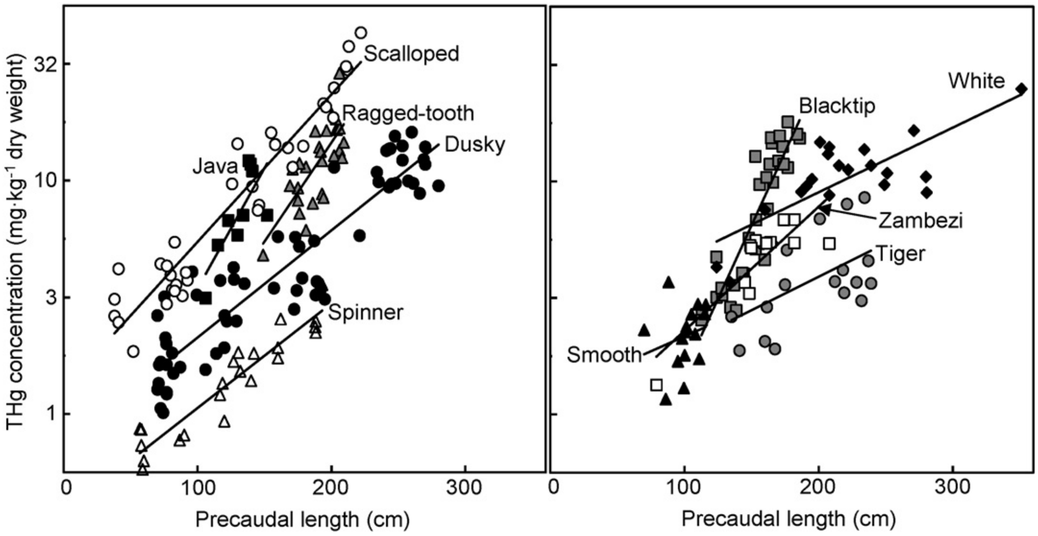

Fig. 1. Muscle total mercury (THg) levels (log-scale) versus precaudal length in 10 shark species collected from the east coast of South Africa from 2005 to 2010. Symbols represent blacktip ($r^2 = 0.81$, $p < 0.001$), dusky ($r^2 = 0.85$, $p < 0.001$), white ($r^2 = 0.60$, $p < 0.001$), Java ($r^2 = 0.59$, $p = 0.02$), ragged-tooth ($r^2 = 0.54$, $p < 0.001$), scalloped ($r^2 = 0.91$, $p < 0.001$), smooth ($r^2 = 0.05$, $p = 0.42$), spinner ($r^2 = 0.89$, $p < 0.001$), tiger ($r^2 = 0.27$, $p = 0.03$) and Zambezi ($r^2 = 0.73$, $p < 0.001$) sharks. Lines represent regression lines for each species. Shark species of $n < 9$ were not included, however, for interest's sake the correlation coefficient for shortfin mako sharks (4 samples, but of highest mean THg) was $r^2 = 0.53$. Data divided into two panels for visibility.

of the sharks are also a significant determinant. In one of the only other studies examining several shark species simultaneously, $\delta^{15}N$ better accounted for THg variation than did size in an assemblage of deepwater chondrichthyans from southeastern Australia (Pethybridge et al. 2012). Due to low sample sizes for most species, however, those authors only examined overall THg-length relationships, which are likely weaker than species-specific relationships, as observed in our data.

Generally speaking, size-corrected THg concentrations agreed with known diets of the individual species. For example, relatively higher THg levels in ragged-tooth, Java, and white sharks agree with feeding mainly on other chondrichthyans, in addition to teleost fish, whereas tiger, smooth hammerhead and spinner sharks with the lowest THg levels feed mainly on teleosts and/or reptiles or cephalopods (Cortés 1999; Hussey et al. 2014). Yet, the size-corrected THg levels in Java, blacktip and scalloped hammerhead sharks were higher than predicted from the THg-$\delta^{15}N$ regression (Fig. 2), suggesting the influence of additional factors. Size-corrected species mean log[THg] was not correlated with overall or usual forage depth (large sharks: $r^2 < 0.1$, $p > 0.38$; small sharks: $r^2 < 0.29$, $p > 0.11$), even though such a relationship has been found in fish assemblages from other regions (Choy et al. 2009; Cossa et al. 2012). Scalloped hammerhead sharks are known to forage at greater depths than other species examined here, except for white sharks (Table 1), which may explain their higher THg levels. In contrast, blacktip and Java sharks inhabit shallow nearshore waters, suggesting instead that elevated THg levels in these sharks, beyond that explained by trophic position, could be a consequence of proximity of foraging habitat to local South African Hg source regions. For example, a Hg processing plant existed until recently in KwaZulu-Natal and reportedly discharged Hg into nearby waterways in 1990 (Papu-Zamxaka et al. 2010). Nonetheless, spinner and Zambezi sharks also inhabit nearshore waters (Table 1) and fall close to the THg-$\delta^{15}N$ regression line. On an intra- specific basis, size-corrected THg concentrations were significantly and weakly positively correlated with $\delta^{15}N$ only for blacktip ($r^2 = 0.15$, $p = 0.03$) and white sharks ($r^2 = 0.25$, $p = 0.02$), and with $\delta^{13}C$ only for blacktip sharks ($r^2 = 0.33$, $p < 0.001$; $\delta^{15}N$ and $\delta^{13}C$ were not correlated for blacktip). Thus, in terms of ecological factors explaining THg concentrations, instead of a single forage depth measure per species, more fine-scale habitat parameters that ideally consider intra- as well as inter-specific variation may be required, such as daily, seasonal and interannual movement data (Hussey et al. 2009).

The slope of the linear regression of log[THg] or log[CH_3Hg] with $\delta^{15}N$ is generally used to assess THg or CH_3Hg biomagnification through a food web, as $\delta^{15}N$ is considered a proxy for trophic position. A recent worldwide meta-analysis found a mean trophic magnification slope (TMS) of 0.16 in aquatic food webs, but with much variation among systems that the authors concluded remains unexplained (Lavoie et al. 2013). We found a TMS nearly twice as high (0.29) as the global mean in our dataset, which consisted of upper trophic level sharks. For such species, diet-tissue discrimination factors ($\Delta^{15}N$) have been shown to be lower than for teleost fish (Hussey et al. 2010). Moreover, variation in $\Delta^{15}N$ has been demonstrated between species and taxa, and with diet (Caut et al. 2009; Overmyer et al. 2008). Thus, results of this and other studies suggest that the direct use of $\delta^{15}N$, or likewise the use of $\delta^{15}N$-based trophic position estimates employing a single $\Delta^{15}N$ value, may not be the most accurate or precise methods to use when calculating Hg biomagnification through a food web. Hussey et al. (2014) have proposed a new method of calculating trophic position that accounts for $\Delta^{15}N$ variation, whereby $\Delta^{15}N$ narrows with increasing trophic position. We suggest that biomagnification of Hg, as well as other contaminants that biomagnify, should be reassessed using these more appropriate trophic position estimates.

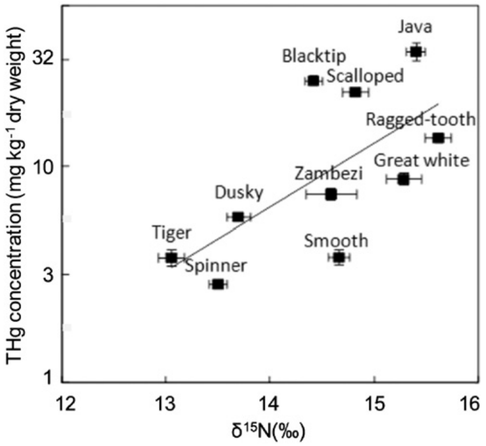

Fig. 2. Mean muscle total mercury (THg) levels (log-scale) versus $\delta^{15}N$ (±SE) in 10 shark species collected from the east coast of South Africa from 2005 to 2010. Mean THg levels are size-corrected to "large" (or old) sharks, i.e. to the upper quartile precaudal length (196.7 cm) using species-specific log[THg]-length relationships. Black line represents the regression line ($r^2 = 0.44$, $p = 0.04$). Shark species of $n < 9$ were not included.

3.4. Implications for shark and human health

The presence of CH_3Hg in human dietary items is of concern due to its neurotoxic effects at low levels, particularly on embryonic and fetal development (Driscoll et al. 2013). The main source of CH_3Hg exposure to humans is fish consumption (Mergler et al. 2007). Regulatory guidelines for the protection of human health generally range from 0.3 to 1 mg kg^{-1} ww as the maximum permissible levels of Hg or CH_3Hg in fish tissues (http://www.chem.unep.ch/). For 12 of the 17 South African shark species, mean THg concentrations were above 1 mg kg^{-1} ww guidelines, and for all but smoothhound and whale sharks, mean THg concentrations were above 0.3 mg kg^{-1} ww guidelines (Fig. 3). Data on Hg concentrations in shark fins is limited, but concentrations may be around an order of magnitude lower than in muscle tissues (Escobar-Sánchez et al. 2010), which would suggest that even fin Hg levels of mako and ragged-tooth sharks from South Africa could be above the 0.3 mg kg^{-1} ww guideline. More data on shark fin Hg levels and ratios to muscle Hg levels are needed, as well as information on CH_3Hg:Hg ratios in shark fins. Although a high molar ratio of selenium (Se) to Hg is thought to potentially offer protective effects against Hg toxicity, the Se:Hg molar ratio in shark muscle appears to be low relative to teleost fish, indicating a health risk or at least very limited health benefit to consuming elasmobranch fish (Burger and Gochfeld 2011; Kaneko and Ralston 2007). It should be noted that some of these sharks are fully (white sharks: Government Gazette-Republic of South Africa. 1998) or partially (whale and ragged-tooth sharks: Government Gazette-Republic of South Africa. 2005; hammerheads: C da Silva, South African Department of Agriculture, Forestry and Fisheries, personal communication) protected in South Africa, and thus consumption of these species may be absent or at least limited. In addition, of the 17 shark species tested, estimated average annual landings exceed 10 t in the case of only three; dusky (range 11–100 t), copper *Carcharhinus brachyurus* (101–200 t) and shortfin mako (301–700 t), and that, in general, sharks weighing between 1.5 and 12 kg are preferred because of lower (size-related) levels of Hg and Cd (da Silva et al., 2015; da Silva and Bürgener 2007). Filets of shortfin mako, however, are valuable regardless of size (da Silva et al., 2015), which is a concern, given the high levels of THg.

Less frequently considered are the toxicological implications of such high Hg levels for the fish themselves. Sub-lethal health effects, including cell and tissue damage and lowered reproduction, occur in freshwater fish at muscle Hg concentrations in the range of 0.5–1.2 mg kg^{-1} ww (Sandheinrich and Wiener 2011). Estimated mean muscle THg levels for all species of shark in this study met or exceeded 0.5 mg kg^{-1} ww, with the exception of smoothhound and whale sharks (Fig. 3). Nonetheless, differences in Hg sensitivity should be expected between sharks and teleosts, as well as among shark species, as has been observed for other vertebrate classes (Sandheinrich and Wiener 2011).

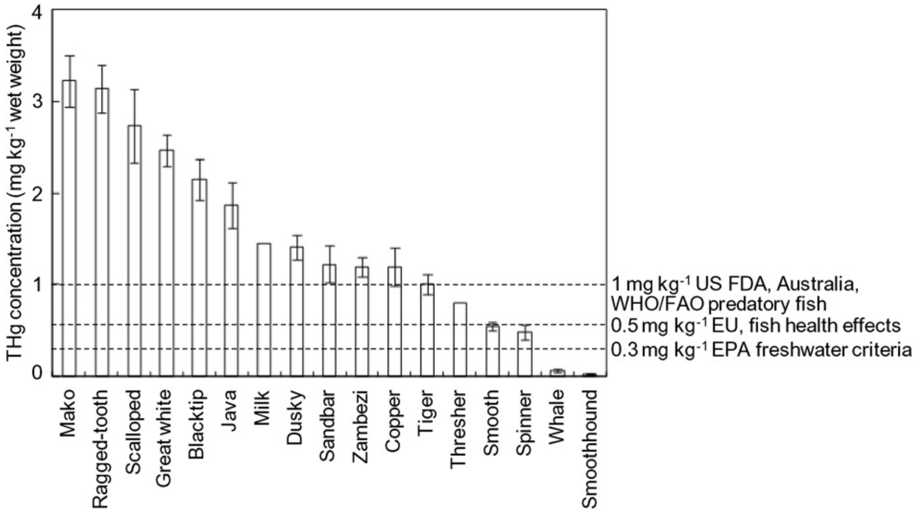

Fig. 3. Mean muscle total mercury (THg) concentrations (± SE) in 17 shark species collected from the east coast of South Africa from 2005 to 2010 relative to regulatory guidelines for human and fish health (http://www.chem.unep.ch/and Sandheinrich and Wiener 2011). Wet weight THg values in this study were estimated as 0.24 × THg (dry weight).

A global treaty, the Minamata Convention on Mercury (http://www.mercuryconvention.org/), was recently (2013) established through UNEP to protect humans and other biota from deleterious effects of Hg through reduced anthropogenic Hg emissions to the environment. It has been predicted that substantial reductions in primary Hg emissions would be required just to stop Hg from increasing beyond current levels in the world's oceans due to the persistence of Hg and to on-going accumulation of burdens from previous emissions (Amos et al. 2013). Thus, substantial near-term reductions in marine predator Hg levels (such as the high levels found in South African sharks in this study) as a consequence of the Convention are unlikely, suggesting an on-going human health concern in the near future.

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