SETAC PRESS

Environmental Toxicology and Chemistry, Vol. 34, No. 9, pp. 2051–2060, 2015

Printed in the USA

COMPARATIVE ORGANOCHLORINE ACCUMULATION IN TWO ECOLOGICALLY SIMILAR SHARK SPECIES (*CARCHARODON CARCHARIAS* AND *CARCHARHINUS OBSCURUS*) WITH DIVERGENT UPTAKE BASED ON DIFFERENT LIFE HISTORY

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(Submitted 6 November 2014; Returned for Revision 10 December 2014; Accepted 15 April 2015)

Abstract: Trophic position and body mass are traits commonly used to predict organochlorine burdens. Sharks, however, have a variety of feeding and life history strategies and metabolize lipid uniquely. Because of this diversity, and the lipid-association of organochlorines, the dynamics of organochlorine accumulation in sharks may be predicted ineffectively by stable isotope-derived trophic position and body mass, as is typical for other taxa. The present study compared ontogenetic organochlorine profiles in the dusky shark (*Carcharhinus obscurus*) and white shark (*Carcharodon carcharias*), which differ in metabolic thermoregulation and trophic position throughout their ontogeny. Although greater organochlorine concentrations were observed in the larger bodied and higher trophic position white shark (e.g., p,p′-dichlorodiphenyldichloroethylene: 20.2 ± 2.7 ng/g vs 9.3 ± 2.2 ng/g in the dusky shark), slopes of growth-dilution corrected concentrations with age were equal to those of the dusky shark. Similar ontogenetic trophic position increases in both species, less frequent white shark seal predation than previously assumed, or inaccurate species-specific growth parameters are possible explanations. Inshore habitat use (indicated by $\delta^{13}C$ values) and mass were important predictors in white and dusky sharks, respectively, of both overall compound profiles and select organochlorine concentrations. The present study clarified understanding of trophic position and body mass as reliable predictors of interspecific organochlorine accumulation in sharks, whereas regional endothermy and diet shifting were shown to have less impact on overall rates of accumulation. *Environ Toxicol Chem* 2015;34:2051–2060. © 2015 SETAC

Keywords: Bioaccumulation Organochlorine Stable isotopes Dusky shark White shark

INTRODUCTION

The global distribution and persistence of organochlorines such as polychlorinated biphenyls (PCBs) and organochlorine pesticides make them some of the most significant contaminants affecting the health of humans and the environment. Accumulation of organochlorines in biota is an outcome of organochlorine hydrophobicity and resistance to breakdown and excretion, which drive diet to be the most important route of exposure for the more hydrophobic organochlorines [1]. Consequently, organochlorine concentrations increase with increasing trophic levels in food webs, a process termed biomagnification. Large-bodied, long-lived, and high–trophic level animals such as seals [2–4], cetaceans [5–7], and sharks [1,8,9], experience the highest organochlorine exposure. Potential outcomes of organochlorine exposure on the sustainability of shark populations is an imperative consideration, because they are subject to many other compounding stressors contributing to their global decline [10,11]. Such stressors include selective capture via commercial fishing operations and shark fin trade-driven incentives [12], habitat degradation [13], and the overarching stressor of climate change [14].

Predicting organochlorine concentrations in sharks is a challenge for several reasons. Sharks are unique among vertebrates in their lipid metabolism and use of their lipid-rich livers for buoyancy control [15]. Shark species also vary greatly in ecological and life history characteristics that would be expected to affect contaminant accumulation, such as growth rates, feeding rates, ontogenetic diet shifts, and metabolic capacity [16,17]. Because of this variation in physiological and ecological processes governing organochlorine accumulation in sharks, thorough assessment of these effects on organochlorine loading is required for a clear understanding of organochlorine exposure to shark populations. When examining shark organochlorine burdens, a method to diminish the variability introduced by 1 of these unique physiological processes is to look specifically at muscle tissue, despite the liver being the primary site for organochlorine accumulation. Shark muscle is buffered from body-condition related lipid depletion [18], whereas liver stores are relied on and can hence dramatically concentrate amounts of contaminant [19]. Because muscle concentrations are expected to reflect the whole-body level of contamination of an individual, this method is used in the present study to provide a robust lifetime-based view of shark organochlorine accumulation and overall feeding ecology, which resists alterations due to organochlorine bioamplification [20].

Dusky (*Carcharhinus obscurus*) and white (*Carcharodon carcharias*) sharks from the marine ecosystem of South Africa possess many similar life history traits that make them prone to organochlorine accumulation. This includes large body mass,

All Supplemental Data may be found in the online version of this article.
* Address correspondence to afisk@uwindsor.ca.
Published online 21 April 2015 in Wiley Online Library (wileyonlinelibrary.com).
DOI: 10.1002/etc.3029

large offspring (increased organochlorine offloading per individual) and late age-at-maturity (longer accumulative period prior to offloading). These species, however, differ in some key characteristics that are expected to influence organochlorine exposure and accumulation directly. First, South African dusky and white sharks are considered secondary and tertiary piscivores, respectively. Accordingly, the 2 species differ by approximately 1 trophic level (TL; dusky averaging TL4 and white TL5) [21]. Second, at a body length of $\geq$2 m, white sharks shift much of their predation efforts from teleost toward elasmobranch and mammal prey (particularly the Cape fur seal, *Arctocephalus pusillus pusillus*) [22]. Conversely, an overall depletion of $\delta^{15}N$ values with increasing dusky shark body mass has been observed [23]. This size-related decline in trophic position is thought to relate to an ontogenetic change from coastal occupation to foraging at the edge of the continental shelf on abundant schools of lower–trophic level fish prey [23]. Older white sharks would therefore be exposed to more contaminated prey than dusky sharks, because of the higher organochlorine biomagnification within the prey consumed. Third, regional endothermy in lamnids is said to drive greater energy requirements [24], as these sharks possess higher basal metabolic rates than ectothermic sharks [24]. This would result in white sharks consuming a large amount of prey biomass [25], greater than that of the ectothermic dusky shark. Appropriately then, it is expected that white sharks grow to a larger body size faster than do dusky sharks ontogenetically [26,27] (Supplemental Data, Figure S1). Because larger body size is correlated with a lower contaminant elimination capacity in fish [28–30], this higher growth rate may also contribute to heightened organochlorine accumulation in the white shark.

Evidence of differential organochlorine accumulation in shark species that differ in their ontogenetic diet shifts, thermal regulation, and growth rate may therefore help determine the effect of these factors on overall organochlorine exposure in sharks. The specific objectives of the present study were to compare observed organochlorine concentrations and profiles between the 2 species; to determine whether these can be explained by relationships with age, body mass, or stable isotope indicators of trophic position and habitat use; and to quantify and contrast the relationships between growth-dilution corrected organochlorine concentration and age in these 2 species.

METHODS

Sample collection

Dusky and white shark tissue samples were taken from animals caught in bather protection nets set approximately 400 m offshore, at depths of 10 m to 14 m at designated beaches along the KwaZulu-Natal coast off South Africa (for specific details, see Dudley [31]). Between 2005 and 2012, white muscle tissue was removed alongside the vertebral column anterior to the first dorsal fin from *C. carcharias* ($n = 53$; 28 female, 25 male, 160–363 cm precaudal length), and *C. obscurus* ($n = 42$; 22 female, 20 male, 106–270 cm precaudal length) during routine dissections by KwaZulu-Natal Sharks Board staff (Convention on International Trade in Endangered Species of Wild Fauna and Flora, South African permits 106704 and 106627). Because of bias in results due to maternal organochlorine transfer [32,33], no neonates (<100 cm precaudal length dusky sharks, <150 cm precaudal length white sharks) were included in the present study.

Precaudal length (mm), mass (kg), and sex were recorded for all individuals, and muscle tissue samples were immediately stored frozen (–20 °C). Samples were lyophilized (48 h), and shipped to the Chemical Tracer Lab at the Great Lakes Institute for Environmental Science (University of Windsor, Ontario, Canada) in 2009 and 2012 for both organochlorine contaminant and stable isotope analysis.

Organochlorine extraction and analysis

Using the method of Lazar et al. [34] with modification, muscle tissue was analyzed for 40 PCB congeners, p,p′-dichlorodiphenyltrichloroethane (DDT) and metabolites (p,p′-dichlorodiphenyldichloroethylene [DDE], p,p′-dichlorodiphenyldichlorethane [DDD]), chlorobenzenes (pentachlorobenzene, 1,2,3,4-tetrachlorobenzene, 1,2,4,5-tetrachlorobenzene and hexachlorobenzene), hexachlorocyclohexanes (α-HCH, β-HCH, γ-HCH), chlordanes (heptachlor epoxide, oxychlordane, *trans*-chlordane, *cis*-chlordane, *trans*-nonachlor and *cis*-nonachlor), octachlorostyrene, mirex, and dieldrin (Supplemental Data, Tables S1 and S2). All materials contacting the sample (glass, aluminum, polytetrafluoroethylene) were rinsed 3 times with acetone and hexane before use.

Briefly, solid/liquid column extraction using 15 mL 1:1 hexane (Hx):dichloromethane (DCM; % v/v) was carried out on 1 g freeze-dried muscle tissue previously weighed and desiccated in 10 g dried anhydrous sodium sulfate (Na_2SO_4). Internal standards of 50 μL of 694 ng/mL PCB 34 and 100 μL of 500 ng/mL BDE 71 were added. Column eluate was concentrated (Heidolph Hei-Vap Advantage, Model 561-01110-00) and brought to 10 mL in Hx. One mL of this solution was dried at 110 °C for 1 h for gravimetric lipid determination. Lipid was removed only from samples containing more than 0.15 g of lipid via gel permeation chromatography. Subsequent florisil separation involved elution through 6 g activated florisil, and elution volumes per fraction included 50 mL of hexane, 50 mL 15:85 DCM:Hx (% v/v), and 130 mL 60:40 DCM:Hx (% v/v). Eluate was rotaevaporated, brought to 1 mL in isooctane, and capped in 2-mL amber glass wide crimp-top autosampler vials (Agilent Technologies) before analysis on a gas chromatography–electron capture detector (GC System Model 6890A, Autosampler Tray Model 7683, Agilent Technologies).

Blanks and reference fish (carp) homogenate were run with every 6 samples to account for variation in results caused by the method itself and were within the criteria for the Great Lakes Institute for Environmental Science Organics Analysis Laboratory, certified by the Canadian Association for Laboratory Accreditation. Verification of compounds was performed on a subset of samples with gas chromatography–mass selective detector. Recovery percentages ranged from 41.96% to 101.54% (mean ± standard deviation; 75.9% ± 12.1 for 50 mL of hexane, 74.8% ± 14.1 for 50 mL 15:85 DCM:Hx [% v/v]), and blank and recovery correction was performed for all samples.

Carbon and nitrogen stable isotope determination

Stable isotope analysis was conducted using the method of Hussey et al. [22]. Briefly, freeze-dried samples were homogenized, lipid-extracted according to standard protocols, and analyzed on a continuous-flow isotope ratio mass spectrometer (Finnigan MAT Delta^{plus}; Thermo Finnigan). Atmospheric nitrogen was used as the reference standard for $\delta^{15}N$ and Pee Dee Belemnite Carbonate for $\delta^{13}C$. Stable isotope values were expressed as the deviation from standards in parts per thousand (‰). Analytical precision expressed as the

standard deviation of internal lab standard fish muscle and National Institute of Standards and Technology (NIST) 8414 bovine standard ($n = 7$ for each) was, respectively, 0.11‰ and 0.10‰ for $\delta^{15}N$ and 0.15‰ and 0.12‰ for $\delta^{13}C$. Analytical accuracy based on difference from NIST standards was within 0.14‰ of certified values for $\delta^{13}C$ of both sucrose (NIST 8542; $n = 58$) and L-glutamic acid (NIST 8573; $n = 57$) and within 0.24‰ of certified values for $\delta^{15}N$ of L-glutamic acid (NIST 8573; $n = 57$), ammonium sulfate (NIST 8548; $n = 57$), and nitrate (NIST 8549; $n = 18$).

Data analyses

All data analyses were performed using the statistical software, R (Ver 3.0.1 [35]). Appropriate nonparametric tests were selected where log-transformation did not normalize distributions or homogenize variance between species. Shark ages were calculated from precaudal length using published von Bertalanffy parameters for the 2 species (white 1–13 yr, parameters from Wintner and Cliff [26]; dusky 2–61 yr, parameters from Natanson et al. [27]). For comparison with shark species from different geographic locations (Table 1), approximate wet weight concentrations were calculated by applying a moisture content of 75.4% to dry weight concentrations. This value was determined from the regression line of the relationship between previously measured wet and freeze-dried weights of muscle from KwaZulu-Natal Sharks Board sharks, which was the same for all 8 species ($y = 3.9255x + 0.1863$, $r^2 = 0.97$, $p < 0.001$).

Differences in $\delta^{15}N$ values and log-transformed mass (kg) between species were determined using an independent samples *t*-test, whereas a 2-tailed Exact Wilcoxon Rank-Sum Test (package "exactRankTests," [36]) was used to compare differences in $\delta^{13}C$ between species.

To determine whether differences in ontogenetic contaminant profiles occurred between species, nontransformed compound concentrations were converted to relative proportions via division by the most abundant and highly recalcitrant compound, p,p′-DDE (with nondetects set to 0; Supplemental Data, Table S2). Proportions were then assessed for difference using ANOSIM on a Bray-Curtis dissimilarity matrix, followed by SIMPER (Package "vegan" [37]) to determine compounds contributing most to species' dissimilarity. The effect of life stage on intraspecific organochlorine profiles was also examined by converting nontransformed concentrations into p_i—that is, the proportion of a compound's concentration in an individual to the summed concentrations for that contaminant in the species overall (also setting nondetects to 0). A Shannon's Diversity Index value was then computed for individual sharks using $H = -(\Sigma p_i ln p_i)$, to assess variance in the diversity of compound profiles as a function of body mass (i.e., differential mobility and consequent habitat range) according to compound presence/absence and abundance. The H values were then log-transformed, and the relationships between mass and H scores were tested for the different species using a linear regression.

To determine whether overall organochlorine profiles within each species correlated with shark $\delta^{15}N$, $\delta^{13}C$, mass and sex, 2 separate principal component analyses (1 for each species) were performed using vegan on correlation matrices of white and dusky shark contaminant proportions (i.e., p,p′-DDE standardized) following removal of the 3 most extreme outliers (1 juvenile, and 2 adult dusky sharks; median absolute deviation test statistic ≥ 15). Proportions were used to account for contamination variability across the suite of contaminants as a whole (a more highly contaminated individual would tend to have higher concentrations of all compounds and vice versa). Shark $\delta^{15}N$, $\delta^{13}C$, mass, and sex (predictor variables) were then regressed (using additive multiple linear regression) against principle component scores from the first 3 axes (principle component 1, principle component 2, principle component 3: dependent variables) to explore how variations in contaminant profiles in each species were related to these shark characteristics. Because of concerns over multicollineary—because $\delta^{15}N$ and $\delta^{13}C$, as well as $\delta^{15}N$ and mass, were highly correlated—we used separate sets of regressions ($\delta^{15}N$ and sex were included in 1 set of regressions and $\delta^{13}C$, mass, and sex were included in a separate set of regressions) to explore the significance of individual predictors on principle component scores. For this principle component analysis, and in the calculation of average concentrations (Supplemental Data, Table S1), nondetects were set to the limit of quantification value.

To test for species differences in actual compound concentrations, only the 4 compounds that were found in every individual of each species were considered (p,p′-DDE, *t*-nonachlor, and PCBs 180 and 187), and concentrations were log-transformed prior to analysis. Accumulation differences between species and relationships between concentrations and predictor variables are expected to be well-exemplified by these congeners because they readily biomagnify, as expected for compounds at an octanol−water partition coefficient (log K_{ow}) near 7 (range, 6.35–6.96 [38,39]) [28]. Concentration differences in these compounds between species were tested using either a 2-tailed Exact Wilcoxon Rank-Sum Test, Student's *t*-test, or Welch's *t*-test, depending on data normality and variance homogeneity. To examine ontogenetic trends, correlations between concentrations of these compounds and shark mass, $\delta^{15}N$, and $\delta^{13}C$ were investigated (excluding neonates) using either linear regression or Spearman's ρ.

Finally, due to the potential for differences in growth-dilution to confound a proper comparison of uptake rates between species, concentrations of the 4 compounds found in all samples were growth-dilution corrected by standardizing to shark mass. Because observed concentrations could have been growth-diluted only to an extent equal to shark mass, concentrations were multiplied by the masses of the particular individuals in which they were found. Species' differences in slopes of the relationship between concentrations and age were then tested via analysis of covariance (ANCOVA). This analysis was undertaken both with white shark ages calculated according to Wintner and Cliff [26] and with these white shark ages doubled given potential age underestimation [40]. Dusky shark individuals with ages calculated as per Natanson et al. [27] were included in each species comparison, only as they corresponded to the white shark age range: first 1 yr to 13 yr of age and secondly 2 yr to 26 yr of age (8 dusky sharks were between 13–26 yr of age; therefore these 8 were added to the latter analysis).

RESULTS

Body mass, age, and stable isotopes

White (≥ 1.5 m) and dusky (≥ 1 m) sharks were found to differ significantly in $\delta^{15}N$ ($F = 109.6$), mass ($F = 11.3$), and $\delta^{13}C$ ($W = 232$; $p < 0.01$ for all, Figure 1). Significant polynomial regressions were found between log mass and both $\delta^{13}C$ and $\delta^{15}N$ in the dusky shark ($r^2 = 0.26$, $p < 0.05$, and $r^2 = .58$, $p < 0.001$, respectively) and between log mass and $\delta^{13}C$ in the white shark ($r^2 = .12$, $p < 0.05$). The relationship between white shark log mass and $\delta^{15}N$ approximated a parabolic curve but was not significant (Figure 1).

Table 1. Comparison of organochlorine concentrations in white and dusky sharks sampled from 2005 to 2012 from the KwaZulu-Natal, South Africa, with published concentrations in other marine predators

Class–tissue citation	Location	Species	*n*	Compound (ng/g, mean ± SE)													
				H. Epox	γ-HCH	*t*-non	*cis*-non	*t*-chl	*cis*-chl	Diel	p,p′-DDE	p,p′-DDT	PCB 149	PCB 118	PCB 153	PCB 138	PCB 180	PCB 170
Chondrichthyes—Muscle																		
Present Study	KwaZulu-Natal, South Africa	Dusky shark *Carcharhinus obscurus*	42[a]	0.04 ± 0.01	0.01 ± 0.002	0.3 ± 0.03	0.01 ± 0.002	0.1 ± 0.01	0.04 ± 0.004	0.1 ± 0.01	2.3 ± 0.6	0.1 ± 0.01	0.1 ± 0.01	0.2 ± 0.02	1.3 ± 0.2	1.1 ± 0.2	1.1 ± 0.3	0.4 ± 0.1
Present Study	KwaZulu-Natal, South Africa	White shark *Carcharodon carcharias*	53[a]	0.1 ± 0.01	0.02 ± 0.004	1.1 ± 0.1	0.1 ± 0.01	0.2 ± 0.02	0.1 ± 0.01	0.2 ± 0.03	5.0 ± 0.7	0.1 ± 0.01	0.3± 0.03	0.3± 0.04	1.4± 0.1	1.3± 0.1	0.8± 0.1	0.3± 0.02
Present Study	KwaZulu-Natal, South Africa	Dusky shark *C. obscurus*	42[b]	35.0 ± 19.4	5.8 ± 1.4	133.1 ± 26.4	6.4 ± 1.7	47.1 ± 9.5	25.8 ± 6.4	45.4 ± 14.8	1114.3 ± 302.7	35.8 ± 17.1	34.3± 14.5	107.0± 36.7	614.0± 153.4	542.9± 125.7	520.6± 153.5	174.8± 50.2
Present Study	KwaZulu-Natal, South Africa	White shark *C. carcharias*	53[b]	20.1 ± 6.9	5.8 ± 1.0	373.3 ± 34.8	17.9 ± 3.1	52.3 ± 7.0	26.5 ± 3.0	95.0 ± 18.8	1802.9 ± 339.2	18.2 ± 3.2	89.3± 13.7	102.8± 19.4	462.4± 67.1	422.7± 53.4	278.4± 32.1	87.3± 11.4
[41]	Kwazulu-Natal, South Africa	White shark *C. carcharias*	3[c]	1.5 ± 0.6	0.9 ± 0.4	–	–	–	–	–	1.6 ± 0.2	1.8 ± 0.6	–	–	–	–	–	–
[8]	Florida Bay, Tampa Bay, Apalachicola Bay, USA	Bonnethead shark *Sphyrna tiburo*	50[c]	0.4 ± 0.3	-	3.1 ± 0.6	2.2 ± 0.8	6.5 ± 0.7	4.0 ± 0.5	0.2 ± 0.9	4.5 ± 0.5	13.9 ± 2.0	–	–	–	–	–	–
Chondrichthyes—Liver																		
[41]	Kwazulu-Natal, South Africa	White shark *C. carcharias*	3[b]	26.1 ± 12.4	3.8 ± 0.2	–	–	–	–	–	1114.2± 379.0	55.5± 14.0	–	–	–	–	–	–
[43]	Canary Islands, North African Atlantic Ocean	Portuguese dogfish *Centroscymnus coelolepis*	9[b]	–	–	–	–	–	–	–	–	–	2.3± 0.7	20.6± 6.0	45.7± 18.3	45.0± 27.1	29.3±11.3	2.5± 0.9
[43]	Canary Islands, North African Atlantic Ocean	Shortnose velvet dogfish *Centroscymnus cryptacanthus*	1[b]	–	–	–	–	–	–	–	–	–	NA	NA	297.8	253.3	124.4	23.4
[43]	Canary Islands, North African Atlantic Ocean	Leafscale gulper shark *Centrophorus squamosus*	1[b]	–	–	–	–	–	–	–	–	–	138.9	1250.5	1056.5	67.3	12.2	56.8
[9]	USA East Coast	Sandbar shark *Carcharhinus plumbeus*	23[c]	2.5 ± 0.3	–	65.4± 10.1	36.6± 5.8	2.0± 0.9	7.7± 1.4	2.7± 1.4	445.6± 71.8	16.5± 4.3	–	–	–	–	–	–
[9]	USA East Coast	Blacktip shark *Carcharhinus limbatus*	25[c]	0.68 ± 0.1	–	41.7± 6.8	19.7± 3.2	1.7± 0.2	10.2± 1.8	1.7± 0.6	127.0± 29.2	4.5± 2.1	–	–	–	–	–	–
Mammalia—Blubber																		
[50]	Durban Whaling Station KwaZulu-Natal, South Africa	Minke whale *Balaenoptera acutorostrata*	28[b]	–	–	–	–	–	–	24.7± 4.8	64.5 ± 18.0	215.6 ± 69.6	–	–	–	–	–	–
[50]	Durban Whaling Station KwaZulu-Natal, South Africa	Fin whale *Balaenoptera physalus*	6[b]	–	–	–	–	–	–	ND	91.9 ± 40.9	115.0 ± 72.6	–	–	–	–	–	–
[50]	Durban Whaling Station KwaZulu-Natal, South Africa	Sperm whale *Physeter macrocephalus*	12[b]	–	–	–	–	–	–	ND	351.3 ± 38.8	364.9 ± 106.7	–	–	–	–	–	–
[42]	Crozet Archipelago, southern Indian Ocean	Killer whale *Orcinus orca*	9[b]	–	–	–	–	–	–	–	–	–	860 ± 180	–	2670 ± 380	1580 ± 260	860 ± 150	–

[a]Data are in ng/g wet wt with weight-based moisture content 75.40%, taken from previously determined regression line of relationship, between wet and freeze-dried weights of muscle tissue of KwaZulu-Natal Sharks Board sharks. The relationship applied to all species; $y = 3.9255x + 0.1863$, $r^2 = 0.97$, $p < 0.001$.

[b]Data are in ng/g lipid wt.

[c]Data are in ng/g wet weight.

SE = standard error; H.Epox = heptachlor epoxide; Non = nonachlor; Chl = chlordane; Diel = dieldrin; DDE = dichlorodiphenyldichloroethylene; DDT = dichlorodiphenyltrichloroethane; PCB = polychlorinated biphenyl.

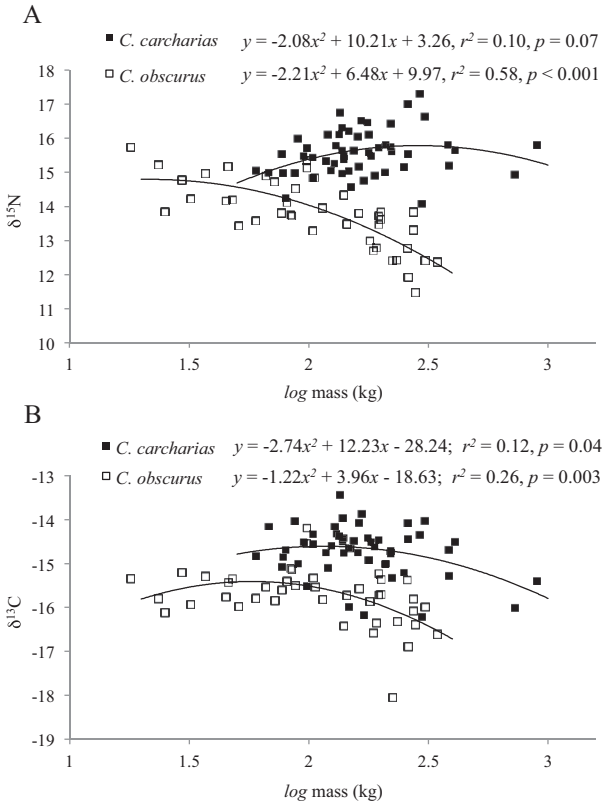

Figure 1. Relationship between mass and stable isotope values in dusky (*Carcharhinus obscurus*; open boxes) and white (*Carcharodon carcharias*; solid boxes) sharks sampled from 2005 to 2012 from the KwaZulu-Natal coast off South Africa: (**A**) $\delta^{15}N$ versus log mass, (**B**) $\delta^{13}C$ versus log mass.

Organochlorine compound ontogenetic profiles

Comparison of overall organochlorine accumulation pattern via ANOSIM revealed a significant difference in organochlorine profiles between species ($R = 0.5$, $p < 0.01$; Figure 2). Nine compounds were responsible for 52.5% of the dissimilarity between species: PCBs 138 (11.1%), 153 (7.9%), and 180 (7.3%); *t*-nonachlor (6.2%); and PCBs 33 (4.8%), 52 (4.6%), 44 (3.8%), 187 (3.4%), and 31/28 (3.4%; SIMPER, Figure 2). For white sharks, 13 compounds were found in all individuals (*cis*-chlordane, *t*-nonachlor, *cis*-nonachlor, p,p′-DDE, and PCBs 31/28, 153, 156/171, 170, 180, 183, 187, 194, 199). This contrasts with only 4 compounds found in every dusky shark (p,p′-DDE, *t*-nonachlor, and PCBs 180 and 187). No significant relationship was found between body mass and H scores of white or dusky sharks ($p > 0.05$ for all), indicating compound profiles were consistent over ontogeny.

Intraspecific compound profiles and predictor variables

The first 3 principle component axes extracted from the PCAs performed on white and dusky shark contaminant proportions explained 69.7% and 53.6% of the total contaminant variation, respectively. For white sharks, only $\delta^{13}C$ was significantly related to principle component 2 (no other significant relationships were found for any predictor with principle component 1, principle component 2, or principle component 3) via the following univariate relationship: principle component $2 = 0.100 \times \delta^{13}C + 1.477$, $r^2 = 0.170$, ($p < 0.01$). Proportions of PCBs 153, 138, 128, 180, 170, 194, and 206 in white sharks all loaded positively and heavily on principle component 2 (loadings >0.714; Supplemental Data, Table S3), and PCBs 128, 180, and 206 were positively correlated with $\delta^{13}C$ (Pearson's $r > .29$, $p < 0.05$). For dusky sharks, principle component 1 was positively related to mass (principle component $1 = 0.236 \times log_{10}[\text{mass}] + [-0.474]$, $r^2 = 0.227$, $p < 0.01$). Only mirex and PCB 194 loaded positively, although weakly (loading = 0.16 and 0.12, respectively) on principle component 1 of the dusky shark principle component analysis, but only mirex was significantly positively correlated to dusky shark mass ($r = 0.44$, $p < 0.05$). Proportions of several compounds (PCBs 31/28, 44, 95, 99, *c*-chlordane, and *t*-nonachlor) loaded negatively and heavily (loadings < –0.70; Supplemental Data, Table S3) on principle component 1 but only PCBs 95, 99, and *c*-chlordane were significantly, negatively correlated with dusky shark mass ($r =$ –0.40, –0.35, –0.57, respectively, all $p < 0.05$). No other relationships were found between principle component axes and mass, $\delta^{15}N$, $\delta^{13}C$, or sex in dusky sharks.

Organochlorine concentrations and predictor variables

Organochlorine concentrations were different between species for 2 of the 4 examined compounds, *t*-nonachlor and p,p′-DDE, with elevated concentrations in the white shark ($p < 0.001$; log K_{OW} 6.35 and 6.96, respectively). Concentrations of

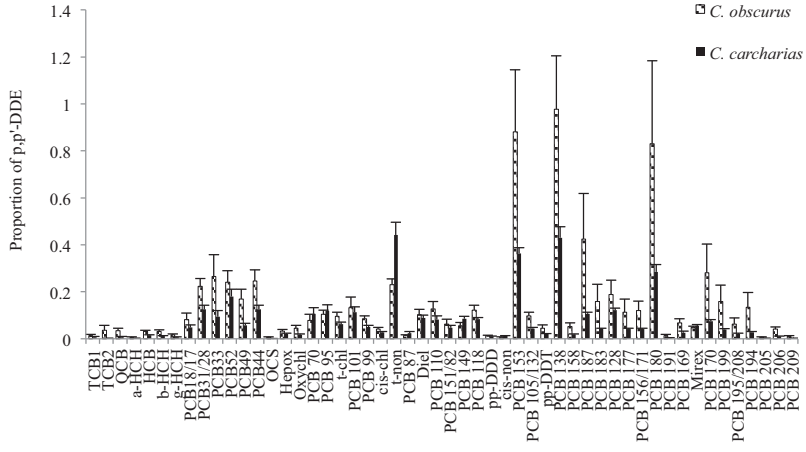

Figure 2. Organochlorine contaminant profile comparison between dusky (*Carcharhinus obscurus*; hatched bars) and white (*Carcharodon carcharias*; solid bars) sharks sampled from 2005 to 2012 from the KwaZulu-Natal coast off South Africa. DDE = dichlorodiphenyldichloroethylene; TCB = tetrachlorobenzene; OCB = penatchlorobenzene; HCH = hexachlorocyclohexanes; HCB = hexachlorobenzene; PCB = polychlorinated biphenyls; OCS = octachlorostyrene; Diel = dieldrin; DDD = dichlorodiphenyldichlorethane; DDT = dichlorodiphenyltrichloroethane.

t-nonachlor were 4.6 ± 0.3 ng/g and 1.0 ± 0.1 ng/g in white and dusky sharks, respectively, and p,p′-DDE concentrations were 20.2 ± 2.7 ng/g and 9.3 ± 2.2 ng/g, respectively (Supplemental Data, Table S1). In dusky sharks, significant negative relationships were found between $\delta^{15}N$ and p,p′-DDE ($r^2 = 0.11$), and between $\delta^{13}C$ and p,p′-DDE ($r_s{}^2 = 0.10$), PCB 180 ($r^2 = 0.15$), and PCB 187 ($r^2 = 0.12$) concentrations ($p < 0.05$ for all), whereas a positive relationship was found between mass and p,p′-DDE ($r^2 = 0.22$, $p < 0.01$; Figure 3A). For white sharks, a positive relationship was found between PCB 180 and $\delta^{13}C$ ($r^2 = 0.17$, $p < 0.01$; Figure 3B). Among all sharks, *t*-nonachlor and p,p′-DDE had positive relationships with $\delta^{15}N$ ($r^2 = 0.38$ [Figure 3C], $r_s{}^2 = 0.06$, respectively), $\delta^{13}C$ ($r^2 = 0.34$, $r_s{}^2 = 0.08$, respectively), and mass ($r_s{}^2 = 0.06$, $r^2 = 0.12$, respectively; $p < 0.05$ for all).

Differences in organochlorine accumulation with age

Slopes of the relationship between growth-dilution corrected organochlorine concentrations and age as calculated from Wintner and Cliff [26] and Natanson et al. [27] did not differ significantly between species for any of the 4 compounds, whether or not white shark ages were doubled according to Hamady et al. [40] (Figure 4; ANCOVA interaction, $p > 0.05$ for all).

DISCUSSION

The present study compared organochlorine ontogenetic profiles, concentrations, and accumulation patterns in 2 shark species that contrast each other in important ecological and life history parameters. Among all individuals, organochlorine concentrations had significant positive relationships with variables generally predicting organochlorine dynamics in fish (body mass and stable isotopes), but fewer were found intraspecifically. Despite the distinct life history characteristics of these species, organochlorine concentrations did not increase at different rates between the species with age. However, concentrations of the highly biomagnifying compounds, *t*-nonachlor and p,p′-DDE, were significantly elevated in the higher–trophic level and larger-bodied white shark. The present study provides insight into the trophic ecology of these species, indicating that differences in influential life history characteristics, such as trophic level and body mass, are weak intraspecific predictors of organochlorine dynamics, but remain accurate predictors at the food web level.

Muscle p,p′-DDE concentrations of dusky and white sharks in the present study were higher than blubber concentrations reported from other large marine predators caught in KwaZulu-Natal (Minke whale [*Balaenoptera* sp.], Fin whale [*B. physalus*], and Sperm whale [*Physeter macrocephalus*]), but were similar to concentrations reported previously in KwaZulu-Natal white sharks [41] (Table 1, lipid-based). Notably, lipid-based concentrations of p,p′-DDT in sharks from the present study were approximately half the amount found previously in KwaZulu-Natal white sharks (Table 1 and Schlenk et al. [41]). Because this previous study included only 3 white shark individuals, it is likely that this difference is due to the high variation in levels actually present in the KwaZulu-Natal white shark population being insufficiently captured in this small sample size. Lipid-based concentrations of PCBs 138, 149, 153, and 180 quantified in the present study are less than half the concentration observed in blubber of the killer whale, *Orcinus orca*, from the Crozet Archipelago, the most southerly comparable location [42], but are more than an order of magnitude higher than previously found in Portuguese dogfish liver (*Centroscymnus coelolepis*) from the northwestern African Atlantic Ocean [43]. Although this suggests higher contamination in the Southern Hemisphere, comparable concentrations of PCBs were found in the shortnose velvet dogfish (*Centroscymnus cryptacanthus*), and nearly double the amount of PCB 153, as well as over an order of magnitude higher PCB 118 were found in the leafscale gulper shark (*Centrophorus squamosus*), both also from the northwestern African Atlantic. It is therefore likely that such low concentrations in the Portuguese dogfish are instead a result of different feeding ecology.

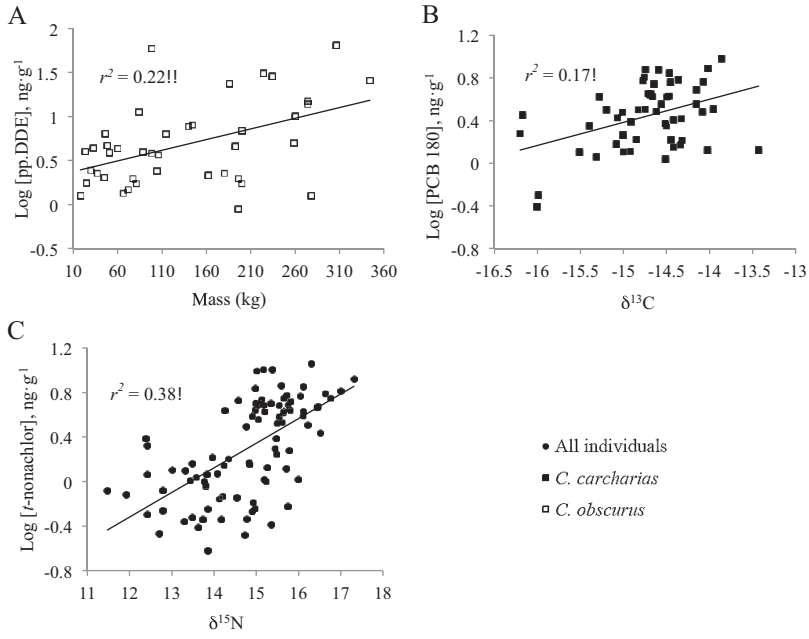

Figure 3. Significant relationships between organochlorine concentrations and predictor variables: (**A**) mass versus p,p′-dichlorodiphenyldichloroethylene in *Carcharhinus obscurus* (open boxes); (**B**) $\delta^{13}C$ versus polychlorinated biphenyls 180 in *Carcharodon carcharias* (closed boxes); and (**C**) $\delta^{15}N$ versus *t*-nonachlor over all individuals of both species (closed circles). p,p′-DDE = p,p′-dichlorodiphenyldichloroethylene; PCB = polychlorinated biphenyls.

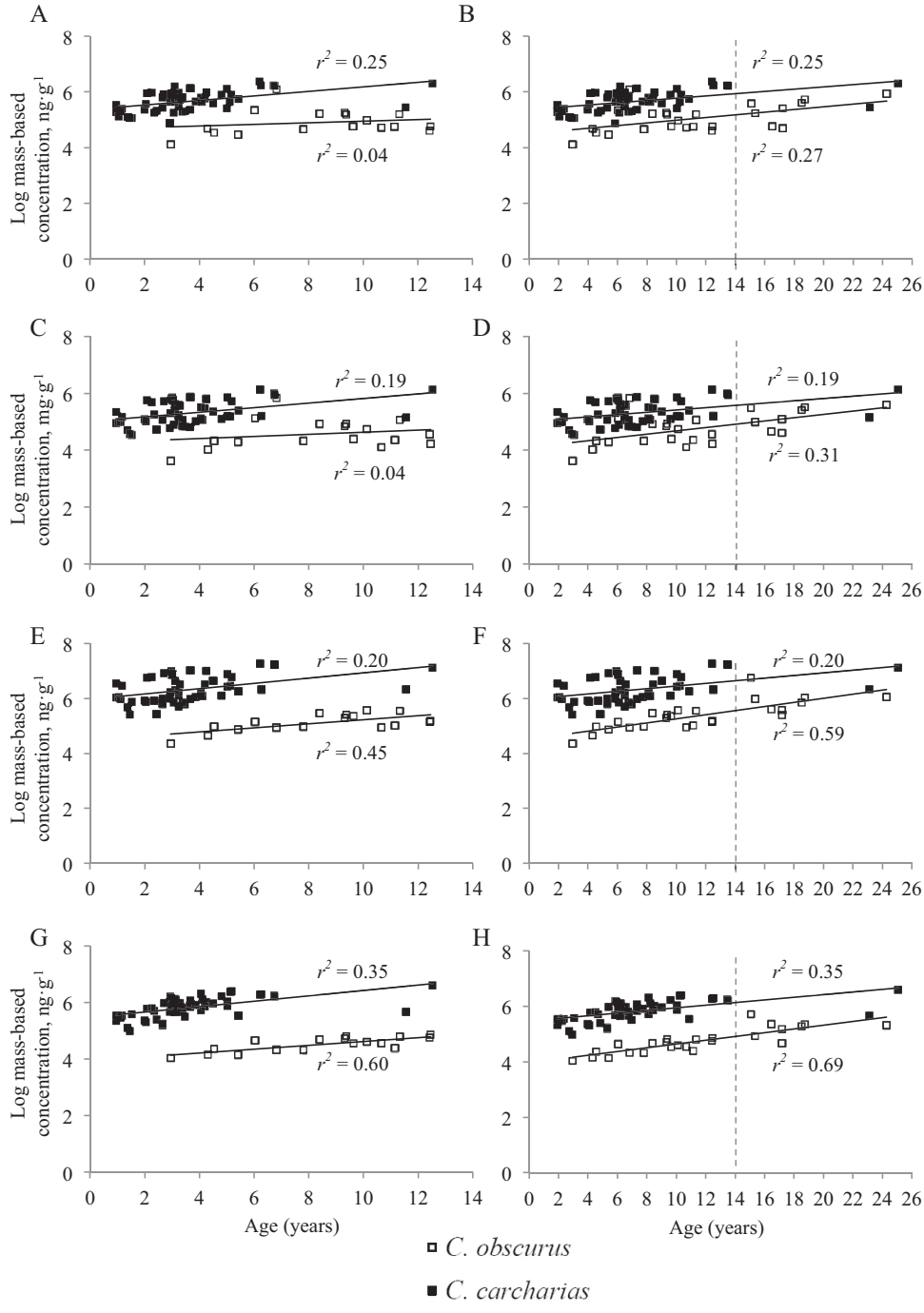

Figure 4. Slope comparison of growth-dilution corrected concentrations: (**A**,**C**,**E**,**G**) *Carcharodon carcharias* ages as per [36]; (**B**,**D**,**F**,**H**) *C. carcharias* ages doubled [35] and 8 additional *Carcharhinus obscurus* added to comparison. (**A**,**B**) Polychlorinated biphenyl (PCB) 180, (**C**,**D**) PCB 187, (**E**,**F**) p,p′-dichlorodiphenyldichloroethylene, (**G**,**H**) *t*-nonachlor.

White shark KwaZulu-Natal p,p′-DDE concentrations were nearly equal to concentrations reported from a short-lived, low–trophic level species (bonnethead sharks, *Sphyrna tiburo*) sampled from Florida [8]. Dusky and previously sampled white sharks from KwaZulu-Natal [41] contained approximately half this amount. Higher–trophic level sharks (blacktip, *Carcharhinus limbatus* and sandbar, *Carcharhinus plumbeus*) from the Florida and Georgia regions of the United States were found to have high wet weight concentrations of p,p′-DDE in the liver, as would be expected; but because these values were not expressed in lipid weight, a portion of this increase is a result of higher lipid concentration in liver than muscle tissue. Regardless, the present study's findings may indicate that for Southern Hemisphere animals, only those compounds with the highest propensity for biomagnification are likely to reach similar concentrations to low–trophic level organisms in the Northern Hemisphere. Furthermore, the above-mentioned conversion to the p,p′-DDE metabolite now provides white sharks with similar concentrations of this particular compound.

White and dusky sharks had significantly different organochlorine profiles, indicating differential habitat use or feeding ecology of these species throughout their ontogeny. Differences in $\delta^{13}C$ profiles between dusky and white sharks also indicate potential variation in habitat use across all life stages, with the

higher $\delta^{13}C$ values of the white shark suggesting more inshore feeding, a behavior supported by previous satellite tracking data [44]. The overall contaminant profile of white sharks is in agreement with this feeding behavior, given a comparatively more even contamination across the full suite of analyzed compounds (i.e., as proportions of p,p′-DDE) and a proportionate increase in select contaminants (PCBs 128, 180, and 206) with increased inshore predominance. Both of these may be attributed to higher organochlorine concentrations in inshore habitats. Conversely, dusky shark contamination was dominated by the 3 compounds contributing most to dissimilarity between the species' profiles: PCB 153, 138, and 180. These congeners, along with p,p′-DDE, are highly persistent and may be expected to predominate in typically less-contaminated pelagic food webs (hexa- and hepta-chlorinated PCB congeners are shown to predominate in pelagic species [45]). This is supported by the fact that subadult and adult dusky sharks predominantly occur and feed in the pelagic habitat at the edge of the continental shelf [22,46]. Decreasing proportions of PCBs 95, 99, and *c*-chlordane with dusky body mass are likely the combined result of these compounds not being very highly bioaccumulative and decreasing in contamination with ontogenetic movement of the dusky offshore.

Although a positive relationship between body mass and organochlorine concentrations is well-documented for fish species [47], the absence of positive relationships between organochlorine concentration and body mass observed in the white shark is not an unusual finding when compared with other top–trophic level marine predators. High–trophic level mammal and shark species commonly have a negative or null relationship between organochlorine concentrations and body mass, for example in *O. orca* (southern Indian Ocean [42]) and in the salmon shark, *Lamna ditropis*; thresher shark, *Alopias vulpinus*; and white shark sampled from southern California [48]. As Lyons et al. [48] suggested, positive relationships between organochlorine concentrations and mass in lamnids may occur only after growth rate has decreased enough to reduce the effect of dilution on bioaccumulation. White sharks in the present study, however, included only individuals of life stages that would correspond to positive growth (Supplemental Data, Table S1). Given the scenario Lyon et al. suggested [48], it is possible that dusky sharks may indeed grow much more slowly than white sharks, as was previously thought [26]. This results in the positive relationships of both the overall compound profile principle component analysis response, and p,p′-DDE concentrations observed with dusky mass. Alternatively, the greater ability for mass to predict organochlorine concentrations in dusky sharks may be due to the fact that those sampled in the present study included animals more than 40 yr old, meaning slight changes in mass could represent many years of accumulation.

Our expectation that key differences in life history traits between species would result in higher white shark organochlorine accumulation with age was not supported by slope differences. Ontogenetic diet shifting to higher–trophic level and more contaminated mammal prey, regional endothermy driving increased biomass intake per unit time, and a likely higher growth rate enhancing organochlorine uptake and assimilation in white sharks were all insufficient to drive an increased slope of organochlorine accumulation with age when compared with the dusky shark. The high similarity in slopes overall may be partly explained by the fact that declining $\delta^{15}N$ values with mass in the dusky shark may not be a result of switching to feeding on only lower−trophic level prey (i.e., small schooling fish). Instead, dusky sharks may feed on large pelagic prey with comparable concentrations of organochlorine contamination to that in white shark prey. Slopes may also be similar because of a lower frequency of seal-predation by white sharks than expected; because of high individual variability in predation success on mammal prey [22]; or because adult, large dusky sharks also incorporate elasmobranch, dolphin, and other mammal prey [23] in similar proportions to subadult white sharks. Consequently, the observed decline in $\delta^{15}N$ values of larger dusky sharks is likely a result of a lower ecosystem $\delta^{13}C$ baseline value in the pelagic food web [23]. This is further supported by the positive linear relationship between concentrations of p,p′-DDE and mass, and none with $\delta^{15}N$, providing evidence that body mass is a better predictor of organochlorine concentrations in the dusky shark. Although these scenarios may account for this unexpected comparative organochlorine accumulation, the possibility that currently available von Bertalanffy growth parameters [26,27] are inaccurate for these species may also be a confounding factor.

Our hypothesis that white sharks contain higher concentrations of biomagnifying organochlorines than the dusky shark was supported by elevated concentrations of p,p′-DDE, and *t*-nonachlor. This is a result of their higher trophic position, as indicated by significantly higher $\delta^{15}N$ values and body mass of white sharks. Because of a positive relationship between these compounds and both $\delta^{15}N$ and mass among all shark individuals, it is apparent that body mass also plays a role in determining organochlorine burdens among shark species of different sizes. This is expected because of decreasing elimination capacity with fish body mass [28–30]. As detailed, although positive relationships between organochlorine concentrations and white shark mass were not observed, this effect was likely due to not sampling larger individuals and resultant masking by growth-dilution [48]. Because fish (including sharks) have indeterminate growth, verifying the importance of body mass by comparing species with similar trophic positions but different body masses is difficult [49]. It has been well demonstrated in the present study, however, given the high similarity in accumulation patterns with age but overall higher concentrations in the large-bodied white shark. This does not preclude, however, differing trophic positions as an important determinant of organochlorine concentrations. Although relationships between organochlorine concentration and $\delta^{15}N$ were absent within each species, a positive overall trend among all shark individuals was observed with $\delta^{15}N$. Given the fundamental mechanism of organochlorine biomagnification and the structure of trophic systems, trophic level is a consistently reliable indicator of organochlorine concentrations in food webs [49].

SUMMARY

With growth-dilution correction, slopes of white and dusky shark organochlorine accumulation with age were similar. Examples of significantly higher concentrations of some readily biomagnifying compounds were found in the white shark, attributable to higher–trophic level feeding and larger body mass. No correlations were found in white sharks between mass and organochlorine concentrations, but this relationship was positive in dusky sharks and may be attributed to either more rapid growth-dilution in white sharks or the inclusion of much older dusky shark individuals in the analyses. Inshore habitat use was found to be an important predictor of organochlorine

variation in white sharks, and this was absent in dusky sharks because of the predominant occurrence of subadult and mature individuals in pelagic food webs at the edge of the continental shelf. In conclusion, although $\delta^{15}N$ and mass did not predict organochlorine concentrations in both species intraspecifically, they adequately predicted concentrations of some of the most highly biomagnifying organochlorine compounds between these species. The present study has contributed to assessing the variability in physiological and ecological processes governing organochlorine accumulation in sharks, determining that although regional endothermy and diet shifts have lesser impact on overall accumulation rates, trophic position and body mass may be regarded as reliable predictors of interspecific organochlorine accumulation.

SUPPLEMENTAL DATA

Tables S1–S3.
Figure S1. (159 KB DOC).

Acknowledgment—The authors acknowledge the financial support of the Natural Sciences and Engineering Research Council (NSERC) during the present study, through the Canada Research Chair programme (CRC 950-203515) and NSERC Discovery Grant (DG 05490) to A.T. Fisk, and NSERC Canada Graduate Scholarships-Master's to M. Beaudry. Thank you to the KwaZulu-Natal Sharks Board operations and laboratory staff for providing the sharks and for dissections and sample collection, which enabled the present study to be performed. Thank you also to A. Hussey for stable isotope analysis and to S. Isaac, J. Nix, and A. Tanner for organochlorine extractions.

Data availability—Data are available on request to the corresponding author, A.T. Fisk (afisk@uwindsor.ca).

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