Legacy and emerging organic contaminants in two sympatric shark species from Reunion Island (Southwest Indian Ocean): Levels, profiles and maternal transfer

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1. Introduction

Marine apex predators, such as marine mammals, tunas, billfish and sharks, act as both drivers of the ecosystems in which they live and sentinels of environmental health (Green and Larson, 2016; Ramos and González-Solís, 2012). Due to their long lifespans and position at the top of the food chain, they tend to exhibit high concentrations of persistent and hydrophobic chemical contaminants in tissue *via* bioaccumulation and biomagnification (Borgå et al., 2004; Kelly et al., 2007; Mackay and Fraser, 2000).

Contaminants that are persistent, bio-accumulative and toxic, such as persistent organic pollutants (POPs), may inflict the greatest amount of damage on ecosystems (UNEP, 2001; Green and Larson, 2016; Jepson and Law, 2016). POPs, which are essentially synthesized by humans are characterized by their ability to travel long distances, resulting in their transfer far from emission sources (Beyer et al., 2000; Brown and Wania, 2008; Corsolini et al., 2014), mainly through atmospheric volatilization and condensation cycles referred to as global distillation (Dachs et al., 2002; Wania and MacKay, 1996). POPs are hence widespread in the environment; they occur in various matrices and regions around the world (Dachs et al., 2002; Pozo et al., 2006; Reid et al., 2000) and as such are considered as global pollutants (Pozo et al., 2006). POPs include well-studied polychlorinated biphenyls (PCBs) and various organochlorine pesticides (OCPs), as well as other substances listed more recently in the Stockholm Convention (UNEP, 2001), such as perfluorooctane sulfonate (PFOS). Historically, OCPs, in particular hexachlorocyclohexane (HCH) and dichlorodiphenyltrichloroethane (DDT), were synthesized in high quantities (Li, 1999; Li and Macdonald, 2005). Although the production of legacy POPs has ceased in most countries, contemporary inputs into the environment continue to originate from secondary sources such as open burning, disposal of products containing PCBs, recycling of electric and electronic devices or PCB-containing pigments used in household paints (Breivik et al., 2011; Grimm et al., 2015), or from direct use, e.g. DDT against vectors of diseases (Qiu et al., 2005; van den

Berg et al., 2017). The ban on certain chemical substances has led to their replacement with new substances, classified as contaminants of emerging concern (CECs). For example, the ban on PFOS has led to the use of long-chain (> 7 carbon atoms) perfluorinated carboxylic acids (PFCAs), found in increasing amounts in the environment (Wang et al., 2017). Oceans are recognized as the main reservoirs of these compounds (Johansson et al., 2019). In this context, both legacy POPs and CECs need to be studied in many oceanic regions, in particular those remote from major sources of direct pollution, such as the southern Indian Ocean (Corsolini et al., 2016; Hoydal et al., 2015; Roscales et al., 2016; Trumble et al., 2012).

The tiger shark (*Galeocerdo cuvier*: Péron & Lesueur, 1822) and bull shark (*Carcharhinus leucas*: Müller & Henle, 1839) are apex predators inhabiting both coastal and oceanic tropical ecosystems (Compagno, 1984), where they play a major role (Heithaus et al., 2008). Both species are generalist feeders that can forage on prey of aquatic or terrestrial origin (Cliff and Dudley, 1991; Dicken et al., 2017; Trystram et al., 2017). More specifically, in Reunion Island (RUN), an oceanic island in the Southwest Indian Ocean, these two sympatric species differ in terms of prey and trophic habitat specialization (Le Croizier et al., 2020;Trystram et al., 2017). Both species are characteristic of the marine fauna of RUN (Fricke et al., 2009) and play a key role in the island's coastal ecosystem dynamics. However, no rigorous assessment of their exposure to organic contaminants has so far been conducted, despite the fact that they feed at high trophic levels, as do marine mammals (Jepson and Law, 2016), which are subject to excessively high concentrations of PCBs associated with long-term population declines (Dirtu et al., 2016; Mwevura et al., 2010; Tanabe, 2002). Actually, the contamination of marine ecosystems by organic contaminants in RUN has been the subject of a very limited number of studies (Dirtu et al., 2016; Munschy et al., 2016), in particular with regards to CECs. More generally, organic contaminant occurrence in large carnivorous shark species has been poorly documented (Cagnazzi et al., 2019; Fisk et al., 2002; Gelsleichter et al., 2005; Lee et al., 2015; Schlenk et al., 2005; Weijs et al., 2015), despite the fact that most of these species are threatened with overexploitation and habitat

destruction on a global scale (Dulvy et al., 2017; Ferretti et al., 2010). Studying the accumulation of toxic pollutants in large sharks in general, and in tiger and bull sharks in particular, is therefore essential. In addition, sharks have been shown to offload significant amounts of contaminants to their offspring, potentially resulting in female depuration (Lyons and Adams, 2015). Maternal transfer of toxic substances bioaccumulated in mothers is of particular concern, as i) it may represent a risk for offspring due to early-stage exposure; ii) it contributes to the global organic contaminant cycle *via* maternal offloading and transfer to offspring prior to other external exposure (Lyons and Adams, 2015; Mull et al., 2013). In elasmobranchs, pregnant females transfer chemical contaminants to their offspring through various pathways related to their reproductive strategy (oviparity, ovoviviparity, aplacental or placental viviparity), which may in turn affect contaminant levels and profiles in offspring (Cagnazzi et al., 2019; Lyons and Adams, 2015; Lyons and Lowe, 2013; Mull et al., 2013). The bull shark is a viviparous species whose embryos are fed by direct maternal inputs *via* a placental bond after absorption of yolk reserves, while in the ovoviviparous tiger shark, embryonic development is solely ensured by a yolk, with additional nutritional inputs during gestation (Castro et al., 2016). Both of these modes of gestation incur maternal transfer of hydrophobic pollutants (Lyons and Adams, 2015; Mull et al., 2013; Olin et al., 2014; Weijs et al., 2015). Our study aimed to characterize the contamination of tiger and bull sharks from RUN by legacy POPs and CECs in order to identify contaminant sources and explore the potential use of contaminants as trophic habitat tracers. The influence of biological parameters (size as a proxy of age, sex, lipid content) on contaminant bioaccumulation and maternal transfer were also studied. The presented data on the contamination of two top predator sharks from the Indian Ocean could constitute an essential benchmark for further studies.

2. Materials and methods

2.1. Sample collection

Tiger sharks (*Galeocerdo cuvier,* n = 21) and bull sharks (*Carcharhinus leucas*, n = 18) were collected in 2018 and 2019 as part of a shark control program implemented following an increase in shark attacks on the West coast of RUN (Lagabrielle et al., 2018). All individuals were caught along the West coast of the island, where most nautical activities take place, at depths between 10 m and 70 m (ca. 0.2 to 2 km from the shore) using bottom setlines and SMART (shark management alert in real-time) drumlines (Guyomard et al. 2019) and were dissected less than 36 h after capture. On the basis of demographic parameters for the two species in RUN (Pirog et al., 2019a, 2020), the studied specimens included both mature and immature individuals, including one 82 cm female bull shark specimen considered as offspring of that year (referred to as "young-of-the-year" later in the text). Our sampling strategy focused on a wide range of morphometrics for the purpose of studying organic contaminant bioaccumulation as a function of individual size. Total mass (W in kg) and total length (measured from the tip of the nose to the end of the tail, T_L in cm) were measured in all studied individuals to the nearest gram and centimetre respectively, and sex was determined through visual observation (presence/absence of claspers in males/females respectively). In addition, all non-empty or regurgitated stomach contents were analysed to describe diet. In order to assess shark physiological condition, the Fulton's condition factor (K) was calculated as follows:

$$K = 100 \times ((W \times 10^3) \times {T_L}^{-3})$$

whereby W is mass (kg) and T_L is total length (cm) (Gilliers et al., 2004).

Muscle was chosen as a good integrator of organic contaminants over time due to its slower turnover rate compared to liver (Cullen et al., 2019). White muscle was collected between the dorsal fins of adults. Embryos were also collected from one female of each species. A total of eight embryos were collected from the bull shark mother and ten embryos from the tiger

shark mother. Whole dorsal muscle was taken and pooled from 2 individuals of same sex and similar sizes to obtain a total of 4 pooled samples for tiger and bull sharks; two tiger sharks embryos were also analysed individually. Shark embryos are good indicators of maternal transfer of pollutants as they are not exposed to external food.

All samples were stored in amber glass vials at -20°C until further treatment. Prior to total lipid content and organic contaminant analyses, the samples were homogenized using a blender, freeze-dried for 72 h and finely ground using a MM200 ball mill (Retsch).

2.2. Total lipid content analysis

Extractable organic matter, used as a proxy for total lipid content (TLC), was determined with 0.5 g dry weight (dw) of sample extracted with a mixture of n-hexane and acetone (80/20 volume/volume [v/v]) using pressurized liquid extraction (PLE) at 100 °C under 10 MPa by means of an ASE 350 (Dionex©) (Munschy et al., 2020). The extracts were evaporated to dryness and TLC was determined gravimetrically and expressed in % of wet weight (ww).

2.3. Organic contaminant analysis

PCBs and OCPs were determined as described by Munschy et al., 2016. Briefly, 5-10 g of samples were extracted by PLE with dichloromethane. Prior to extraction, $^{13}C_{12}$-labelled compounds (18 PCBs, including 12 dioxin-like (dl-), 6 indicator (i-) PCBs, 5 DDT isomers, aldrin, dieldrin, endrin, isodrin, α-, β-endosulfan and endosulfan-sulfate) were added to the sample for internal standard calibration and quantification using the isotopic dilution method. The extracts were successively purified using gel permeation chromatography, a silica and alumina adsorption chromatography column and two-dimensional HPLC system with two columns coupled in series. Four fractions were obtained (F1: non-coplanar PCBs and *p,p'*-DDE, F2: coplanar PCBs, F3: OCPs, F4: remaining OCPs, treated with concentrated sulphuric acid). Analyses were performed by gas chromatography coupled with high resolution mass spectrometry (GC–HRMS) using a Hewlett-Packard 6890 gas chromatograph fitted with an SGE HT-8 capillary column (50 m × 0.22 mm × 0.2 µm) and

coupled to an AutoSpec Ultima mass spectrometer (Waters Corp.). The samples were analysed for 30 PCBs ranging from trichlorinated to decachlorinated congeners, including the 12 dioxin-like (dl) -PCBs (CB-77, -81, -105, -114, -118, -123, -126, -156, -157, -167, -169, -189), the 6 indicator (i)-PCBs (CB-28, -52, -101, -138, -153, -180) and various OCPs (*p,p′*-DDT, *o,p′*-DDT, *o,p′*-DDD, *p,p′*-DDD, *p,p′*-DDE, dieldrin, aldrin, isodrin, mirex and hexachlorobenzene -HCB). Compounds were quantified by isotopic dilution using the corresponding $^{13}C_{12}$-labelled isomers (except mirex, quantified using ^{13}C *p,p'*-DDE) and internal standard method was used to quantify samples. Prior to injection, a solution containing $^{13}C_{12}$-labeled CB-70, -111 and -170, d_8-labeled *p,p'*-DDD and *o,p'*-DDT was added to the final purified extracts for signal correction. Perfluoroalkylated substances (PFASs) were determined according to Munschy et al., 2019. Briefly, one gram of a freeze-dried sample, to which an internal standard mixture of nine labelled compounds was added prior to agitation, was extracted using liquid-solid extraction with MeOH/KOH (0.01 M of KOH), purified onto two consecutive SPE cartridges (a WAX weak anion exchange stationary phase and an Envicarb charcoal stationary phase, evaporated to dryness and reconstituted in 200 µL of a mixture of MeOH:H_2O (50:50, v/v), to which PFOS $^{13}C_8$ was added. The following compounds were analysed, including five C_4- to C_{10}-perfluoroalkyl sulfonates (PFSAs) and nine C_6- to C_{14} perfluorocarboxylic acids (PFCAs): perfluorobutane sulfonate (PFBS); perfluorohexane sulfonate (PFHxS); perfluoroheptane sulfonate (PFHpS); perfluorooctane sulfonate (PFOS); perfluorodecane sulfonate (PFDS); perfluorohexanoic acid (PFHxA); perfluoroheptanoic acid (PFHpA); perfluorooctanoic acid (PFOA); perfluorononanoic acid (PFNA); perfluorodecanoic acid (PFDA); perfluoroundecanoic acid (PFUnDA); perfluorododecanoic acid (PFDoDA); perfluorotridecanoic acid (PFTrDA) and perfluorotetradecanoic acid (PFTeDA). Targeted analytes were quantified using the corresponding isotope labelled standard, unless otherwise stated. The labelled standards were PFHxS $^{18}O_2$ (used to quantify PFBS and PFHxS), PFOS $^{13}C_4$ (used to quantify PFHpS, PFOS and PFDS), PFHxA $^{13}C_2$ (used to quantify PFHxA and PFHpA), PFOA $^{13}C_4$, PFNA $^{13}C_5$, PFDA $^{13}C_2$, PFUnDA $^{13}C_2$, PFDoDA $^{13}C_2$, and PFTeDA $^{13}C_2$ (used to quantify PFTrDA

and PFTeDA). PFOS $^{13}C_8$ was added to the purified extracts before injection and used as an injection standard. Analysis was performed using an Acquity ultra-performance liquid chromatograph (UPLC®, Waters Corp.) coupled to a triple quadrupole mass spectrometer (Xevo® TQ-S micro, Waters Corp.) interfaced with a Z-sprayTM (Waters Corp.) electrospray ionization source. UPLC separation was achieved using an Acquity UPLC BEH C18 reversed-phase column (50 mm x 2.1 mm; 1.7µm particle size) eluted with ammonium acetate in water (20 mM) (A) and methanol (B).

2.4. QA/QC

Analyses were conducted under fume hoods with positive pressure in a low-dust laboratory to minimize external and cross-contamination. For each series of 10 samples for PCBs and OCPs and of 15-20 samples for PFASs, one in-house quality control (QC) sample and one procedural blank were processed.

The QC for PCBs and OCPs was prepared from fish muscle spiked with OCPs at 0.1 - 0.2 ng g^{-1} dw, while method performances were assessed using PCBs already present in the sample. For PFASs, the QC was prepared using commercially-purchased whole mussel tissue spiked with the targeted compounds at 0.2 to 0.3 ng g^{-1} dw. Trueness, calculated as the relative bias compared to targeted values, was 0.3 - 23% for PCBs, 0.4 - 23% for OCPs and 5 - 33% for PFASs.

Procedural blanks, incorporating all steps of the protocol, were analysed with each series of 12 samples (n = 4). Contaminants detected in more than 50% of the procedural blanks included various PCB congeners (CB-28, -31, -44, -49, -52, -66, -77, -87, -101, -105, -110, -118, -132, -138, -149, -151, -153, -156, -170, -180, -183 and -187), HCB and dieldrin for OCPs, and PFHxA, PFHpA, PFOA, PFNA, PFTrDA and PFTeDA for PFASs. Concentrations in blanks, calculated with the extracted sample masses, were 0.22 - 3.2 pg g^{-1} ww for PCBs, 1.9 - 26.8 pg g^{-1} ww for OCPs and 1.4 - 37.7 pg g^{-1} ww for PFASs. Concentrations in samples were corrected using the blank value of each sample batch. The recovery (n = 49) of each labelled compound ranged from 67 ± 8% (CB-52) to 85 ± 16% (CB-167) for PCBs, 42 ± 14%

186 (α-endosulfan) to 115 ± 22% (dieldrin) for OCPs and 38 ± 10% (PFTeDA) to 85 ± 10%
187 (PFHxA) for PFASs.
188 Limits of quantification (LOQs) were calculated for each target compound in each sample
189 using a signal-to-noise ratio of 3 (peak-to-peak) on the less intensive raw data signal
190 (qualifier ion) (Wenzl et al., 2016), taking into account injection volume, the volume of
191 concentrated extract prior to injection and extracted sample weight. LOQs ranged from 0.02
192 pg g^{-1} ww to 1.36 pg g^{-1} ww for PCBs, 0.1 pg g^{-1} ww to 27.4 pg g^{-1} ww for OCPs and 0.2 pg g^{-1}
193 ww to 19.0 pg g^{-1} ww for PFASs.

194 2.5. Statistical analysis

195 In view of the small number of studied individuals and non-homogeneous nature of dataset
196 variances, statistical analyses were conducted using non-parametric tests. Comparisons of
197 mean contaminant concentrations between species and between sexes of each species
198 were conducted using the Wilcoxon-Mann-Whitney test. The correlation between biological
199 parameters (size, mass, % lipids and K index) and organic contaminant concentrations was
200 assessed with the Spearman rank test. POP profiles were investigated using the Mann-
201 Whitney test and a standardized Principal Component Analysis (PCA) was additionally
202 performed using Euclidean distances for PCBs.
203 The data were analysed with R software (version 3.4.4). Mean comparisons and correlation
204 tests were conducted with the stat and Hmisc packages (Hollander et al., 2013). PCAs were
205 performed using the FactorMiner package (Husson et al., 2017).

206 **3. Results and discussion**

207 3.1. Biological parameters
208 Free swimming bull sharks ranged from 82 cm to 327 cm in size; tiger sharks ranged from
209 157 cm to 387 cm in size (Table 1). Embryos pertaining to each species were similar in size /
210 mass, but significant differences were found between the two species, i.e. 33 ± 1 cm / 0.12 ±

211 0.01 kg and 63 ± 1 cm / 1.7 ± 0.1 kg in tiger and bull sharks, respectively. Embryo sizes
212 corresponded to embryo ages of ca. 6 months and 10 months for tiger and bull sharks,
213 respectively (Pirog et al., 2019a, 2020; Whitney and Crow, 2007), i.e. at the third of total
214 gestation time for tiger shark (Pirog et al., 2020) and at the end of gestation for bull shark
215 (Pirog et al., 2019).
216 As no significant differences were found in any of the four biological parameters (T_L, W, K,
217 TLC) across sexes from either species, both sexes were considered together for
218 morphometric and lipid content comparison between species. Tiger sharks showed a similar
219 average total length and body mass to bull sharks. However, tiger shark maximum size (387
220 cm versus 327 cm) and mass (402 kg versus 299 kg) exceeded that of bull sharks. Fulton's
221 condition factor mean value was 0.7 times lower in tiger sharks versus bull sharks ($p = 5.10^{-7}$),
222 while TLCs were not significantly different between both species. K is often used as a
223 global indicator of habitat quality or food availability and a positive correlation with lipid
224 content has previously been described in juvenile white sharks (*Carcharodon carcharias*)
225 (Logan et al., 2018). The absence of correlation between K and TLCs in the studied species
226 (tiger sharks: $p = 0.45$; bull sharks: $p = 0.12$) may be due to age (large juveniles and adults)
227 or differences in the physiology and dynamics of lipid storage in the two species.
228 Fish, cephalopods, birds and unidentified remains were found in the non-empty stomachs of
229 bull sharks (64%) and tiger sharks (45%). These observations are coherent with data
230 previously reported by Trystram et al. (2017) and confirm a dominant piscivorous dietary
231 habit for both species, with tiger sharks exhibiting a more generalist foraging behaviour than
232 bull sharks.

233 3.2. Influence of biological parameters on organic contaminant bioaccumulation

234 Concentrations of the various families of studied organic contaminants in tiger and bull
235 sharks are presented in Table 2. Detailed results per individual compound are given in Table
236 S1.

237 The highest contaminant concentrations were found in tiger shark embryos, with the
238 exception of PFOS. In adults, OCPs were predominant in tiger sharks, followed by PCBs and
239 PFASs, while in bull sharks, concentrations ranked in the following order PCBs > OCPs >
240 PFASs. All concentrations were low in the young-of-the-year bull shark.
241

242 3.2.1. Chlorinated POPs

243 TLCs were not correlated with concentrations of the different contaminant families in either
244 species (whether only mature individuals were considered or not), indicating that TLC had a
245 limited influence on chlorinated POP bioaccumulation in white muscle. Chlorinated POP
246 concentrations were therefore primarily compared on a ww basis.
247 Each PCB congener was quantified in over 60% of shark samples, with the exception of CB-
248 31 and -209, which were detected in 45% of bull shark samples and CB-28, -31, -44 and -49,
249 which were detected in 15%, 15%, 40% and 40% of tiger shark samples, respectively.
250 Among OCPs, the 5 isomers of DDT, mirex and dieldrin were detected in over 90% of
251 samples from both species; endrin was detected in 50% and 75% of bull shark and tiger
252 shark samples, respectively. Contamination levels in adults ranged from 104 to 9885 pg g^{-1}
253 ww for ∑ 30 PCBs, 50 to 2473 pg g^{-1} ww for ∑ DDTs, 3 to 1562 pg g^{-1} ww for mirex and 6 to
254 49 pg g^{-1} ww for the sum of dieldrin-endrine (Figure 1). A high inter-individual variability in
255 contamination levels was observed in both species probably due to the generalist foraging
256 behaviour of the two species (Trystram et al. 2017) that lead to a high variety of ingested
257 prey with potential different degrees of contamination. Besides, a wide range of sizes / ages
258 were considered, hence enhancing contamination variability. Organochlorinated compound
259 mean concentrations, on a ww basis, were 3 times higher in male than in female bull sharks
260 (Figure 1), although differences were not statistically significant for PCBs ($p = 0.03$ and 0.06
261 for OCPs and PCBs respectively) due to the high variability of contamination levels across
262 individuals (OCP concentrations were 1455.5 ± 1359.7 pg g^{-1} ww in males and 419.6 ± 388.5
263 pg g^{-1} ww in females; PCB concentrations were 4602.7 ± 4146.1 pg g^{-1} ww in males and

1502.8 ± 1331.7 pg g^{-1} ww in females). When normalized to lipid content, these concentrations were significantly higher in males for both OCPs and PCBs ($p = 0.006$ for OCPs and $p = 0.024$ for PCBs): 243.7 ± 189.9 ng g^{-1} lw versus 68.4 ± 59.3 ng g^{-1} lw in females for OCPs and 745.7 ± 606.0 ng g^{-1} lw in males versus 261.9 ± 240.0 ng g^{-1} in females for PCBs. Conversely, PCB and OCP concentrations were similar ($p = 0.35$) in both sexes in tiger sharks: OCP concentrations were 756.6 ± 756.5 pg g^{-1} ww in males and 1268.4 ± 1066.1pg g^{-1} ww in females; PCB concentrations were 298.6 ± 155.0 pg g^{-1} ww in males and 375.4 ± 348.1 pg g^{-1} ww in females. When normalized to lipid content, these concentrations were 114.7 ± 105.6 ng g^{-1} lw in males versus 208.0 ± 173.5 ng g^{-1} lw in females for OCPs and 43.4 ± 17.3.0 ng g^{-1} lw in males versus 59.0 ± 49.0 ng g^{-1} lw in females for PCBs ($p = 0.17$ and $p = 0.76$ respectively). Differences in contamination levels between sexes may be caused by various factors such as reproductive loss, growth rate or diet (Larsson et al., 1993; Madenjian et al., 2010; Ng and Gray, 2009; Rypel et al., 2007). No sex-related dietary differences have been reported between the two species (Trystram et al., 2017); moreover, males have a higher growth rate than females (Cruz-Martínez et al., 2004; Kneebone et al., 2008). Neither of these factors would lead to the differences observed in contaminant accumulation in male and female bull sharks. The lack of sex-related influences on POP contamination in tiger sharks could indicate that reproduction has less impact on hydrophobic pollutant concentrations in ovoviviparous female tiger sharks versus viviparous female bull sharks. The results suggest that the ovoviviparous mode of gestation induces less decontamination in female shark muscle than viviparity. Actually, the continuous supply of nutrients during bull shark gestation could result in higher maternal contaminant offloading in comparison to egg-forming species such as tiger sharks (Castro et al., 2016). However, further investigations would be needed in order to confirm this hypothesis.

No significant differences in DDT concentrations were found between the two species (bull shark: 584 ± 648 pg g^{-1} ww; tiger shark: 585 ± 580 pg g^{-1} ww, $p = 0.9$), while PCB mean concentrations were 7 times higher in bull sharks (2597 ± 2968 pg g^{-1} ww) than in tiger sharks (339 ± 270 pg g^{-1} ww; $p = 10^{-6}$). The higher PCB contamination levels found in bull

292 sharks, combined with their more coastal habitat in RUN (Trystram et al., 2017), indicates
293 exposure of these coastal environments to more significant PCB sources. Conversely, the
294 similar levels of DDT accumulation found in both species suggests similar sources in both
295 coastal (bull sharks) and offshore (tiger sharks) feeding habitats. Among OCPs, mirex
296 concentrations were 2.5 times higher in tiger sharks than in bull sharks (tiger shark: 403 ±
297 431 pg g^{-1} ww; bull shark: 180 ± 334 pg g^{-1} ww, $p = 0.028$), while dieldrin concentrations were
298 1.5 times higher in tiger sharks than in bull sharks (tiger shark: 36 ± 14 pg g^{-1} ww; bull shark:
299 24 ± 14 pg g^{-1} ww, $p = 0.016$). As tiger sharks migrate long distances across the Indian
300 Ocean (Pirog et al., 2019a), these differences suggest that certain insecticides, such as
301 mirex and dieldrin, may be subject to different patterns of use on the Indian Ocean scale,
302 despite the fact that these compounds have been banned in most countries. Although mirex
303 has been detected in air and various matrices in the Indian Ocean, no specific source has
304 been clearly identified (Bouwman et al., 2012; Qiu et al., 2020; Srimurali et al., 2015), while
305 dieldrin may still be in use in Kenya (Barasa et al., 2008).
306 Only PCBs were significantly correlated to individual size in bull sharks ($p = 0.038$), while
307 PCB, DDT and mirex concentrations showed positive correlations with tiger shark size ($p =$
308 0.0081, 0.00016 and 0.001 respectively). Contaminant accumulation over fish lifetime
309 reflects a combination of various factors, such as fish growth rate, contaminant accumulation
310 and elimination rates, and reproduction. In sharks, previously-reported results showed
311 species-related relationships between contaminant accumulation and size (Lyons et al.,
312 2019).

313 3.2.2. PFASs

314 Among the analysed PFASs, PFUnDA, PFTrDA and PFTeDA were more frequently detected
315 in both species (95 - 100% of the samples), followed by PFDoDA (79%), PFNA (69%) and
316 PFDA (59%). PFOS was above LOQs in 83% of bull sharks and only 24% of tiger sharks due
317 to the low concentrations encountered. Other PFASs were below LOQs in all samples.
318 Overall, PFOS concentrations above LOQs were in the 6.0 - 148.0 pg g^{-1} ww range (mean of

319 31.9 ± 34.1 pg g^{-1} ww, one male bull shark exhibiting a higher concentration of 555 pg g^{-1} ww
320 was excluded), while ∑ PFCA concentrations were in the 69.0 - 353.5 pg g^{-1} ww range, with
321 mean concentrations of 135.0 ± 38.2 pg g^{-1} ww in tiger sharks and 212.0 ± 97.8 pg g^{-1} ww in
322 bull sharks.
323 PFOS and ∑ PFCA concentrations were not statistically different between sexes in either
324 species ($p = 0.12$ and 0.24 respectively for bull shark; PFOS not tested for tiger sharks and p
325 $= 0.39$ for ∑ PFCAs). Unlike organochlorine compounds, PFASs did not show higher
326 concentrations in male bull sharks ($p = 0.32$); this could indicate that female contamination by
327 these compounds is less influenced by reproduction and mode of gestation versus PCBs and
328 OCPs. PFAS concentrations were significantly higher in bull sharks versus tiger sharks and
329 differences were significant for each individual compound ($p = 0.00051$ to 0.02), with the
330 exception of PFNA and PFTrDA ($p = 0.051$ and 1 respectively). The higher PFAS levels
331 found in bull sharks suggest that their sources may be local rather than distant. High inter-
332 individual variability was observed (a factor of 5 between minimum and maximum PFAS
333 concentrations), as expected in view of the wide range of sizes / ages. However, no
334 significant correlation was found between PFAS concentrations and fish sizes in the two
335 species.

336 3.3. Contaminant profiles and ratios as tracers of contamination sources

337 No differences in PCB, DDT and PFAS profiles were found between sexes in either species,
338 suggesting that they share similar trophic habitats, as hypothesised by Trystram et al.,
339 (2017). Both sexes were thus examined together.

340 3.3.1. PCBs

341 PCBs can be classified according to the number and position of chlorine atoms, which affect
342 their hydrophobicity (Hawker and Connell, 1988) and propensity to be metabolized. Chlorine
343 atom position defines five structure-activity groups (SAGs), characterizing congener
344 metabolization capability (Boon et al., 1997; Yunker et al., 2011). Groups I and II are not

345 metabolized, group III is metabolized in some mammals but not in fish, and groups IV and V
346 are metabolized in some fish (Buckman et al., 2007).
347 To assess contamination profiles, a standard PCA (Figure 2) was performed on all samples
348 except the young-of-the-year bull shark, which was included for information purposes only
349 (additional individual). In this analysis, the two first components representing 90.9% of total
350 variance were used according to the Kaiser criterion (component eigenvalue > 1, Figure 2).
351 Additional variables were groups poorly-represented in the factorial plan of the first two
352 components (inertia of the variable in a component < mean inertia of that component).
353 PCB congeners predominating in bull sharks were higher chlorinated (7 - 8 chlorine atoms)
354 and non-metabolizable (SAG I), while tiger sharks were characterized by a higher variability
355 in their profiles (Figure 2), corresponding to a higher diversity of tiger shark prey in RUN
356 (Trystram et al., 2017). Univariate analyses confirmed that the relative abundance of these
357 compounds differed significantly between the two species ($p = 0.00064$ and $p = 0.012$ for 7
358 and 8 chlorine PCBs respectively). The PCB contamination profile of the young-of-the-year
359 bull shark was completely different to that of the other bull shark individuals.
360 PCBs exhibited a contamination profile dominated by higher chlorinated and non-
361 metabolizable congeners in bull sharks. As the heavier congeners are less mobile and bind
362 to organic matter near the coast, leading to preferential accumulation of these compounds in
363 food webs close to their sources (Salvadó et al., 2013), bull sharks would appear to be more
364 highly-impacted by local, coastal PCB sources. This surprising result, in view of the island's
365 low level of industrialization, is corroborated by the study of Dirtu et al. (2016), which showed
366 higher PCB levels in coastal Indo-Pacific bottlenose dolphins (*Tursiops aduncus*) than in
367 spinner dolphins (*Stenella longirostris)* living further offshore in RUN. Conversely, tiger
368 sharks were predominantly impacted by lower chlorinated PCBs, which can be transported
369 further offshore.

3.3.2. DDTs

DDT profiles were dominated by p,p'-DDE followed by o,p'-DDT in tiger sharks and by p,p'-DDT in bull sharks (Figure 3). Bull sharks showed significantly higher p,p'-DDE contributions than tiger sharks (97 ± 3% > 92 ± 5%; $p = 9.4\ 10^{-6}$).

The relative proportions of *p,p'*-DDE were higher than those reported in other high trophic level predators in RUN by Munschy et al. (2016) and Dirtu et al. (2016). The ratio of (*p,p'*-DDE + *p,p'*-DDD) / ∑ DDTs concentrations, which is an indicator of *p,p'*-DDT degradation in the environment, was not significantly different in the two species (0.95 ± 0.04 on average, p = 0.88) and indicated that DDT sources in the studied sharks were old (Suárez et al., 2013).

The *o,p'*-DDT / *p,p'*-DDT concentration ratio, providing information on the source of DDT contamination, showed values above 0.34 in both species, suggesting that dicofol may be a contributor to contamination (Suárez et al., 2013). This ratio was significantly higher in tiger sharks than in bull sharks (2.19 ± 2.17 > 0.55 ± 0.52; p = 0.00034), possibly indicating a dicofol source outside RUN contaminating the Indian Ocean.

3.3.3. OCP / PCB concentration ratios

Various organic contaminant ratios, when used in combination with data on trophic habitats, can be used to trace the trophic origin of contamination. The ∑ OCP / ∑ PCB concentration ratio has been commonly-used in various species to provide an initial evaluation of industrial or agricultural sources (Munschy et al., 2016; Storelli et al., 2006; Suárez et al., 2013). This ratio differed significantly in the two species, i.e. 0.37 ± 0.24 in bull sharks and 2.93 ± 1.96 ($p= 6.3\ 10^{-10}$) in tiger sharks, indicating that contamination profiles were dominated by pollutants of different origin (industrial in bull sharks, pesticides in tiger sharks). Genetic population and trophic ecology studies in RUN have shown that tiger sharks have an offshore habitat and can navigate throughout the Indian Ocean basin (Pirog et al., 2019b; Trystram et al., 2017). Conversely, bull sharks live in coastal environments, although some individuals engage in wide scale movements in the Southwest Indian Ocean (Daly et al.,

2014; Lea et al., 2015; Pirog et al., 2019c; Trystram et al., 2017). In addition, the two species feed on different prey in RUN. These information on the trophic habits of the two species suggest that the coastal prey of bull sharks could be more highly-impacted by PCBs, although the sources and the physicochemical dynamics of these pollutants in the tissues of the two species could be an alternative explanation to the observed differences. Concentration ratios were positively correlated with tiger shark size (rho = 0.60; p = 0.0073) suggesting changes in contamination sources according to age. Similarly, this could either be due to movements throughout the Indian Ocean or to physiology-related changes.

3.3.4. PFASs

Concentrations of ∑PFCAs, including long-chain compounds (C ≥ 8), were higher than PFOS concentrations (on average by a factor of 11 and 17 in bull and tiger sharks, respectively) in all samples (the above-mentioned male bull shark outlier excluded), consistent with the higher bioaccumulative abilities of these compounds versus their short-chain counterparts (Kelly et al., 2009; Martin et al., 2003; Pan et al., 2014). More specifically, PFCA profiles showed a predominance of odd-chain length compounds (PFTrDA and PFUnDA) versus even-chain (PFDA, PFDoDA and PFTeDA) (Figure 4), as previously observed in fish (Martin et al., 2003; Sturm and Ahrens, 2010), including in the Indian Ocean (Munschy et al., 2020). Their presence has been partially explained by the degradation of fluorotelomer alcohols (FTOHs) (Ellis et al., 2004; Martin et al., 2004). PFUnDA and other long-chain PFCAs could originate from the degradation of 10:2 FTOH (Hart et al., 2008). Odd-chain PFCAs such as PFUnDA and PFTrDA have also been reported as impurities in PFNA resulting from the oxidation of fluorotelomer olefins (Prevedouros et al., 2006; Rotander et al., 2012).

Tiger shark and bull shark long-chain PFCA profiles differed, with higher proportions of odd-chain PFCAs in tiger sharks (80 ± 9%) versus bull sharks (66 ± 13%; p = 0.00094), suggesting that dietary differences could result in different contamination patterns.

3.4. Maternal transfer

3.4.1. Contamination levels

In both species, organic contaminant concentrations showed low variability between embryo pools (6 - 19% relative standard deviation for the various contaminant families within each species), consistent with the fact that they originated from the same female, were of the same age and were all exposed to similar contaminant sources (ie, yolk sac or placental bond), with no external exposure.

In bull shark embryos, concentrations (in ww) were 4 to 25 times lower than those of their mother for all chlorinated compounds except dieldrin (ratio mother / embryos of 1) (Table 3). When normalized to TLC, embryo contaminations were 6 to 35 times lower than those of their mother. Among perfluorinated compounds, PFCAs showed higher concentrations in the mother, while PFOS was 3 times more concentrated in embryos. These results demonstrate that lipophilic and amphiphilic molecules are not transmitted to embryos in the same manner, indicating that molecule physico-chemical properties and affinity with major biological macromolecules are determining parameters in the maternal transfer of organic contaminants in bull sharks. Conversely, concentrations of all chlorinated compounds and PFCAs expressed in ww (Table 3) were 2 to 7 times (and 3 times in lw) higher in tiger shark embryos than in the mother, while PFOS was not detected in the mother and in 33% of embryos only. In tiger sharks, embryonic development is ensured by a yolk sac and a high energy uterine fluid (named the embryotrophe), as no placental connection exists (Castro et al., 2016). The differences in modes of gestation and mean TLCs in embryos between the two species (TLCs in tiger shark embryos were 2 times higher than in bull shark embryos ($p < 0.001$)) could partly explain the fact that tiger shark embryos were exposed to higher levels of lipophilic contaminants than bull shark embryos. However, further investigations of embryos of both species at different stages of gestation would be necessary as both embryos differed in their embryonic stage.

448 In bull sharks, a significant negative linear regression between the octanol / water partition
449 coefficient (log $K_{OW;}$ Hawker and Connell, 1988; Shen and Wania, 2005) and log10-
450 transformed ratio between the average concentration of organochlorinated compounds in
451 embryos versus the mother (i.e. the partition ratio), both normalized to lipid content, was
452 observed (r = -0.85, p = 0.0005, Figure 5A). Conversely, no significant relationship was found
453 in tiger sharks (Figure 5B). These results show that the maternal transfer of
454 organochlorinated compounds in viviparous sharks depends on molecule hydrophobicity. As
455 shown previously by Lyons and Adams (2015) in another placental shark species, our results
456 show that the most hydrophobic molecules were less transferred to the muscle of bull shark
457 embryos than the low hydrophobic ones. On the opposite, tiger shark embryo contamination
458 relative to their mother was not dependent on the molecule hydrophobicity (Figure 5B).
459 PFOS showed a higher partition ratio than PFCAs (0.45 for PFOS versus -0.17 ± 0.08 for
460 PFCAs) and PFCA partition ratios were negatively-correlated with the number of carbon
461 atoms (rho = -0.9; p = 0.016) in bull sharks. All PFASs were detected in tiger shark embryos,
462 while in the mother, only PFUnDA and PFTrDA (partition ratio 1.29 and 1.19, respectively)
463 were detected, suggesting that PFASs were efficiently transferred to offspring. The
464 differences observed between the two species could be related to the biochemical
465 composition of tiger shark eggs versus exchanges driven by the placental bond of bull
466 sharks.
467

468 3.4.2. Contamination profiles

469 PCB profiles were similar in the tiger shark mother and embryos (Figure 2), while higher
470 relative concentrations of *p,p'*-DDE were observed in the mother (mother: 95%; embryos: 86
471 ± 0.5 %; Figure 6A). Conversely, hexachlorinated congeners belonging to SAG III were less-
472 prevalent in the bull shark mother than in embryos (Figure 2), while similar DDT profiles were
473 observed (Figure 6A). The (*p,p'*-DDE + *p,p'*-DDD) / *p,p'*-DDT ratio was respectively 2 and 4
474 times higher in bull and tiger shark mothers than in their embryos, suggesting that DDT

475 transfer and accumulation in embryo muscle was isomer-specific. Similar PFAS profiles were
476 found in the bull shark mother and embryos, with the exception of PFOS, which showed
477 higher relative concentrations in embryos (9% in the mother and 31 ± 4% in her embryos;
478 Figure 6B). Only PFUnDA and PFTrDA were detected in the tiger shark mother, whereas
479 PFASs were detected in 100% of embryos, with the exception of PFOS and PFDoDA
480 detected in 33% and 83% of embryos, respectively. Our results show that the different
481 molecule families have different fates in the two studied species, suggesting that pollutant
482 family and mode of gestation (known to be different in the two species) are important factors
483 in the maternal transfer of organic contaminants.

484 3.5. Organic contaminant levels in sharks from Reunion Island versus worldwide levels

485 Large inter-individual variations in organic contaminant concentrations in sharks have been
486 found in previous studies worldwide (Table S2 and S3). Various factors, such as size, sex
487 and lipid levels of studied individuals, which are not always reported, the number of
488 compounds considered, the decrease in legacy POP concentrations and potential increase in
489 CECs over time (Tanabe and Ramu, 2012) could explain these variations.
490 Sharks sampled in various locations in the Southern Hemisphere have shown PCB
491 concentrations between 2 and 100 times higher than sharks from RUN (Cagnazzi et al.,
492 2019; Marsili et al., 2016). A pregnant bull shark and blacktip reef sharks (*Carcharhinus*
493 *melanopterus*) sampled in Australia and on the east coast of South Africa showed
494 concentrations similar to those of RUN bull sharks, but 10 times lower than those of tigers
495 sharks (Beaudry, 2014; Cagnazzi et al., 2019). PCB and DDT concentrations reported in
496 another top predator species (albacore tuna *Thunnus alalunga*) from the coast of South
497 Africa were lower than those found in RUN due to higher industrialization and urbanization in
498 South Africa (Dirtu et al., 2016; Munschy et al., 2016; Mwevura et al., 2010). DDT
499 concentrations were 5 to 100 times higher in sharks from the Southern Hemisphere oceans
500 than RUN (Beaudry, 2014; Cagnazzi et al., 2019; Marsili et al., 2016; Schlenk et al., 2005).
501 POP concentrations were therefore globally lower in individuals sampled in RUN versus

other locations in the Southern Hemisphere, probably associated with the island's low urbanization and industrialization and its remote oceanic position far from landmasses.

High industrialization and urbanization in many areas of the Northern Hemisphere have fostered major sources of POPs, leading in turn to higher concentrations in Northern Hemisphere biota versus the Southern Hemisphere (Aguilar et al., 2002). Indeed, this is clearly reflected in PCBs (Table S2) and, to a lesser extent, in DDTs (Table S3), indicating that these banned compounds are still used in some countries in the Southern Hemisphere, probably for mosquito control (van den Berg et al., 2017). Sharks caught in the Northern Hemisphere showed 10 to 1000 times higher PCB concentrations than sharks caught in RUN, with the exception of individuals caught near Portugal and in the Northeast Pacific Ocean, which showed similar concentrations to bull sharks from RUN (Alves et al., 2016; Boldrocchi et al., 2020; Cullen et al., 2019; Johnson-Restrepo et al., 2005; Lee et al., 2015; Storelli et al., 2003). Mean DDT concentrations in RUN sharks were similar to those in various shark species caught in the Northeast Pacific Ocean, with the exception of oceanic whitetip sharks (*Carcharhinus longimanus*), characterised by 100-fold lower DDT concentrations (Table S3; Lee et al., 2015). In contrast, RUN sharks showed 100 times lower DDT concentrations than those of whale sharks (*Rhincodon typus*) from the Red Sea, associated with the contemporary use of DDT in Djibouti (Boldrocchi et al., 2020), indicating that DDT concentrations are strongly influenced by local sources and can therefore vary greatly from one ocean basin to another. Mirex concentrations in the muscle of bonnethead sharks (*Sphyrna tiburo*) from Florida were below LOQ (Gelsleichter et al., 2005), whereas this compound was quantified in 100% of samples in our study. The mirex ban and absence of this pesticide in Florida, where it has been widely used in the past (Alley, 1973), suggest recent regional use of this compound in Southern Indian Ocean, possibly to control invasive ants (Blard, 2006; Delabie and Blard, 2002). In the same study, dieldrin concentrations were found to be 10 times higher than in tiger and bull sharks from RUN.

Regarding PFASs, little data are available on the accumulation of these emerging pollutants in sharks. Alves et al. (2016) studied blue sharks in Portugal (*Prionace glauca*) and recorded

PFCAs and PFOS concentrations 5 times higher than those in sharks from RUN. However, concentrations can be expected to be higher in the Northern Hemisphere, similarly to those of other POPs from industrial sources, such as PCBs. PFASs are more readily-transported in oceans than POPs and are hence likely to be more globally-dispersed throughout the oceans, resulting in smaller differences between hemispheres versus PCBs (Prevedouros et al., 2006).

4. Conclusion

This study provides the first data on the contamination of two shark species collected along the West coast of Reunion Island. Our results showed that POP contamination levels in RUN were lower overall than worldwide levels. PCB and DDT contamination levels were below environmental thresholds, except in bull sharks, which exhibited concentrations of four PCB congeners similar to environmental thresholds.

Contamination profiles differed between the two species, indicating, as reported in previous trophic ecology studies, that these two sympatric and opportunistic top predators do not exploit the same trophic niches.

Results suggest the presence of PCB sources in Reunion Island, leading to higher contamination levels in bull sharks than in tiger sharks. Tiger sharks were mainly contaminated by OCPs, which are distributed more globally throughout the Indian Ocean, particularly in offshore ecosystems. Contaminant transfer from mother to embryos was species- and contaminant-dependant, suggesting that organic contaminant transfer to offspring is driven by mode of gestation and molecule physico-chemical properties.

In view of the limited data available on POPs and CECs in Reunion Island coastal ecosystems, a study on their accumulation in tiger and bull shark preys would be necessary to gain a better understanding of accumulation dynamics. In addition, more investigations would be needed in order to fully understand the maternal transfer of organic contaminants, including CECs, in these species.

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References

Aguilar, A., Borrell, A., Reijnders, P.J.H., 2002. Geographical and temporal variation in levels of organochlorine contaminants in marine mammals. Mar. Environ. Res. 53, 425–452. https://doi.org/10.1016/S0141-1136(01)00128-3

Alley, E.G., 1973. The Use of Mirex in Control of the Imported Fire Ant 1. J. Environ. Qual. 2, 52–61. https://doi.org/10.2134/jeq1973.00472425000200010005x

Alves, L.M.F., Nunes, M., Marchand, P., Le Bizec, B., Mendes, S., Correia, J.P.S., Lemos, M.F.L., Novais, S.C., 2016. Blue sharks (*Prionace glauca*) as bioindicators of pollution and health in the Atlantic Ocean: Contamination levels and biochemical stress responses. Sci. Total Environ. 563–564, 282–292. https://doi.org/10.1016/j.scitotenv.2016.04.085

Barasa, M.W., Lalah, J.O., Wandiga, S.O., 2008. Seasonal variability of persistent organochlorine pesticide residues in marine fish along the Indian Ocean coast of Kenya. Toxicol. Environ. Chem. 90, 535–547. https://doi.org/10.1080/02772240701556312

Beaudry, M.C., 2014. Comparative analysis of organochlorine accumulation in two top predator shark species with contrasting life history traits. University of Windsor. Electronic Theses and Dissertations. 5154. https://scholar.uwindsor.ca/etd/5154.

Beyer, A., Mackay, D., Matthies, M., Wania, F., Webster, E., 2000. Assessing Long-Range Transport Potential of Persistent Organic Pollutants. Environ. Sci. Technol. 34, 699–703. https://doi.org/10.1021/es990207w

Blard, F., 2006. Les fourmis envahissantes de l'île de la Réunion : interactions compétitives et facteurs d'invasion. Ph D thesis, Université de la Réunion, 97p.

Boldrocchi, G., Monticelli, D., Butti, L., Omar, M., Bettinetti, R., 2020. First concurrent assessment of elemental- and organic-contaminant loads in skin biopsies of whale sharks from Djibouti. Sci. Total Environ. 722, 137841. https://doi.org/10.1016/j.scitotenv.2020.137841

593 Boon, J.P., Meer, J. van der, Allchin, C.R., Law, R.J., Klungsøyr, J., Leonards, P.E.G., Spliid,
594 H., Storr-Hansen, E., Mckenzie, C., Wells, D.E., 1997. Concentration-Dependent
595 Changes of PCB Patterns in Fish-Eating Mammals: Structural Evidence for Induction
596 of Cytochrome P450. Arch. Environ. Contam. Toxicol. 33, 298–311.
597 https://doi.org/10.1007/s002449900257
598 Borgå, K., Fisk, A.T., Hoekstra, P.F., Muir, D.C.G., 2004. Biological and chemical factors of
599 importance in the bioaccumulation and trophic transfer of persistent organochlorine
600 contaminants in arctic marine food webs. Environ. Toxicol. Chem. 23, 2367–2385.
601 https://doi.org/10.1897/03-518
602 Bouwman, H., Kylin, H., Choong Kwet Yive, N.S., Tatayah, V., Løken, K., Utne Skaare, J.,
603 Polder, A., 2012. First report of chlorinated and brominated hydrocarbon pollutants in
604 marine bird eggs from an oceanic Indian Ocean island. Environ. Res. 118, 53–64.
605 https://doi.org/10.1016/j.envres.2012.05.009
606 Breivik, K., Gioia, R., Chakraborty, P., Zhang, G., Jones, K.C., 2011. Are Reductions in
607 Industrial Organic Contaminants Emissions in Rich Countries Achieved Partly by
608 Export of Toxic Wastes? Environ. Sci. Technol. 45, 9154–9160.
609 https://doi.org/10.1021/es202320c
610 Brown, T.N., Wania, F., 2008. Screening Chemicals for the Potential to be Persistent Organic
611 Pollutants: A Case Study of Arctic Contaminants. Environ. Sci. Technol. 42, 5202–
612 5209. https://doi.org/10.1021/es8004514
613 Buckman, A.H., Brown, S.B., Small, J., Muir, D.C.G., Parrott, J., Solomon, K.R., Fisk, A.T.,
614 2007. Role of Temperature and Enzyme Induction in the Biotransformation of
615 Polychlorinated Biphenyls and Bioformation of Hydroxylated Polychlorinated
616 Biphenyls by Rainbow Trout (*Oncorhynchus mykiss*). Environ. Sci. Technol. 41,
617 3856–3863. https://doi.org/10.1021/es062437y
618 Cagnazzi, D., Consales, G., Broadhurst, M.K., Marsili, L., 2019. Bioaccumulation of
619 organochlorine compounds in large, threatened elasmobranchs off northern New

South Wales, Australia. Mar. Pollut. Bull. 139, 263–269. https://doi.org/10.1016/j.marpolbul.2018.12.043

Castro, J.I., Sato, K., Bodine, A.B., 2016. A novel mode of embryonic nutrition in the tiger shark, *Galeocerdo cuvier*. Marine Biology Research 12, 200–205. https://doi.org/10.1080/17451000.2015.1099677

Cliff, G., Dudley, S.F.J., 1991. Sharks caught in the protective gill nets off Natal, South Africa. 4. The bull shark *Carcharhinus leucas* Valenciennes. South Afr. J. Mar. Sci. 10, 253–270. https://doi.org/10.2989/02577619109504636

Compagno, L.J.V., 1984. FAO species catalogue. Sharks of the world. An annotated and illustrated catalogue of shark species known to date. II. Carcharhiniformes. 382p

Corsolini, S., Ancora, S., Bianchi, N., Mariotti, G., Leonzio, C., Christiansen, J.S., 2014. Organotropism of persistent organic pollutants and heavy metals in the Greenland shark *Somniosus microcephalus* in NE Greenland. Mar. Pollut. Bull. 87, 381–387. https://doi.org/10.1016/j.marpolbul.2014.07.021

Corsolini, S., Pozo, K., Christiansen, J.S., 2016. Legacy and emergent POPs in the marine fauna of NE Greenland with special emphasis on the Greenland shark *Somniosus microcephalus*. Rendiconti Lincei 27, 201–206. https://doi.org/10.1007/s12210-016-0541-7

Cruz-Martínez, A., Chiappa-Carrara, X., Arenas-Fuentes, V., 2004. Age and Growth of the Bull Shark, *Carcharhinus leucas* , from Southern Gulf of Mexico. J. Northwest Atl. Fish. Sci. 35, 367–374. https://doi.org/10.2960/J.v35.m481

Cullen, J.A., Marshall, C.D., Hala, D., 2019. Integration of multi-tissue PAH and PCB burdens with biomarker activity in three coastal shark species from the northwestern Gulf of Mexico. Sci. Total Environ. 650, 1158–1172. https://doi.org/10.1016/j.scitotenv.2018.09.128

Dachs, J., Lohmann, R., Ockenden, W.A., Méjanelle, L., Eisenreich, S.J., Jones, K.C., 2002. Oceanic Biogeochemical Controls on Global Dynamics of Persistent Organic Pollutants. Environ. Sci. Technol. 36, 4229–4237. https://doi.org/10.1021/es025724k

Daly, R., Smale, M.J., Cowley, P.D., Froneman, P.W., 2014. Residency Patterns and Migration Dynamics of Adult Bull Sharks (*Carcharhinus leucas*) on the East Coast of Southern Africa. PLOS ONE 9, e109357. https://doi.org/10.1371/journal.pone.0109357

Delabie, J.H.C., Blard, F., 2002. The tramp ant Hypoponera punctatissima (Roger) (Hymenoptera: Formicidae: Ponerinae): new records from the southern hemisphere. Neotrop. Entomol. 31, 149–151. https://doi.org/10.1590/S1519-566X2002000100020

Dicken, M.L., Hussey, N.E., Christiansen, H.M., Smale, M.J., Nkabi, N., Cliff, G., Wintner, S.P., 2017. Diet and trophic ecology of the tiger shark (*Galeocerdo cuvier*) from South African waters. PLOS ONE 12, e0177897. https://doi.org/10.1371/journal.pone.0177897

Dirtu, A.C., Malarvannan, G., Das, K., Dulau-Drouot, V., Kiszka, J.J., Lepoint, G., Mongin, P., Covaci, A., 2016. Contrasted accumulation patterns of persistent organic pollutants and mercury in sympatric tropical dolphins from the south-western Indian Ocean. Environ. Res. 146, 263–273. https://doi.org/10.1016/j.envres.2016.01.006

Dulvy, N.K., Simpfendorfer, C.A., Davidson, L.N.K., Fordham, S.V., Bräutigam, A., Sant, G., Welch, D.J., 2017. Challenges and Priorities in Shark and Ray Conservation. Curr. Biol. 27, R565–R572. https://doi.org/10.1016/j.cub.2017.04.038

Ellis, D.A., Martin, J.W., De Silva, A.O., Mabury, S.A., Hurley, M.D., Sulbaek Andersen, M.P., Wallington, T.J., 2004. Degradation of Fluorotelomer Alcohols: A Likely Atmospheric Source of Perfluorinated Carboxylic Acids. Environ. Sci. Technol. 38, 3316–3321. https://doi.org/10.1021/es049860w

Ferretti, F., Worm, B., Britten, G.L., Heithaus, M.R., Lotze, H.K., 2010. Patterns and ecosystem consequences of shark declines in the ocean. Ecol. Lett. 13, 1055–1071. https://doi.org/10.1111/j.1461-0248.2010.01489.x

Fisk, A.T., Tittlemier, S.A., Pranschke, J.L., Norstrom, R.J., 2002. Using Anthropogenic Contaminants and Stable Isotopes to Assess the Feeding Ecology of Greenland

675 Sharks. Ecology 83, 2162–2172. https://doi.org/10.1890/0012-
676 9658(2002)083[2162:UACASI]2.0.CO;2
677 Fricke, R., Mulochau, T., Durville, P., Chabanet, P., Tessier, E., Letourneur, Y., 2009.
678 Annotated checklist of the fish species (Pisces) of La Réunion, including a Red List of
679 threatened and declining species. Stuttg. Beitr. Zur Naturkunde A 2, 1–168.
680 Gelsleichter, J., Manire, C.A., Szabo, N.J., Cortés, E., Carlson, J., Lombardi-Carlson, L.,
681 2005. Organochlorine Concentrations in Bonnethead Sharks (Sphyrna tiburo) from
682 Four Florida Estuaries. Arch. Environ. Contam. Toxicol. 48, 474–483.
683 https://doi.org/10.1007/s00244-003-0275-2
684 Gilliers, C., Amara, R., Bergeron, J.P., Le Pape, O., 2004. Comparison of growth and
685 condition indices of juvenile flatfish in different coastal nursery grounds. Environ. Biol.
686 Fishes 71, 189-198.
687 Guyomard D., Perry, C., Tournoux P.U., Cliff, G., Peddemors, V., Jaquemet, S. 2019. An
688 innovative fishing gear to enhance the release of non-target species in coastal shark-
689 control programs: The SMART (shark management alert in real-time) drumline. Fish.
690 Res. 216, 6-17. https://doi.org/10.1016/j.fishres.2019.03.011
691 Green, A., Larson, S., 2016. A Review of Organochlorine Contaminants in Nearshore Marine
692 Mammal Predators. J. Environ. Anal. Toxicol. 06. https://doi.org/10.4172/2161-
693 0525.1000370
694 Grimm, F.A., Hu, D., Kania-Korwel, I., Lehmler, H.-J., Ludewig, G., Hornbuckle, K.C., Duffel,
695 M.W., Bergman, Å., Robertson, L.W., 2015. Metabolism and metabolites of
696 polychlorinated biphenyls. Crit. Rev. Toxicol. 45, 245–272.
697 https://doi.org/10.3109/10408444.2014.999365
698 Hart, K., Gill, V.A., Kannan, K., 2008. Temporal Trends (1992–2007) of Perfluorinated
699 Chemicals in Northern Sea Otters (Enhydra lutris kenyoni) from South-Central
700 Alaska. Arch. Environ. Contam. Toxicol. 56, 607. https://doi.org/10.1007/s00244-008-
701 9242-2

Hawker, D.W., Connell, D.W., 1988. Octanol-water partition coefficients of polychlorinated biphenyl congeners. Environ. Sci. Technol. 22, 382–387. https://doi.org/10.1021/es00169a004

Heithaus, M.R., Frid, A., Wirsing, A.J., Worm, B., 2008. Predicting ecological consequences of marine top predator declines. Trends Ecol. Evol. 23, 202–210. https://doi.org/10.1016/j.tree.2008.01.003

Hollander, M., Wolfe, D.A., Chicken, E., 2013. Nonparametric Statistical Methods. John Wiley & Sons.

Hoydal, K.S., Letcher, R.J., Blair, D.A.D., Dam, M., Lockyer, C., Jenssen, B.M., 2015. Legacy and emerging organic pollutants in liver and plasma of long-finned pilot whales (Globicephala melas) from waters surrounding the Faroe Islands. Sci. Total Environ. 520, 270–285. https://doi.org/10.1016/j.scitotenv.2015.03.056

Husson, F., Le, S., Pagès, J., Le, S., Pagès, J., 2017. Exploratory Multivariate Analysis by Example Using R. Chapman and Hall/CRC. https://doi.org/10.1201/b21874

Jepson, P.D., Law, R.J., 2016. Persistent pollutants, persistent threats. Science 352, 1388–1389. https://doi.org/10.1126/science.aaf9075

Johansson, J.H., Salter, M.E., Navarro, J.C.A., Leck, C., Nilsson, E.D., Cousins, I.T., 2019. Global transport of perfluoroalkyl acids via sea spray aerosol. Environ. Sci. Process. Impacts 21, 635–649. https://doi.org/10.1039/C8EM00525G

Johnson-Restrepo, B., Kannan, K., Addink, R., Adams, D.H., 2005. Polybrominated Diphenyl Ethers and Polychlorinated Biphenyls in a Marine Foodweb of Coastal Florida. Environ. Sci. Technol. 39, 8243–8250. https://doi.org/10.1021/es051551y

Kelly, B.C., Ikonomou, M.G., Blair, J.D., Morin, A.E., Gobas, F.A.P.C., 2007. Food Web Specific Biomagnification of Persistent Organic Pollutants. Science 317, 236–239. https://doi.org/10.1126/science.1138275

Kelly, B.C., Ikonomou, M.G., Blair, J.D., Surridge, B., Hoover, D., Grace, R., Gobas, F.A.P.C., 2009. Perfluoroalkyl Contaminants in an Arctic Marine Food Web: Trophic

729 Magnification and Wildlife Exposure. Environ. Sci. Technol. 43, 4037–4043.
730 https://doi.org/10.1021/es9003894
731 Kneebone, J., Natanson, L.J., Andrews, A.H., Howell, W.H., 2008. Using bomb radiocarbon
732 analyses to validate age and growth estimates for the tiger shark, *Galeocerdo cuvier*,
733 in the western North Atlantic. Mar. Biol. 154, 423–434.
734 https://doi.org/10.1007/s00227-008-0934-y
735 Lagabrielle, E., Allibert, A., Kiszka, J.J., Loiseau, N., Kilfoil, J.P., Lemahieu, A., 2018.
736 Environmental and anthropogenic factors affecting the increasing occurrence of
737 shark-human interactions around a fast-developing Indian Ocean island. Sci. Rep. 8,
738 3676. https://doi.org/10.1038/s41598-018-21553-0
739 Larsson, P., Okla, L., Collvin, L., 1993. Reproductive status and lipid content as factors in
740 PCB, DDT and HCH contamination of a population of pike (*Esox lucius* L.). Environ.
741 Toxicol. Chem. 12, 855–861. https://doi.org/10.1002/etc.5620120509
742 Lea, J.S.E., Humphries, N.E., Clarke, C.R., Sims, D.W., 2015. To Madagascar and back:
743 long-distance, return migration across open ocean by a pregnant female bull shark
744 Carcharhinus leucas. J. Fish Biol. 87, 1313–1321. https://doi.org/10.1111/jfb.12805
745 Le Croizier, G., Lorrain, A., Sonke, J.E., Jaquemet, S., Schaal, G., Renedo, M., Besnard, L.,
746 Cherel, Y., Point, D., 2020. Mercury isotopes as tracers of ecology and metabolism in
747 two sympatric shark species. Environmental Pollution 114931.
748 https://doi.org/10.1016/j.envpol.2020.114931
749 Lee, H.-K., Jeong, Y., Lee, S., Jeong, W., Choy, E.-J., Kang, C.-K., Lee, W.-C., Kim, S.-J.,
750 Moon, H.-B., 2015. Persistent organochlorines in 13 shark species from offshore and
751 coastal waters of Korea: Species-specific accumulation and contributing factors.
752 Ecotoxicol. Environ. Saf. 115, 195–202. https://doi.org/10.1016/j.ecoenv.2015.02.021
753 Li, Y.F., 1999. Global technical hexachlorocyclohexane usage and its contamination
754 consequences in the environment: from 1948 to 1997. Sci. Total Environ. 232, 121–
755 158. https://doi.org/10.1016/S0048-9697(99)00114-X

Li, Y.F., Macdonald, R.W., 2005. Sources and pathways of selected organochlorine pesticides to the Arctic and the effect of pathway divergence on HCH trends in biota: a review. Sci. Total Environ. 342, 87–106. https://doi.org/10.1016/j.scitotenv.2004.12.027

Logan, R.K., White, C.F., Winkler, C., Jorgensen, S.J., O'Sullivan, J.B., Lowe, C.G., Lyons, K., 2018. An evaluation of body condition and morphometric relationships within southern California juvenile white sharks *Carcharodon carcharias*. J. Fish Biol. 93, 842–849. https://doi.org/10.1111/jfb.13785

Lyons, K., Lowe, C.G., 2013. Quantification of Maternal Offloading of Organic Contaminants in Elasmobranchs Using the Histotrophic Round Stingray (*Urobatis halleri*) as a Model. Environ. Sci. Technol. 47, 12450–12458. https://doi.org/10.1021/es402347d

Lyons, K., Adams, D.H., 2015. Maternal offloading of organochlorine contaminants in the yolk-sac placental scalloped hammerhead shark (*Sphyrna lewini*). Ecotoxicology 24, 553–562. https://doi.org/10.1007/s10646-014-1403-7

Lyons, K., Kacev, D., Preti, A., Gillett, D., Dewar, H., Kohin, S., 2019. Species-Specific Characteristics Influence Contaminant Accumulation Trajectories and Signatures Across Ontogeny in Three Pelagic Shark Species. Environ. Sci. Technol. 53, 6997–7006. https://doi.org/10.1021/acs.est.8b07355

Mackay, D., Fraser, A., 2000. Bioaccumulation of persistent organic chemicals: mechanisms and models. Environ. Pollut. 110, 375–391. https://doi.org/10.1016/S0269-7491(00)00162-7

Madenjian, C.P., Schrank, C.S., Begnoche, L.J., Elliott, R.F., Quintal, R.T., 2010. Sexual difference in PCB concentrations of coho salmon (*Oncorhynchus kisutch*). Sci. Total Environ. 408, 1719–1724. https://doi.org/10.1016/j.scitotenv.2009.12.023

Marsili, L., Coppola, D., Giannetti, M., Casini, S., Fossi, M., Van WYK, J., Sperone, E., Tripepi, S., Micarelli, P., Rizzuto, S., 2016. Skin Biopsies as a Sensitive Non-Lethal Technique for the Ecotoxicological Studies of Great White Shark (*Carcharodon*

783 *carcharias*) Sampled in South Africa. Expert Opin. Environ. Biol. 04.
784 https://doi.org/10.4172/2325-9655.1000126
785 Martin, J.W., Mabury, S.A., Solomon, K.R., Muir, D.C.G., 2003. Dietary accumulation of
786 perfluorinated acids in juvenile rainbow trout (*Oncorhynchus mykiss*). Environ.
787 Toxicol. Chem. 22, 189–195.
788 Martin, J.W., Smithwick, M.M., Braune, B.M., Hoekstra, P.F., Muir, D.C.G., Mabury, S.A.,
789 2004. Identification of Long-Chain Perfluorinated Acids in Biota from the Canadian
790 Arctic. Environ. Sci. Technol. 38, 373–380. https://doi.org/10.1021/es034727+
791 Mull, C.G., Lyons, K., Blasius, M.E., Winkler, C., O'Sullivan, J.B., Lowe, C.G., 2013.
792 Evidence of Maternal Offloading of Organic Contaminants in White Sharks
793 (*Carcharodon carcharias*). PLOS ONE 8, e62886.
794 https://doi.org/10.1371/journal.pone.0062886
795 Munschy, C., Bodin, N., Potier, M., Héas-Moisan, K., Pollono, C., Degroote, M., West, W.,
796 Hollanda, S.J., Puech, A., Bourjea, J., Nikolic, N., 2016. Persistent Organic Pollutants
797 in albacore tuna (Thunnus alalunga) from Reunion Island (Southwest Indian Ocean)
798 and South Africa in relation to biological and trophic characteristics. Environ. Res.
799 148, 196–206. https://doi.org/10.1016/j.envres.2016.03.042
800 Munschy, C., Bely, N., Pollono, C., Aminot, Y., 2019. Perfluoroalkyl substances (PFASs) in
801 the marine environment: Spatial distribution and temporal profile shifts in shellfish
802 from French coasts. Chemosphere 228, 640–648.
803 https://doi.org/10.1016/j.chemosphere.2019.04.205
804 Munschy, C., Vigneau, E., Bely, N., Héas-Moisan, K., Olivier, N., Pollono, C., Hollanda, S.,
805 Bodin, N., 2020. Legacy and emerging organic contaminants: Levels and profiles in
806 top predator fish from the western Indian Ocean in relation to their trophic ecology.
807 Environmental Research 188, 109761. https://doi.org/10.1016/j.envres.2020.109761
808 Mwevura, H., Amir, O.A., Kishimba, M., Berggren, P., Kylin, H., 2010. Organohalogen
809 compounds in blubber of Indo-Pacific bottlenose dolphin *(Tursiops aduncus*) and
810 spinner dolphin (Stenella longirostris) from Zanzibar, Tanzania. Environ. Pollut.,

811 Advances of air pollution science: from forest decline to multiple-stress effects on
812 forest ecosystem services 158, 2200–2207.
813 https://doi.org/10.1016/j.envpol.2010.02.027
814 Ng, C.A., Gray, K.A., 2009. Tracking bioaccumulation in aquatic organisms: A dynamic
815 model integrating life history characteristics and environmental change. Ecol. Model.
816 220, 1266–1273. https://doi.org/10.1016/j.ecolmodel.2009.02.007
817 Olin, J.A., Beaudry, M., Fisk, A.T., Paterson, G., 2014. Age-related polychlorinated biphenyl
818 dynamics in immature bull sharks (*Carcharhinus leucas*): PCB dynamics in immature
819 bull sharks. Environ. Toxicol. Chem. 33, 35–43. https://doi.org/10.1002/etc.2402
820 Pan, G., Zhou, Q., Luan, X., Fu, Q.S., 2014. Distribution of perfluorinated compounds in Lake
821 Taihu (China): Impact to human health and water standards. Sci. Total Environ. 487,
822 778–784. https://doi.org/10.1016/j.scitotenv.2013.11.100
823 Pirog, A., Magalon, H., Poirout, T., Jaquemet, S., 2019a. Reproductive biology, multiple
824 paternity and polyandry of the bull shark Carcharhinus leucas. J. Fish Biol. 95, 1195–
825 1206. https://doi.org/10.1111/jfb.14118
826 Pirog, A., Jaquemet, S., Ravigné, V., Cliff, G., Clua, E., Holmes, B.J., Hussey, N.E., Nevill,
827 J.E.G., Temple, A.J., Berggren, P., Vigliola, L., Magalon, H., 2019b. Genetic
828 population structure and demography of an apex predator, the tiger shark Galeocerdo
829 cuvier. Ecol. Evol. 9, 5551–5571. https://doi.org/10.1002/ece3.5111
830 Pirog, A., Ravigné, V., Fontaine, M.C., Rieux, A., Gilabert, A., Cliff, G., Clua, E., Daly, R.,
831 Heithaus, M.R., Kiszka, J.J., Matich, P., Nevill, J.E.G., Smoothey, A.F., Temple, A.J.,
832 Berggren, P., Jaquemet, S., Magalon, H., 2019c. Population structure, connectivity,
833 and demographic history of an apex marine predator, the bull shark Carcharhinus
834 leucas. Ecol. Evol. 9, 12980–13000. https://doi.org/10.1002/ece3.5597
835 Pirog, A., Magalon, H., Poirout, T., Jaquemet, S., 2020. New insights into the reproductive
836 biology of the tiger shark Galeocerdo cuvier and no detection of polyandry in Reunion
837 Island, western Indian Ocean. Mar. Freshw. Res. https://doi.org/10.1071/MF19244

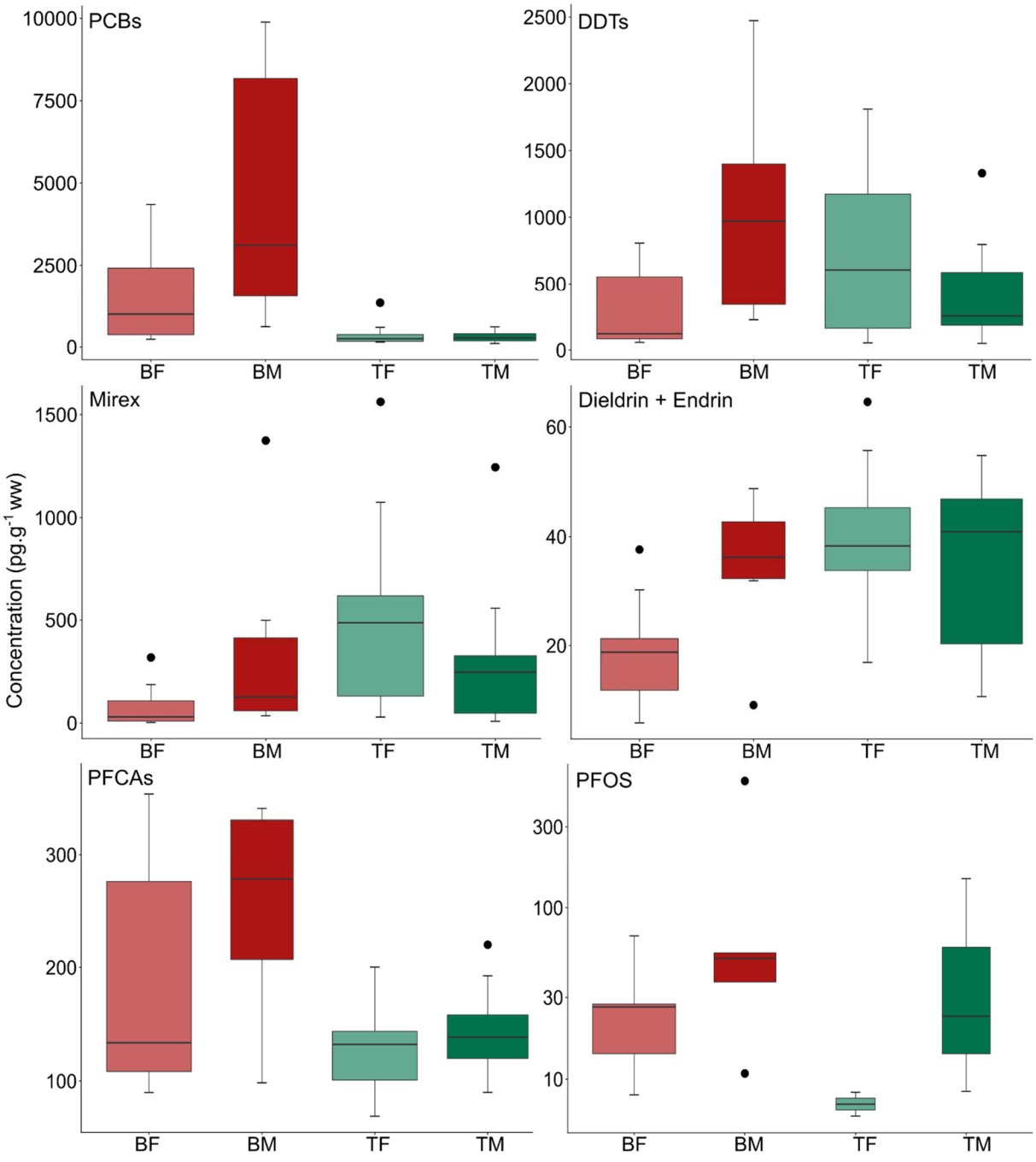
838 Pozo, K., Harner, T., Wania, F., Muir, D.C.G., Jones, K.C., Barrie, L.A., 2006. Toward a
839 Global Network for Persistent Organic Pollutants in Air: Results from the GAPS
840 Study. Environ. Sci. Technol. 40, 4867–4873. https://doi.org/10.1021/es060447t
841 Prevedouros, K., Cousins, I.T., Buck, R.C., Korzeniowski, S.H., 2006. Sources, Fate and
842 Transport of Perfluorocarboxylates. Environ. Sci. Technol. 40, 32–44.
843 https://doi.org/10.1021/es0512475
844 Qiu, X., Zhu, T., Yao, B., Hu, J., Hu, S., 2005. Contribution of Dicofol to the Current DDT
845 Pollution in China. Environ. Sci. Technol. 39, 4385–4390.
846 https://doi.org/10.1021/es050342a
847 Qiu, Y.-W., Wang, D.-X., Zhang, G., 2020. Assessment of persistent organic pollutants
848 (POPs) in sediments of the Eastern Indian Ocean. Sci. Total Environ. 710, 136335.
849 https://doi.org/10.1016/j.scitotenv.2019.136335
850 Ramos, R., González-Solís, J., 2012. Trace me if you can: the use of intrinsic
851 biogeochemical markers in marine top predators. Front. Ecol. Environ. 10, 258–266.
852 https://doi.org/10.1890/110140
853 Reid, B.J., Jones, K.C., Semple, K.T., 2000. Bioavailability of persistent organic pollutants in
854 soils and sediments—a perspective on mechanisms, consequences and assessment.
855 Environ. Pollut. 108, 103–112. https://doi.org/10.1016/S0269-7491(99)00206-7
856 Roscales, J.L., González-Solís, J., Zango, L., Ryan, P.G., Jiménez, B., 2016. Latitudinal
857 exposure to DDTs, HCB, PCBs, PBDEs and DP in giant petrels (Macronectes spp.)
858 across the Southern Ocean. Environ. Res. 148, 285–294.
859 https://doi.org/10.1016/j.envres.2016.04.005
860 Rotander, A., Kärrman, A., Bavel, B. van, Polder, A., Rigét, F., Auðunsson, G.A., Víkingsson,
861 G., Gabrielsen, G.W., Bloch, D., Dam, M., 2012. Increasing levels of long-chain
862 perfluorocarboxylic acids (PFCAs) in Arctic and North Atlantic marine mammals,
863 1984–2009. Chemosphere 86, 278–285.
864 https://doi.org/10.1016/j.chemosphere.2011.09.054

865 Rypel, A.L., Findlay, R.H., Mitchell, J.B., Bayne, D.R., 2007. Variations in PCB
866 concentrations between genders of six warmwater fish species in Lake Logan Martin,
867 Alabama, USA. Chemosphere 68, 1707–1715.
868 https://doi.org/10.1016/j.chemosphere.2007.03.046
869 Salvadó, J.A., Grimalt, J.O., López, J.F., Durrieu de Madron, X., Pasqual, C., Canals, M.,
870 2013. Distribution of organochlorine compounds in superficial sediments from the
871 Gulf of Lion, northwestern Mediterranean Sea. Prog. Oceanogr., Integrated study of a
872 deep submarine canyon and adjacent open slopes in the Western Mediterranean
873 Sea: an essential habitat 118, 235–248. https://doi.org/10.1016/j.pocean.2013.07.014
874 Schlenk, D., Sapozhnikova, Y., Cliff, G., 2005. Incidence of organochlorine pesticides in
875 muscle and liver tissues of South African great white sharks *Carcharodon carcharias*.
876 Mar. Pollut. Bull. 50, 208–211. https://doi.org/10.1016/j.marpolbul.2004.11.032
877 Shen, L., Wania, F., 2005. Compilation, Evaluation, and Selection of Physical–Chemical
878 Property Data for Organochlorine Pesticides. J. Chem. Eng. Data 50, 742–768.
879 https://doi.org/10.1021/je049693f
880 Srimurali, S., Govindaraj, S., Krishna Kumar, S., Babu Rajendran, R., 2015. Distribution of
881 organochlorine pesticides in atmospheric air of Tamilnadu, southern India. Int. J.
882 Environ. Sci. Technol. 12, 1957–1964. https://doi.org/10.1007/s13762-014-0558-3
883 Storelli, M.M., Ceci, E., Storelli, A., Marcotrigiano, G.O., 2003. Polychlorinated biphenyl,
884 heavy metal and methylmercury residues in hammerhead sharks: contaminant status
885 and assessment. Mar. Pollut. Bull. 46, 1035–1039. https://doi.org/10.1016/S0025-
886 326X(03)00119-X
887 Storelli, M.M., Barone, G., Santamaria, N., Marcotrigiano, G.O., 2006. Residue levels of
888 DDTs and toxic evaluation of polychlorinated biphenyls (PCBs) in Scyliorhinus
889 canicula liver from the Mediterranean Sea (Italy). Mar. Pollut. Bull. 52, 696–700.
890 https://doi.org/10.1016/j.marpolbul.2006.02.013
891 Sturm, R., Ahrens, L., 2010. Trends of polyfluoroalkyl compounds in marine biota and in
892 humans. Environ. Chem. 7, 457–484. https://doi.org/10.1071/EN10072

893 Suárez, P., Ruiz, Y., Alonso, A., San Juan, F., 2013. Organochlorine compounds in mussels
894 cultured in the Ría of Vigo: Accumulation and origin. Chemosphere 90, 7–19.
895 https://doi.org/10.1016/j.chemosphere.2012.02.030
896 Tanabe, S., 2002. Contamination and toxic effects of persistent endocrine disrupters in
897 marine mammals and birds. Mar. Pollut. Bull. 45, 69–77.
898 https://doi.org/10.1016/S0025-326X(02)00175-3
899 Tanabe, S., Ramu, K., 2012. Monitoring temporal and spatial trends of legacy and emerging
900 contaminants in marine environment: Results from the environmental specimen bank
901 (es-BANK) of Ehime University, Japan. Mar. Pollut. Bull. 64, 1459–1474.
902 https://doi.org/10.1016/j.marpolbul.2012.05.013
903 Trumble, S.J., Robinson, E.M., Noren, S.R., Usenko, S., Davis, J., Kanatous, S.B., 2012.
904 Assessment of legacy and emerging persistent organic pollutants in Weddell seal
905 tissue (*Leptonychotes weddellii*) near McMurdo Sound, Antarctica. Sci. Total Environ.
906 439, 275–283. https://doi.org/10.1016/j.scitotenv.2012.09.018
907 Trystram, C., Rogers, K.M., Soria, M.M., Jaquemet, S., 2017. Feeding patterns of two
908 sympatric shark predators in coastal ecosystems of an oceanic island. Can. J. Fish.
909 Aquat. Sci. 74, 216–227. https://doi.org/10.1139/cjfas-2016-0105
910 UNEP, 2001. United Nations Environment Programme, The Stockholm Convention on
911 Persistent Organic Pollutants [WWW Document]. URL
912 http://chm.pops.int/default.aspx (accessed 1.25.20).
913 van den Berg, H., Manuweera, G., Konradsen, F., 2017. Global trends in the production and
914 use of DDT for control of malaria and other vector-borne diseases. Malar. J. 16.
915 https://doi.org/10.1186/s12936-017-2050-2
916 Wang, Z., DeWitt, J.C., Higgins, C.P., Cousins, I.T., 2017. A Never-Ending Story of Per- and
917 Polyfluoroalkyl Substances (PFASs)? Environ. Sci. Technol. 51, 2508–2518.
918 https://doi.org/10.1021/acs.est.6b04806
919 Wania, F., MacKay, D., 1996. Peer Reviewed: Tracking the Distribution of Persistent Organic
920 Pollutants. Environ. Sci. Technol. 30, 390A-396A. https://doi.org/10.1021/es962399q

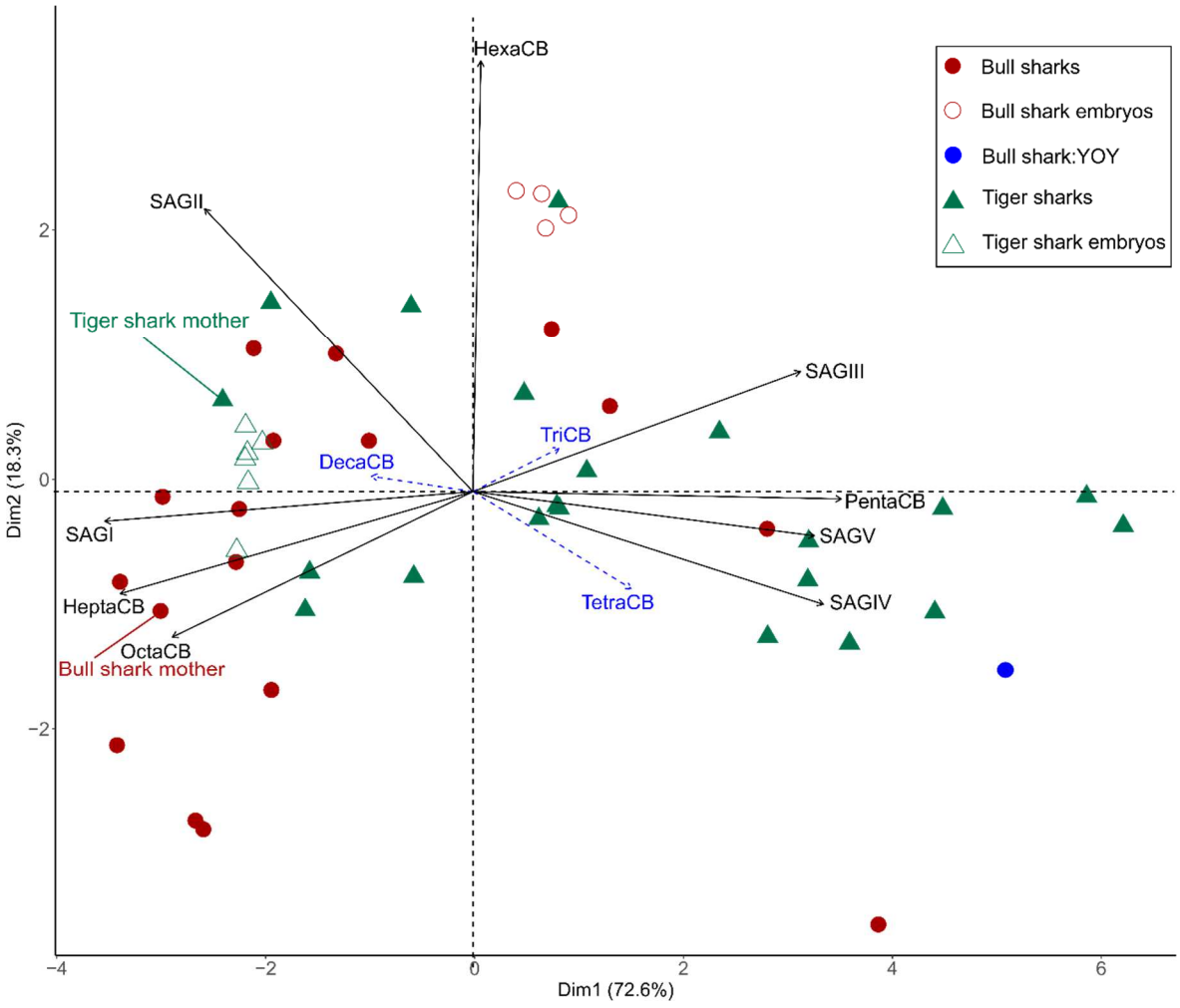
921 Weijs, L., Briels, N., Adams, D.H., Lepoint, G., Das, K., Blust, R., Covaci, A., 2015. Maternal
922 transfer of organohalogenated compounds in sharks and stingrays. Mar. Pollut. Bull.
923 92, 59–68. https://doi.org/10.1016/j.marpolbul.2014.12.056
924 Wenzl, T., Haedrich, J., Schaechtele, A., Robouch, P., Stroka, J., 2016. Guidance Document
925 for the Estimation of LOD and LOQ for Measurements in the Field of Contaminants in
926 Feed and Food, Publications Office of the European Union. ed, EUR. Luxembourg.
927 https://doi.org/10.2787/8931
928 Whitney, N.M., Crow, G.L., 2007. Reproductive biology of the tiger shark (*Galeocerdo cuvier*)
929 in Hawaii. Mar. Biol. 151, 63–70. https://doi.org/10.1007/s00227-006-0476-0
930 Yunker, M.B., Ikonomou, M.G., Sather, P.J., Friesen, E.N., Higgs, D.A., Dubetz, C., 2011.
931 Development and Validation of Protocols To Differentiate PCB Patterns between
932 Farmed and Wild Salmon. Environ. Sci. Technol. 45, 2107–2115.
933 https://doi.org/10.1021/es1038529
934

Figure 1: PCB (Σ 30 congeners), DDT (Σ 5 isomers), mirex, dieldrin+endrin, PFCA (Σ 6 compounds) and PFOS concentrations (pg g^{-1} ww) in female bull sharks (BF, n = 11), male bull sharks (BM, n = 6), female tiger sharks (TF, n = 11) and male tiger sharks (TM, n = 10) collected along the West coast of Reunion Island in 2018-2019. Median values (horizontal solid line inside the box), 25^{th} and 75^{th} percentiles (lower and upper ends of the boxes), 95% confidence intervals (whiskers) and outliers (circles) are shown. Box width is proportional to the number of data in each group.


PCBs
DDTs
Mirex
Dieldrin + Endrin
PFCAs
PFOS
Concentration (pg.g-1 ww)
BF
BM
TF
TM

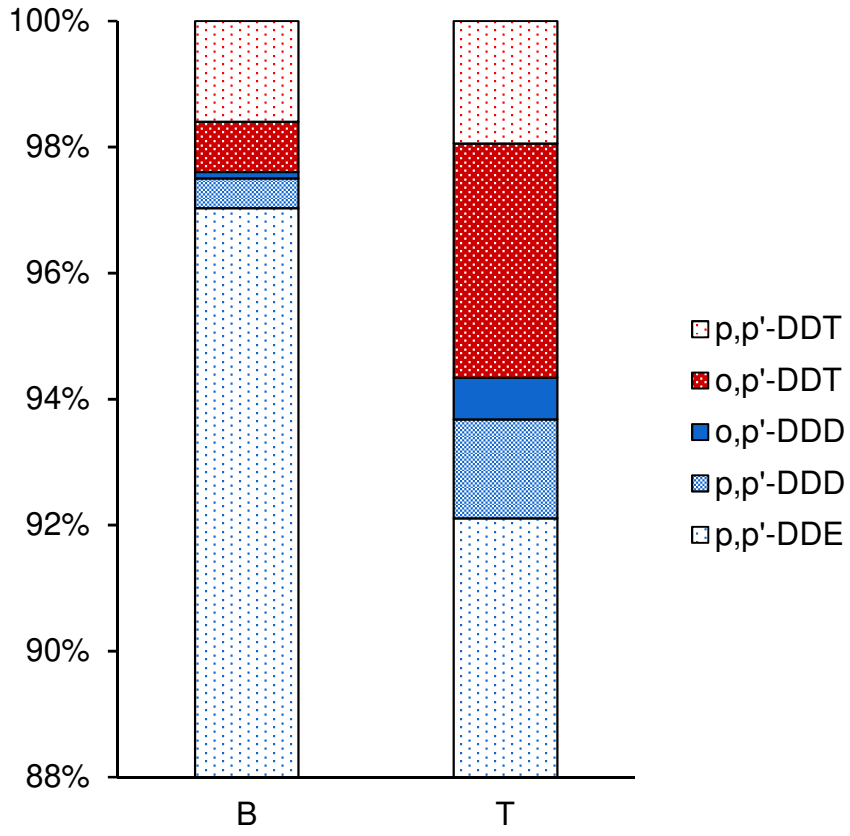
2 columns

Figure 2: Graph of the two principal components of the standardized PCA on the proportions of congeners grouped according to their chlorine numbers and their structure activity group (SAG) (black arrows) in bull shark (n = 17; red filled circles) and their embryos (n = 4; red empty circles), tiger shark (n = 21; green filled triangles) and their embryos (n = 6; green empty triangles). The blue circle represents the young-of-the-year (YOY) bull shark. Congener groups represented by blue arrows are misrepresented in this factorial plane and are added for information purposes without being taken into account in the calculations. TriCB: trichlorinated congeners; TetraCB: tetrachlorinated congeners; PentaCB: pentachlorinated congeners; HexaCB: hexachlorinated congeners; HeptaCB: heptachlorinated congeners; OctaCB: octachlorinated congeners; DecaCB: decachlorinated congeners.



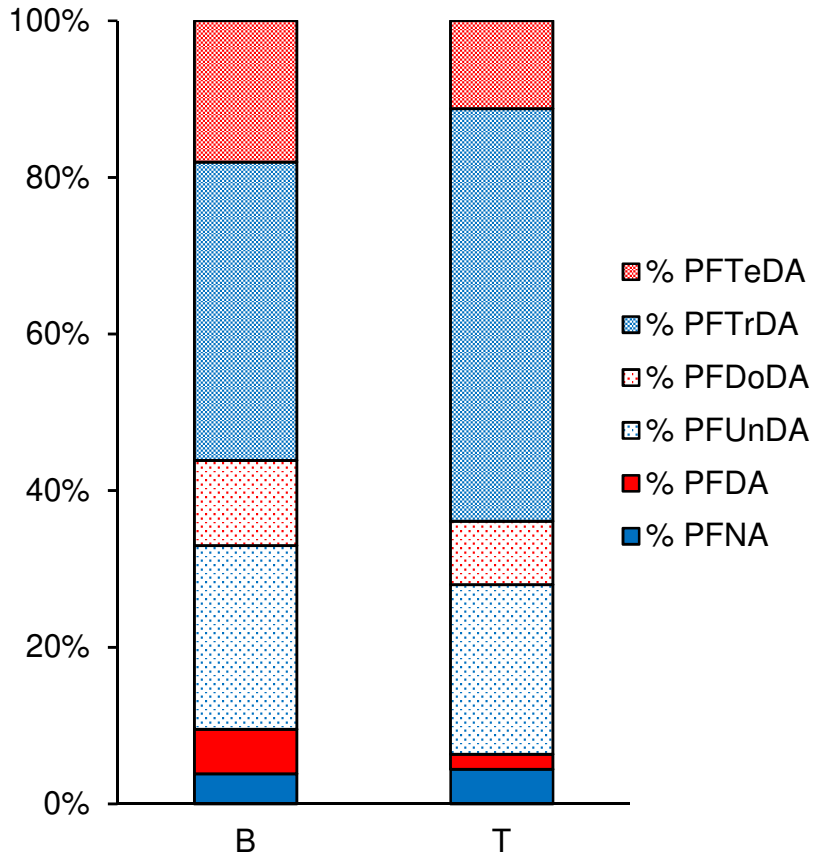
2 columns

Figure 1: Mean DDT contamination profiles (% of the ∑ 5 isomers) in bull sharks (B, n = 17) and tiger sharks (T, n = 21) collected in the West coast of Reunion Island in 2018-2019.



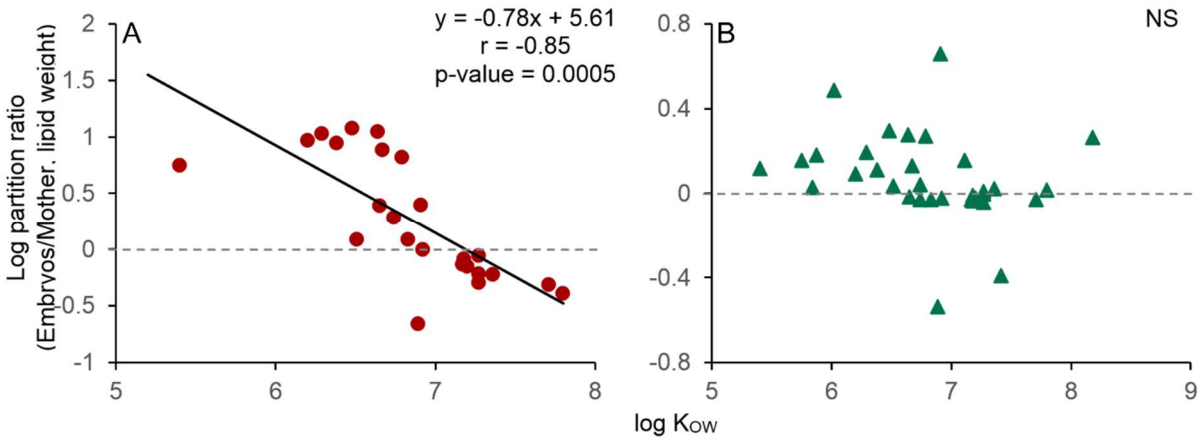
1 column

Figure 1: Mean PFCA contamination profiles in bull sharks (B, n = 17) and adult tiger sharks (T, n = 21) collected along the West coast of Reunion Island in 2018-2019. Odd PFCAs are shown in blue and even PFCAs in red and ranked in ascending carbon number order.



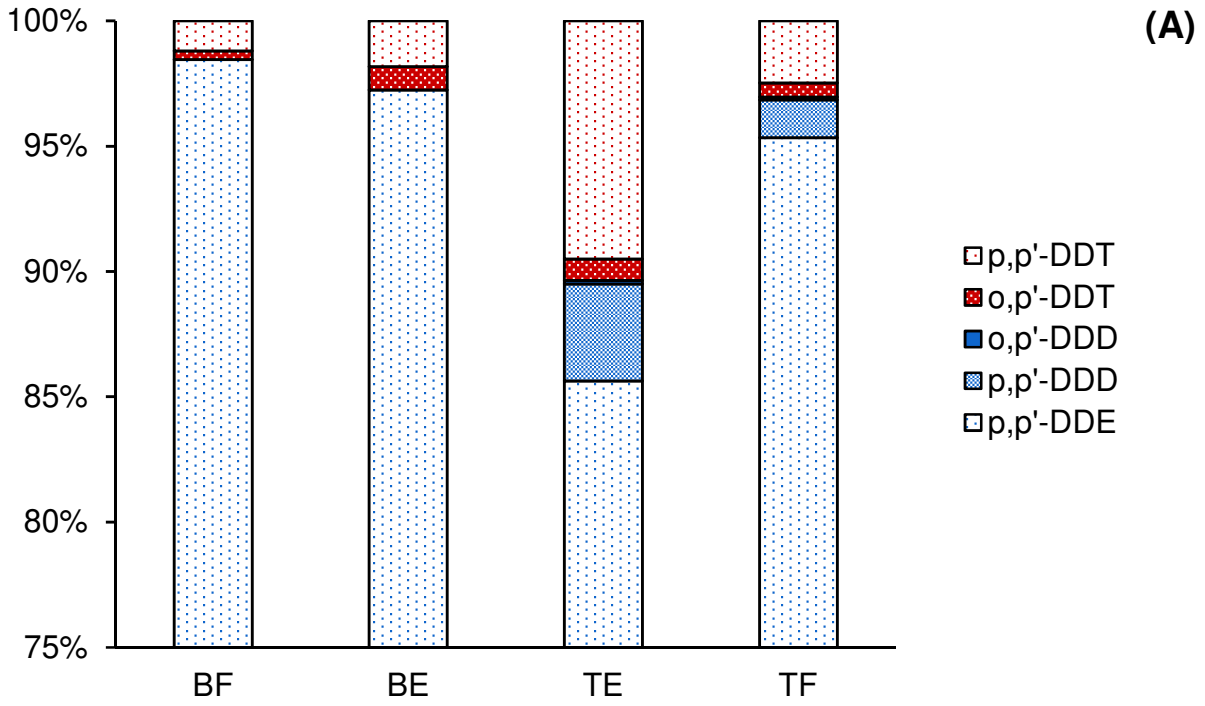
1 column

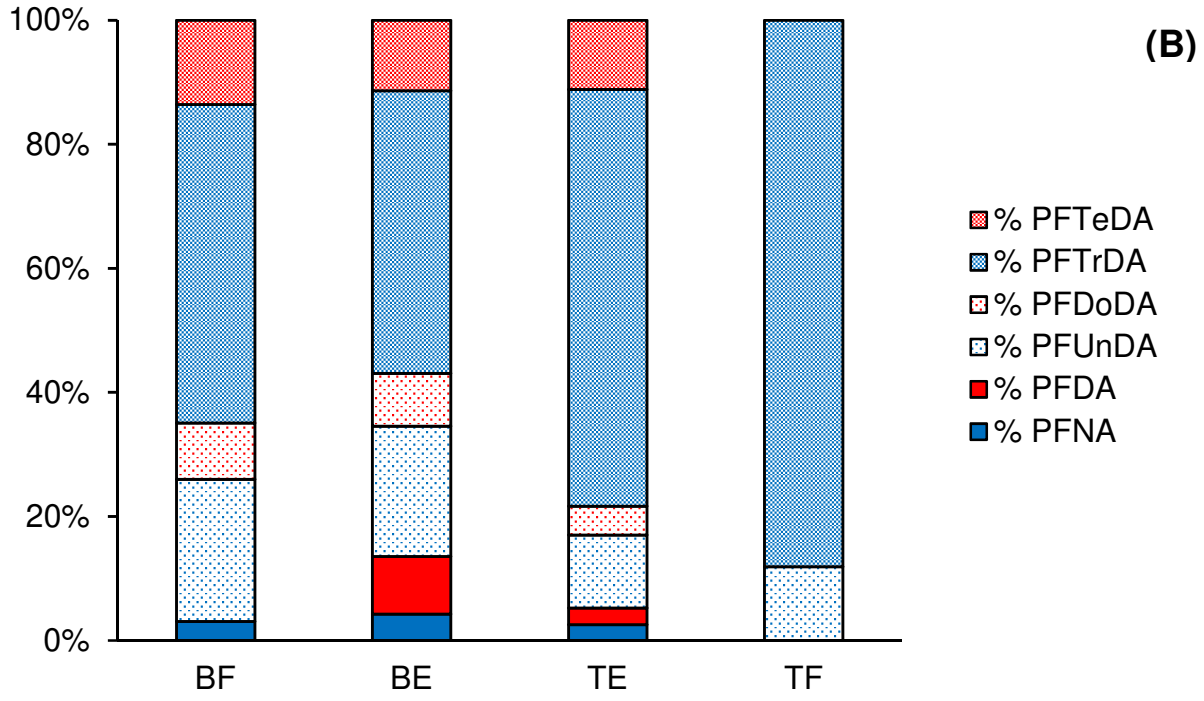
Figure 5: Partition ratios of OCPs and PCBs (pg.g^{-1} lipid weight) plotted against log octanol / water partition coefficient (log K_{OW}; Hawker and Connell, 1988; Shen and Wania, 2005) in bull sharks (red circles, A) and tiger sharks (green triangles, B). Partition ratios were calculated as mean congener concentrations in embryos relative to the concentrations in the mother and log_{10}-transformed. Dashed line: partition ratio = 1. NS = non-significant.



2 columns

Figure 6: Mean contamination profiles of DDT (A) and PFCAs (B) in bull shark mother (BF, n = 1), bull shark embryos (BE, n = 4), tiger shark embryos (TE, n = 6) and tiger shark mother (TF, n = 1) collected along the West coast of Reunion Island in 2018-2019. Odd PFCAs are shown in blue and even PFCAs in red. PFCAs are ranked in ascending order of their carbon number.




(B)
100%
80%
60%
40%
20%
0%
BF
BE
TE
TF
% PFTeDA
% PFTrDA
% PFDoDA
% PFUnDA
% PFDA
% PFNA

1 column

Table 1 : Biological parameters (total length: TL (cm); body mass (kg); Fulton's condition factor: K (g cm^{-3}); total lipid content (TLC in % wet weight)) measured in bull and tiger sharks collected along the West coast of Reunion Island in 2018-2019. Data are expressed as mean ± standard deviation (minimum-maximum).

		Total length (cm)	**Body mass (kg)**	**K (g cm^{-3})**	**TLC (% ww)**
Bull shark	All (n = 17)	231 ± 57 (160 - 327)	109.2 ± 74.7 (27.8 - 299.2)	0.74 ± 0.08 (0.66 - 0.96)	0.57 ± 0.13 (0.19 - 0.76)
	Females (n = 11)	230 ± 62 (202 - 327)	113.2 ± 82.1 (59.0 - 299.2)	0.76 ± 0.09 (0.66 - 0.96)	0.58 ± 0.10 (0.41 - 0.76)
	Males (n = 6)	233 ± 54 (160 - 297)	101.2 ± 63.5 (27.8 - 189.0)	0.71 ± 0.03 (0.68 - 0.74)	0.57 ± 0.20 (0.19 - 0.74)
	Embryos (n = 4)*	63 ± 1 (62 - 64)	1.7 ± 0.1 (1.6 - 1.8)	0.74 ± 0.005 (0.67 - 0.69)	0.68 ± 0.13 (0.49 - 0.77)
	Young-of-the-year (n = 1)	82	4.0	0.70	0.84
Tiger shark	All (n = 21)	275 ± 67 (157 – 387)	133.5 ± 110.9 (14.4 - 402.4)	0.50 ± 0.10 (0.36 - 0.72)	0.65 ± 0.15 (0.53 - 1.22)
	Females (n = 11)	265 ± 66 (157 – 367)	106.7 ± 83.8 (14.4 - 298.3)	0.52 ± 0.12 (0.36 - 0.72)	0.62 ± 0.06 (0.53 - 0.71)
	Males (n = 10)	285 ± 69 (190 - 387)	157.8 ± 130.1 (25.0 - 402.4)	0.48 ± 0.07 (0.37 - 0.60)	0.67 ± 0.20 (0.54 - 1.22)
	Embryos (n = 6)**	33 ± 1 (31 - 34)	0.12 ± 0.01 (0.11 - 0.14)	0.35 ± 0.03 (0.32 - 0.40)	1.67 ± 0.38 (1.17 - 2.23)

*: 4 pools of 2 individuals

**: 4 pools of 2 individuals and 2 individuals

Table 1: Concentrations (pg g^{-1} ww) of the various families of studied POPs in bull and tiger sharks collected along the West coast of Reunion Island in 2018-2019. Data are expressed as mean ± standard deviation (minimum - maximum). LOQ: limit of quantification.

			Bull shark		**Tiger shark**	
		Young-of-the-year n = 1	All individuals n = 17	Embryo n = 4*	All individuals n = 21	Embryo n = 6**
PCBs	**∑i-PCBs**	83	1780 ± 2063 (155 - 7013)	259 ± 17 (240 - 278)	219 ± 186 (67 - 900)	5739 ± 397 (5236 - 6156)
	∑dl-PCBs	20	242 ± 322 (26 - 1171)	47 ± 3 (43 - 50)	36 ± 21 (14 - 91)	558 ± 39 (516 - 605)
	∑30 PCBs	137	2597 ± 2969 (235 - 9885)	370 ± 23 (347 - 397)	339 ± 270 (104 - 1343)	8735 ± 716 (7986 - 9739)
OCPs	**∑DDTs**	89	584 ± 648 (57 - 2473)	83 ± 8 (73 - 92)	585 ± 580 (50 - 1811)	10471 ± 897 (9334 - 11658)
	Mirex	5	180 ± 334 (3 - 1374)	7 ± 1 (6 - 8)	403 ± 431 (9 - 1562)	755 ± 88 (642 - 885)
	Dieldrin+endrin	24	25 ± 14 (6 - 49)	11 ± 1 (11 - 12)	38 ± 15 (11 - 65)	184 ± 19 (157 - 204)
	∑OCPs	118	785 ± 966 (67 - 3879)	102 ± 8 (90 - 111)	1025 ± 946 (90 - 3412)	11409 ± 920 (10351 - 12557)
PFASs	**PFOS**	46	67 ± 142 (< LOQ - 555)	77 ± 5 (< LOQ - 82)	39 ± 61 (< LOQ - 148)	24 ± 7 (< LOQ - 29)
	∑PFCAs	109	212 ± 98 (90 - 354)	177 ± 20 (156 - 200)	135 ± 38 (69 - 220)	1382 ± 254 (1011 - 1685)
	∑PFASs	155	267 ± 194 (90 - 896)	234 ± 36 (187 - 274)	144 ± 53 (69 - 293)	1390 ± 260 (1011 - 1685)

*: 4 pools of 2 individuals

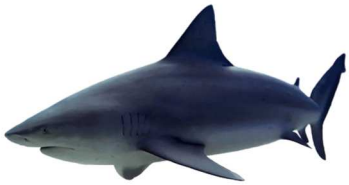
**: 4 pools of 2 individuals and 2 individuals

Table 1: PCB (Σ 30), DDT (Σ 5), mirex, dieldrin-endrin, PFCA (Σ 6) and PFOS concentrations (pg g^{-1} ww) in the white muscle of bull and tiger shark embryos and their mother collected along the West coast of Reunion Island in 2018-2019. Data are expressed as mean ± standard deviation (minimum - maximum).

		PCBs	**DDTs**	**Mirex**	**Dieldrin-endrin**	**PFCAs**	**PFOS**
Bull sharks	Mother	4348	400	187	12	269	27
	Embryos (n = 4)*	370 ± 23 (347 - 397)	83 ± 8 (73 - 92)	7 ± 1 (6 - 8)	11 ± 1 (11 - 12)	177 ± 20 (156 - 200)	77 ± 5 (74 - 82)
Tiger sharks	Mother	1343	1655	496	26	61	< LOQ
	Embryos (n = 6)**	8735 ± 716 (7986 - 9739)	10471 ± 897 (9334 - 11658)	755 ± 88 (642 - 885)	184 ± 19 (157 - 204)	1382 ± 254 (1011 - 1685)	(19 - 29)

*: 4 pools of 2 individuals

**: 4 pools of 2 individuals and 2 individuals

Bull sharks		Tiger sharks
		
Coastal		*Neritic*
++++	PCBs	+
++	DDTs	++
++	PFASs	+