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## **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            1. Introduction

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

8    Contaminants that are persistent, bio-accumulative and toxic, such as persistent organic  
9    pollutants (POPs), may inflict the greatest amount of damage on ecosystems (UNEP, 2001;  
10   Green and Larson, 2016; Jepson and Law, 2016). POPs, which are essentially synthesized  
11   by humans are characterized by their ability to travel long distances, resulting in their transfer  
12   far from emission sources (Beyer et al., 2000; Brown and Wania, 2008; Corsolini et al.,  
13   2014), mainly through atmospheric volatilization and condensation cycles referred to as  
14   global distillation (Dachs et al., 2002; Wania and MacKay, 1996). POPs are hence  
15   widespread in the environment; they occur in various matrices and regions around the world  
16   (Dachs et al., 2002; Pozo et al., 2006; Reid et al., 2000) and as such are considered as  
17   global pollutants (Pozo et al., 2006). POPs include well-studied polychlorinated biphenyls  
18   (PCBs) and various organochlorine pesticides (OCPs), as well as other substances listed  
19   more recently in the Stockholm Convention (UNEP, 2001), such as perfluorooctane sulfonate  
20   (PFOS). Historically, OCPs, in particular hexachlorocyclohexane (HCH) and  
21   dichlorodiphenyltrichloroethane (DDT), were synthesized in high quantities (Li, 1999; Li and  
22   Macdonald, 2005). Although the production of legacy POPs has ceased in most countries,  
23   contemporary inputs into the environment continue to originate from secondary sources such  
24   as open burning, disposal of products containing PCBs, recycling of electric and electronic  
25   devices or PCB-containing pigments used in household paints (Breivik et al., 2011; Grimm et  
26   al., 2015), or from direct use, e.g. DDT against vectors of diseases (Qiu et al., 2005; van den

27 Berg et al., 2017). The ban on certain chemical substances has led to their replacement with  
28 new substances, classified as contaminants of emerging concern (CECs). For example, the  
29 ban on PFOS has led to the use of long-chain (> 7 carbon atoms) perfluorinated carboxylic  
30 acids (PFCAs), found in increasing amounts in the environment (Wang et al., 2017). Oceans  
31 are recognized as the main reservoirs of these compounds (Johansson et al., 2019). In this  
32 context, both legacy POPs and CECs need to be studied in many oceanic regions, in  
33 particular those remote from major sources of direct pollution, such as the southern Indian  
34 Ocean (Corsolini et al., 2016; Hoydal et al., 2015; Roscales et al., 2016; Trumble et al.,  
35 2012).

36 The tiger shark (*Galeocerdo cuvier*: Péron & Lesueur, 1822) and bull shark (*Carcharhinus*  
37 *leucas*: Müller & Henle, 1839) are apex predators inhabiting both coastal and oceanic tropical  
38 ecosystems (Compagno, 1984), where they play a major role (Heithaus et al., 2008). Both  
39 species are generalist feeders that can forage on prey of aquatic or terrestrial origin (Cliff and  
40 Dudley, 1991; Dicken et al., 2017; Trystram et al., 2017). More specifically, in Reunion Island  
41 (RUN), an oceanic island in the Southwest Indian Ocean, these two sympatric species differ  
42 in terms of prey and trophic habitat specialization (Le Croizier et al., 2020; Trystram et al.,  
43 2017). Both species are characteristic of the marine fauna of RUN (Fricke et al., 2009) and  
44 play a key role in the island's coastal ecosystem dynamics. However, no rigorous  
45 assessment of their exposure to organic contaminants has so far been conducted, despite  
46 the fact that they feed at high trophic levels, as do marine mammals (Jepson and Law,  
47 2016), which are subject to excessively high concentrations of PCBs associated with long-  
48 term population declines (Dirtu et al., 2016; Mwevura et al., 2010; Tanabe, 2002). Actually,  
49 the contamination of marine ecosystems by organic contaminants in RUN has been the  
50 subject of a very limited number of studies (Dirtu et al., 2016; Munschy et al., 2016), in  
51 particular with regards to CECs. More generally, organic contaminant occurrence in large  
52 carnivorous shark species has been poorly documented (Cagnazzi et al., 2019; Fisk et al.,  
53 2002; Gelsleichter et al., 2005; Lee et al., 2015; Schlenk et al., 2005; Weijs et al., 2015),  
54 despite the fact that most of these species are threatened with overexploitation and habitat

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

## 79 2. Materials and methods

### 80 2.1. Sample collection

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

$$99 \quad K = 100 \times ((W \times 10^3) \times T_L^{-3})$$

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

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

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

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

## 112 2.2. Total lipid content analysis

113 Extractable organic matter, used as a proxy for total lipid content (TLC), was determined with  
114 0.5 g dry weight (dw) of sample extracted with a mixture of n-hexane and acetone (80/20  
115 volume/volume [v/v]) using pressurized liquid extraction (PLE) at 100 °C under 10 MPa by  
116 means of an ASE 350 (Dionex©) (Munsch et al., 2020). The extracts were evaporated to  
117 dryness and TLC was determined gravimetrically and expressed in % of wet weight (ww).

## 118 2.3. Organic contaminant analysis

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

131 coupled to an AutoSpec Ultima mass spectrometer (Waters Corp.). The samples were  
132 analysed for 30 PCBs ranging from trichlorinated to decachlorinated congeners, including the  
133 12 dioxin-like (dl) -PCBs (CB-77, -81, -105, -114, -118, -123, -126, -156, -157, -167, -169, -  
134 189), the 6 indicator (i)-PCBs (CB-28, -52, -101, -138, -153, -180) and various OCPs (*p,p'*-  
135 DDT, *o,p'*-DDT, *o,p'*-DDD, *p,p'*-DDD, *p,p'*-DDE, dieldrin, aldrin, isodrin, mirex and  
136 hexachlorobenzene -HCB). Compounds were quantified by isotopic dilution using the  
137 corresponding  $^{13}\text{C}_{12}$ -labelled isomers (except mirex, quantified using  $^{13}\text{C}$  *p,p'*-DDE) and  
138 internal standard method was used to quantify samples. Prior to injection, a solution  
139 containing  $^{13}\text{C}_{12}$ -labeled CB-70, -111 and -170,  $\text{d}_8$ -labeled *p,p'*-DDD and *o,p'*-DDT was added  
140 to the final purified extracts for signal correction. Perfluoroalkylated substances (PFASs)  
141 were determined according to Munschy et al., 2019. Briefly, one gram of a freeze-dried  
142 sample, to which an internal standard mixture of nine labelled compounds was added prior to  
143 agitation, was extracted using liquid-solid extraction with MeOH/KOH (0.01 M of KOH),  
144 purified onto two consecutive SPE cartridges (a WAX weak anion exchange stationary phase  
145 and an Envicarb charcoal stationary phase, evaporated to dryness and reconstituted in 200  
146  $\mu\text{L}$  of a mixture of MeOH:H<sub>2</sub>O (50:50, v/v), to which PFOS  $^{13}\text{C}_8$  was added. The following  
147 compounds were analysed, including five C<sub>4</sub>- to C<sub>10</sub>-perfluoroalkyl sulfonates (PFASs) and  
148 nine C<sub>6</sub>- to C<sub>14</sub> perfluorocarboxylic acids (PFCAs): perfluorobutane sulfonate (PFBS);  
149 perfluorohexane sulfonate (PFHxS); perfluoroheptane sulfonate (PFHpS); perfluorooctane  
150 sulfonate (PFOS); perfluorodecane sulfonate (PFDS); perfluorohexanoic acid (PFHxA);  
151 perfluoroheptanoic acid (PFHpA); perfluorooctanoic acid (PFOA); perfluorononanoic acid  
152 (PFNA); perfluorodecanoic acid (PFDA); perfluoroundecanoic acid (PFUnDA);  
153 perfluorododecanoic acid (PFDoDA); perfluorotridecanoic acid (PFTrDA) and  
154 perfluorotetradecanoic acid (PFTeDA). Targeted analytes were quantified using the  
155 corresponding isotope labelled standard, unless otherwise stated. The labelled standards  
156 were PFHxS  $^{18}\text{O}_2$  (used to quantify PFBS and PFHxS), PFOS  $^{13}\text{C}_4$  (used to quantify PFHpS,  
157 PFOS and PFDS), PFHxA  $^{13}\text{C}_2$  (used to quantify PFHxA and PFHpA), PFOA  $^{13}\text{C}_4$ , PFNA  
158  $^{13}\text{C}_5$ , PFDA  $^{13}\text{C}_2$ , PFUnDA  $^{13}\text{C}_2$ , PFDoDA  $^{13}\text{C}_2$ , and PFTeDA  $^{13}\text{C}_2$  (used to quantify PFTrDA



159 and PFTeDA). PFOS  $^{13}\text{C}_8$  was added to the purified extracts before injection and used as an  
160 injection standard. Analysis was performed using an Acquity ultra-performance liquid  
161 chromatograph (UPLC®, Waters Corp.) coupled to a triple quadrupole mass spectrometer  
162 (Xevo® TQ-S micro, Waters Corp.) interfaced with a Z-spray™ (Waters Corp.) electrospray  
163 ionization source. UPLC separation was achieved using an Acquity UPLC BEH C18  
164 reversed-phase column (50 mm x 2.1 mm; 1.7µm particle size) eluted with ammonium  
165 acetate in water (20 mM) (A) and methanol (B).

#### 166 2.4. QA/QC

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

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

177 Procedural blanks, incorporating all steps of the protocol, were analysed with each series of  
178 12 samples ( $n = 4$ ). Contaminants detected in more than 50% of the procedural blanks  
179 included various PCB congeners (CB-28, -31, -44, -49, -52, -66, -77, -87, -101, -105, -110, -  
180 118, -132, -138, -149, -151, -153, -156, -170, -180, -183 and -187), HCB and dieldrin for  
181 OCPs, and PFHxA, PFHpA, PFOA, PFNA, PFTTrDA and PFTeDA for PFASs. Concentrations  
182 in blanks, calculated with the extracted sample masses, were 0.22 - 3.2  $\text{pg g}^{-1}$  ww for PCBs,  
183 1.9 - 26.8  $\text{pg g}^{-1}$  ww for OCPs and 1.4 - 37.7  $\text{pg g}^{-1}$  ww for PFASs. Concentrations in samples  
184 were corrected using the blank value of each sample batch. The recovery ( $n = 49$ ) of each  
185 labelled compound ranged from  $67 \pm 8\%$  (CB-52) to  $85 \pm 16\%$  (CB-167) for PCBs,  $42 \pm 14\%$

186 ( $\alpha$ -endosulfan) to  $115 \pm 22\%$  (dieldrin) for OCPs and  $38 \pm 10\%$  (PFTeDA) to  $85 \pm 10\%$   
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  $\text{pg g}^{-1}$  ww to  $1.36 \text{ pg g}^{-1}$  ww for PCBs,  $0.1 \text{ pg g}^{-1}$  ww to  $27.4 \text{ pg g}^{-1}$  ww for OCPs and  $0.2 \text{ pg g}^{-1}$   
193  $\text{pg g}^{-1}$  ww to  $19.0 \text{ pg g}^{-1}$  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.  $33 \pm 1 \text{ cm}$  /  $0.12 \pm$

211 0.01 kg and  $63 \pm 1$  cm /  $1.7 \pm 0.1$  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<sup>-1</sup>  
253 ww for  $\sum$  30 PCBs, 50 to 2473 pg g<sup>-1</sup> ww for  $\sum$  DDTs, 3 to 1562 pg g<sup>-1</sup> ww for mirex and 6 to  
254 49 pg g<sup>-1</sup> 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  $1455.5 \pm 1359.7$  pg g<sup>-1</sup> ww in males and  $419.6 \pm 388.5$   
263 pg g<sup>-1</sup> ww in females; PCB concentrations were  $4602.7 \pm 4146.1$  pg g<sup>-1</sup> ww in males and

264 1502.8 ± 1331.7 pg g<sup>-1</sup> ww in females). When normalized to lipid content, these  
265 concentrations were significantly higher in males for both OCPs and PCBs (p = 0.006 for  
266 OCPs and p = 0.024 for PCBs): 243.7 ± 189.9 ng g<sup>-1</sup> lw versus 68.4 ± 59.3 ng g<sup>-1</sup> lw in  
267 females for OCPs and 745.7 ± 606.0 ng g<sup>-1</sup> lw in males versus 261.9 ± 240.0 ng g<sup>-1</sup> in  
268 females for PCBs. Conversely, PCB and OCP concentrations were similar (p = 0.35) in both  
269 sexes in tiger sharks: OCP concentrations were 756.6 ± 756.5 pg g<sup>-1</sup> ww in males and  
270 1268.4 ± 1066.1 pg g<sup>-1</sup> ww in females; PCB concentrations were 298.6 ± 155.0 pg g<sup>-1</sup> ww in  
271 males and 375.4 ± 348.1 pg g<sup>-1</sup> ww in females. When normalized to lipid content, these  
272 concentrations were 114.7 ± 105.6 ng g<sup>-1</sup> lw in males versus 208.0 ± 173.5 ng g<sup>-1</sup> lw in  
273 females for OCPs and 43.4 ± 17.3.0 ng g<sup>-1</sup> lw in males versus 59.0 ± 49.0 ng g<sup>-1</sup> lw in  
274 females for PCBs (p = 0.17 and p = 0.76 respectively). Differences in contamination levels  
275 between sexes may be caused by various factors such as reproductive loss, growth rate or  
276 diet (Larsson et al., 1993; Madenjian et al., 2010; Ng and Gray, 2009; Rypel et al., 2007). No  
277 sex-related dietary differences have been reported between the two species (Trystram et al.,  
278 2017); moreover, males have a higher growth rate than females (Cruz-Martínez et al., 2004;  
279 Kneebone et al., 2008). Neither of these factors would lead to the differences observed in  
280 contaminant accumulation in male and female bull sharks. The lack of sex-related influences  
281 on POP contamination in tiger sharks could indicate that reproduction has less impact on  
282 hydrophobic pollutant concentrations in ovoviviparous female tiger sharks versus viviparous  
283 female bull sharks. The results suggest that the ovoviviparous mode of gestation induces  
284 less decontamination in female shark muscle than viviparity. Actually, the continuous supply  
285 of nutrients during bull shark gestation could result in higher maternal contaminant offloading  
286 in comparison to egg-forming species such as tiger sharks (Castro et al., 2016). However,  
287 further investigations would be needed in order to confirm this hypothesis.

288 No significant differences in DDT concentrations were found between the two species (bull  
289 shark: 584 ± 648 pg g<sup>-1</sup> ww; tiger shark: 585 ± 580 pg g<sup>-1</sup> ww, p = 0.9), while PCB mean  
290 concentrations were 7 times higher in bull sharks (2597 ± 2968 pg g<sup>-1</sup> ww) than in tiger  
291 sharks (339 ± 270 pg g<sup>-1</sup> ww; p = 10<sup>-6</sup>). 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:  $403 \pm$   
297  $431 \text{ pg g}^{-1} \text{ ww}$ ; bull shark:  $180 \pm 334 \text{ pg g}^{-1} \text{ ww}$ ,  $p = 0.028$ ), while dieldrin concentrations were  
298 1.5 times higher in tiger sharks than in bull sharks (tiger shark:  $36 \pm 14 \text{ pg g}^{-1} \text{ ww}$ ; bull shark:  
299  $24 \pm 14 \text{ pg g}^{-1} \text{ ww}$ ,  $p = 0.016$ ). 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  $6.0 - 148.0 \text{ pg g}^{-1} \text{ ww}$  range (mean of

319 31.9 ± 34.1 pg g<sup>-1</sup> ww, one male bull shark exhibiting a higher concentration of 555 pg g<sup>-1</sup> ww  
320 was excluded), while ∑ PFCA concentrations were in the 69.0 - 353.5 pg g<sup>-1</sup> ww range, with  
321 mean concentrations of 135.0 ± 38.2 pg g<sup>-1</sup> ww in tiger sharks and 212.0 ± 97.8 pg g<sup>-1</sup> 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 PFTTrDA (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 metabolism 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.



### 370 3.3.2. DDTs

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

374 The relative proportions of *p,p'*-DDE were higher than those reported in other high trophic  
375 level predators in RUN by Munschy et al. (2016) and Dirtu et al. (2016). The ratio of (*p,p'*-  
376 DDE + *p,p'*-DDD) /  $\sum$  DDTs concentrations, which is an indicator of *p,p'*-DDT degradation in  
377 the environment, was not significantly different in the two species ( $0.95 \pm 0.04$  on average,  $p$   
378 = 0.88) and indicated that DDT sources in the studied sharks were old (Suárez et al., 2013).

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

### 384 3.3.3. OCP / PCB concentration ratios

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

396 2014; Lea et al., 2015; Pirog et al., 2019c; Trystram et al., 2017). In addition, the two species  
397 feed on different prey in RUN. These information on the trophic habits of the two species  
398 suggest that the coastal prey of bull sharks could be more highly-impacted by PCBs,  
399 although the sources and the physicochemical dynamics of these pollutants in the tissues of  
400 the two species could be an alternative explanation to the observed differences.  
401 Concentration ratios were positively correlated with tiger shark size ( $\rho = 0.60$ ;  $p = 0.0073$ )  
402 suggesting changes in contamination sources according to age. Similarly, this could either be  
403 due to movements throughout the Indian Ocean or to physiology-related changes.

404

#### 405 3.3.4. PFASs

406 Concentrations of  $\Sigma$ PFCA, including long-chain compounds ( $C \geq 8$ ), were higher than PFOS  
407 concentrations (on average by a factor of 11 and 17 in bull and tiger sharks, respectively) in  
408 all samples (the above-mentioned male bull shark outlier excluded), consistent with the  
409 higher bioaccumulative abilities of these compounds versus their short-chain counterparts  
410 (Kelly et al., 2009; Martin et al., 2003; Pan et al., 2014). More specifically, PFCA profiles  
411 showed a predominance of odd-chain length compounds (PFTrDA and PFUnDA) versus  
412 even-chain (PFDA, PFDoDA and PFTeDA) (Figure 4), as previously observed in fish (Martin  
413 et al., 2003; Sturm and Ahrens, 2010), including in the Indian Ocean (Munsch et al., 2020).  
414 Their presence has been partially explained by the degradation of fluorotelomer alcohols  
415 (FTOHs) (Ellis et al., 2004; Martin et al., 2004). PFUnDA and other long-chain PFCAs could  
416 originate from the degradation of 10:2 FTOH (Hart et al., 2008). Odd-chain PFCAs such as  
417 PFUnDA and PFTrDA have also been reported as impurities in PFNA resulting from the  
418 oxidation of fluorotelomer olefins (Prevedouros et al., 2006; Rotander et al., 2012).  
419 Tiger shark and bull shark long-chain PFCA profiles differed, with higher proportions of odd-  
420 chain PFCAs in tiger sharks ( $80 \pm 9\%$ ) versus bull sharks ( $66 \pm 13\%$ ;  $p = 0.00094$ ),  
421 suggesting that dietary differences could result in different contamination patterns.

## 422 3.4. Maternal transfer

### 423 3.4.1. Contamination levels

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

429 In bull shark embryos, concentrations (in ww) were 4 to 25 times lower than those of their  
430 mother for all chlorinated compounds except dieldrin (ratio mother / embryos of 1) (Table 3).  
431 When normalized to TLC, embryo contaminations were 6 to 35 times lower than those of  
432 their mother. Among perfluorinated compounds, PFCAs showed higher concentrations in the  
433 mother, while PFOS was 3 times more concentrated in embryos. These results demonstrate  
434 that lipophilic and amphiphilic molecules are not transmitted to embryos in the same manner,  
435 indicating that molecule physico-chemical properties and affinity with major biological  
436 macromolecules are determining parameters in the maternal transfer of organic  
437 contaminants in bull sharks. Conversely, concentrations of all chlorinated compounds and  
438 PFCAs expressed in ww (Table 3) were 2 to 7 times (and 3 times in lw) higher in tiger shark  
439 embryos than in the mother, while PFOS was not detected in the mother and in 33% of  
440 embryos only. In tiger sharks, embryonic development is ensured by a yolk sac and a high  
441 energy uterine fluid (named the embryotrophe), as no placental connection exists (Castro et  
442 al., 2016). The differences in modes of gestation and mean TLCs in embryos between the  
443 two species (TLCs in tiger shark embryos were 2 times higher than in bull shark embryos ( $p$   
444  $< 0.001$ )) could partly explain the fact that tiger shark embryos were exposed to higher levels  
445 of lipophilic contaminants than bull shark embryos. However, further investigations of  
446 embryos of both species at different stages of gestation would be necessary as both  
447 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  $\log_{10}$ -  
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  $-0.17 \pm 0.08$  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  $\pm 0.5$  %; 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  $31 \pm 4\%$  in her embryos;  
478 Figure 6B). Only PFUnDA and PFTTrDA 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

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

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

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

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

#### 536 **4. Conclusion**

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

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

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

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

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560

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566 **References**

- 567 Aguilar, A., Borrell, A., Reijnders, P.J.H., 2002. Geographical and temporal variation in levels  
568 of organochlorine contaminants in marine mammals. *Mar. Environ. Res.* 53, 425–452.  
569 [https://doi.org/10.1016/S0141-1136\(01\)00128-3](https://doi.org/10.1016/S0141-1136(01)00128-3)
- 570 Alley, E.G., 1973. The Use of Mirex in Control of the Imported Fire Ant 1. *J. Environ. Qual.* 2,  
571 52–61. <https://doi.org/10.2134/jeq1973.00472425000200010005x>
- 572 Alves, L.M.F., Nunes, M., Marchand, P., Le Bizec, B., Mendes, S., Correia, J.P.S., Lemos,  
573 M.F.L., Novais, S.C., 2016. Blue sharks (*Prionace glauca*) as bioindicators of pollution  
574 and health in the Atlantic Ocean: Contamination levels and biochemical stress  
575 responses. *Sci. Total Environ.* 563–564, 282–292.  
576 <https://doi.org/10.1016/j.scitotenv.2016.04.085>
- 577 Barasa, M.W., Lalah, J.O., Wandiga, S.O., 2008. Seasonal variability of persistent  
578 organochlorine pesticide residues in marine fish along the Indian Ocean coast of  
579 Kenya. *Toxicol. Environ. Chem.* 90, 535–547.  
580 <https://doi.org/10.1080/02772240701556312>
- 581 Beaudry, M.C., 2014. Comparative analysis of organochlorine accumulation in two top  
582 predator shark species with contrasting life history traits. University of Windsor.  
583 *Electronic Theses and Dissertations.* 5154. <https://scholar.uwindsor.ca/etd/5154>.
- 584 Beyer, A., Mackay, D., Matthies, M., Wania, F., Webster, E., 2000. Assessing Long-Range  
585 Transport Potential of Persistent Organic Pollutants. *Environ. Sci. Technol.* 34, 699–  
586 703. <https://doi.org/10.1021/es990207w>
- 587 Blard, F., 2006. Les fourmis envahissantes de l'île de la Réunion : interactions compétitives  
588 et facteurs d'invasion. Ph D thesis, Université de la Réunion, 97p.
- 589 Boldrocchi, G., Monticelli, D., Butti, L., Omar, M., Bettinetti, R., 2020. First concurrent  
590 assessment of elemental- and organic-contaminant loads in skin biopsies of whale  
591 sharks from Djibouti. *Sci. Total Environ.* 722, 137841.  
592 <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

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

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

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

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

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

634 Corsolini, S., Pozo, K., Christiansen, J.S., 2016. Legacy and emergent POPs in the marine  
635 fauna of NE Greenland with special emphasis on the Greenland shark *Somniosus*  
636 *microcephalus*. Rendiconti Lincei 27, 201–206. [https://doi.org/10.1007/s12210-016-](https://doi.org/10.1007/s12210-016-0541-7)  
637 [0541-7](https://doi.org/10.1007/s12210-016-0541-7)

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

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

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

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

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

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

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

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

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

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

673 Fisk, A.T., Tittlemier, S.A., Pranschke, J.L., Norstrom, R.J., 2002. Using Anthropogenic  
674 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

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

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

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

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

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

716 Jepson, P.D., Law, R.J., 2016. Persistent pollutants, persistent threats. *Science* 352, 1388–  
717 1389. <https://doi.org/10.1126/science.aaf9075>

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

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

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

727 Kelly, B.C., Ikonomou, M.G., Blair, J.D., SurrIDGE, B., Hoover, D., Grace, R., Gobas,  
728 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](https://doi.org/10.1016/S0048-9697(99)00114-X)

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

760 Logan, R.K., White, C.F., Winkler, C., Jorgensen, S.J., O’Sullivan, J.B., Lowe, C.G., Lyons,  
761 K., 2018. An evaluation of body condition and morphometric relationships within  
762 southern California juvenile white sharks *Carcharodon carcharias*. *J. Fish Biol.* 93,  
763 842–849. <https://doi.org/10.1111/jfb.13785>

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

767 Lyons, K., Adams, D.H., 2015. Maternal offloading of organochlorine contaminants in the  
768 yolk-sac placental scalloped hammerhead shark (*Sphyrna lewini*). *Ecotoxicology* 24,  
769 553–562. <https://doi.org/10.1007/s10646-014-1403-7>

770 Lyons, K., Kacev, D., Preti, A., Gillett, D., Dewar, H., Kohin, S., 2019. Species-Specific  
771 Characteristics Influence Contaminant Accumulation Trajectories and Signatures  
772 Across Ontogeny in Three Pelagic Shark Species. *Environ. Sci. Technol.* 53, 6997–  
773 7006. <https://doi.org/10.1021/acs.est.8b07355>

774 Mackay, D., Fraser, A., 2000. Bioaccumulation of persistent organic chemicals: mechanisms  
775 and models. *Environ. Pollut.* 110, 375–391. [https://doi.org/10.1016/S0269-](https://doi.org/10.1016/S0269-7491(00)00162-7)  
776 [7491\(00\)00162-7](https://doi.org/10.1016/S0269-7491(00)00162-7)

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

780 Marsili, L., Coppola, D., Giannetti, M., Casini, S., Fossi, M., Van WYK, J., Sperone, E.,  
781 Tripepi, S., Micarelli, P., Rizzuto, S., 2016. Skin Biopsies as a Sensitive Non-Lethal  
782 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](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/S0025-326X\(03\)00119-X](https://doi.org/10.1016/S0025-326X(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](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>

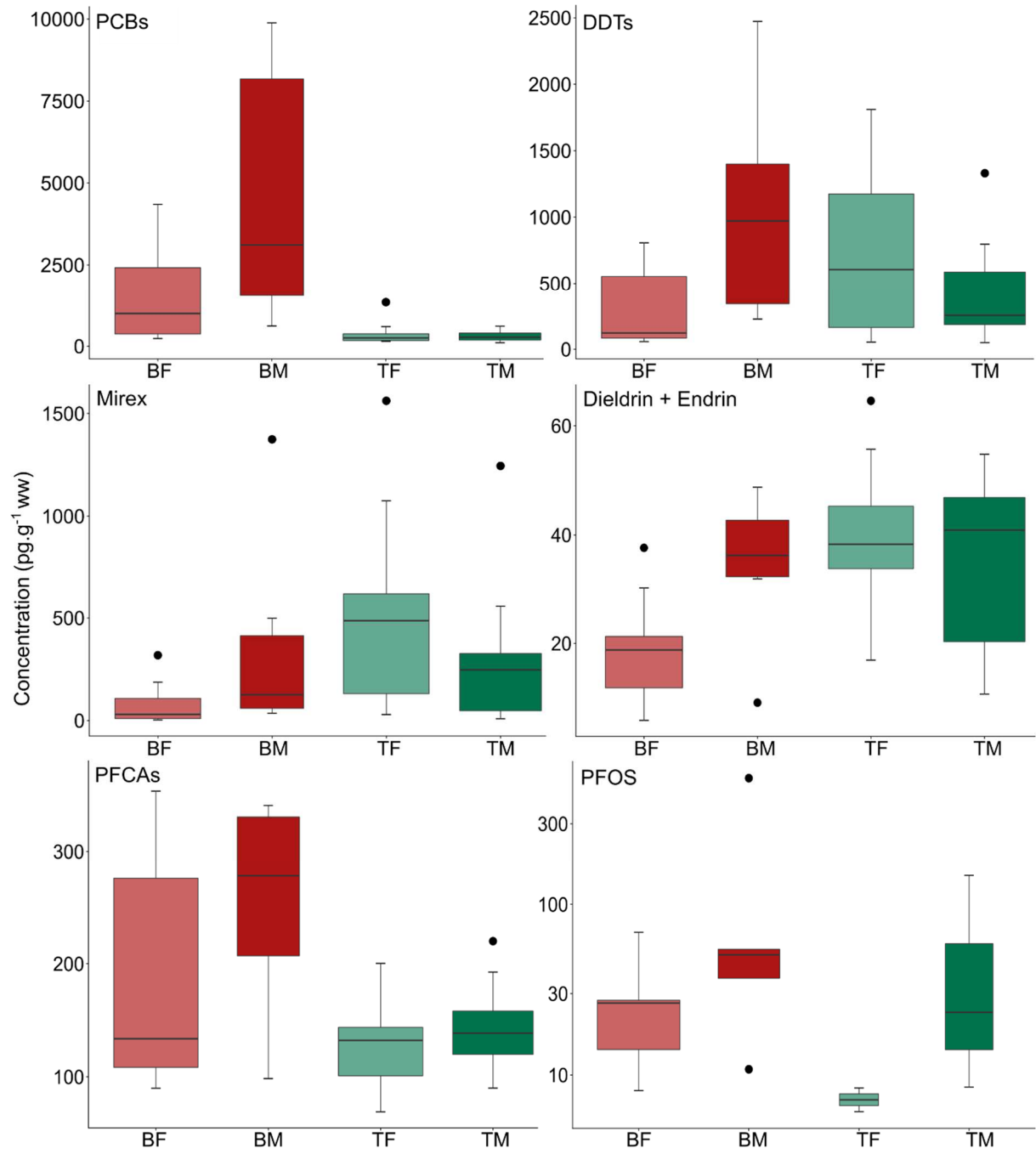
924 Wenzl, T., Haedrich, J., Schaechtele, A., Robouch, P., Stroka, J., 2016. Guidance Document  
925 for the Estimation of LOD and LOQ for Measurements in the Field of Contaminants in  
926 Feed and Food, Publications Office of the European Union. ed, EUR. Luxembourg.  
927 <https://doi.org/10.2787/8931>

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

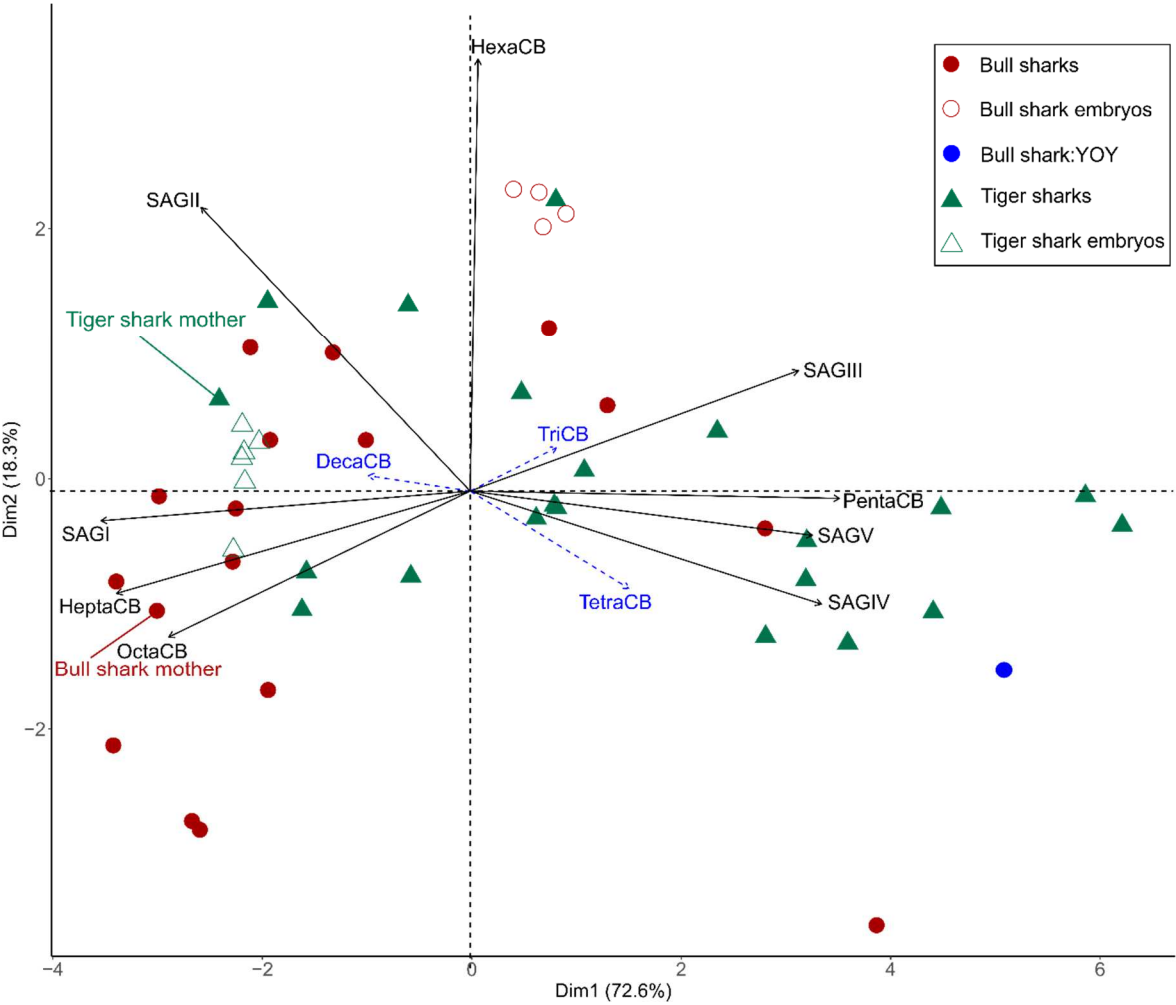
Figure 1: PCB ( $\Sigma$  30 congeners), DDT ( $\Sigma$  5 isomers), mirex, dieldrin+endrin, PFCA ( $\Sigma$  6 compounds) and PFOS concentrations ( $\text{pg g}^{-1}$  ww) in female bull sharks (BF,  $n = 11$ ), male bull sharks (BM,  $n = 6$ ), female tiger sharks (TF,  $n = 11$ ) and male tiger sharks (TM,  $n = 10$ ) collected along the West coast of Reunion Island in 2018-2019. Median values (horizontal solid line inside the box), 25<sup>th</sup> and 75<sup>th</sup> 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.



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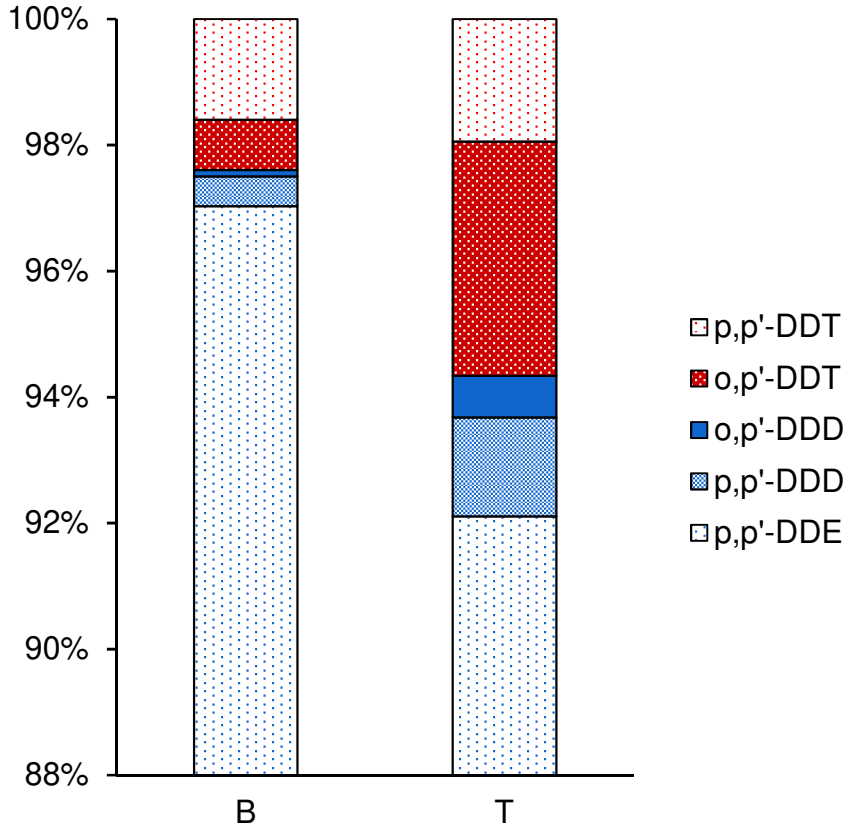


Figure 2: Graph of the two principal components of the standardized PCA on the proportions of congeners grouped according to their chlorine numbers and their structure activity group (SAG) (black arrows) in bull shark (n = 17; red filled circles) and their embryos (n = 4; red empty circles), tiger shark (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.



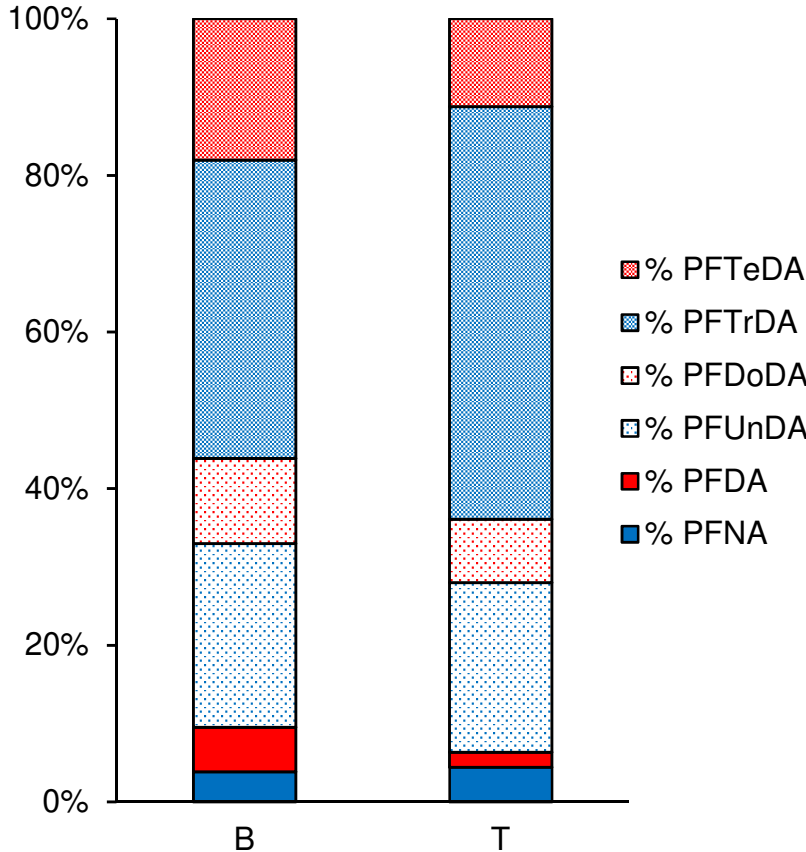
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Figure 1: Mean DDT contamination profiles (% of the  $\Sigma$  5 isomers) in bull sharks (B, n = 17) and tiger sharks (T, n = 21) collected in the West coast of Reunion Island in 2018-2019.



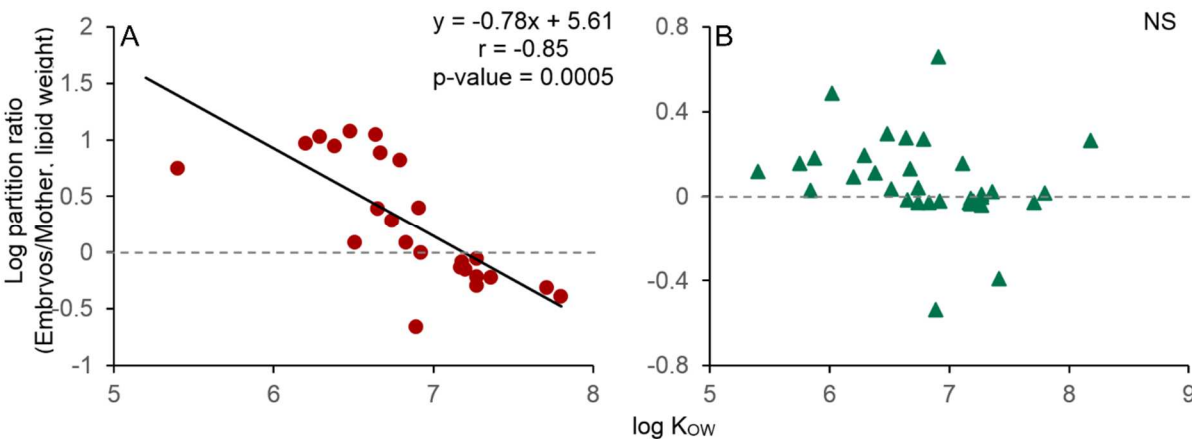
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Figure 1: Mean PFCA contamination profiles in bull sharks (B, n = 17) and adult tiger sharks (T, n = 21) collected along the West coast of Reunion Island in 2018-2019. Odd PFCAs are shown in blue and even PFCAs in red and ranked in ascending carbon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



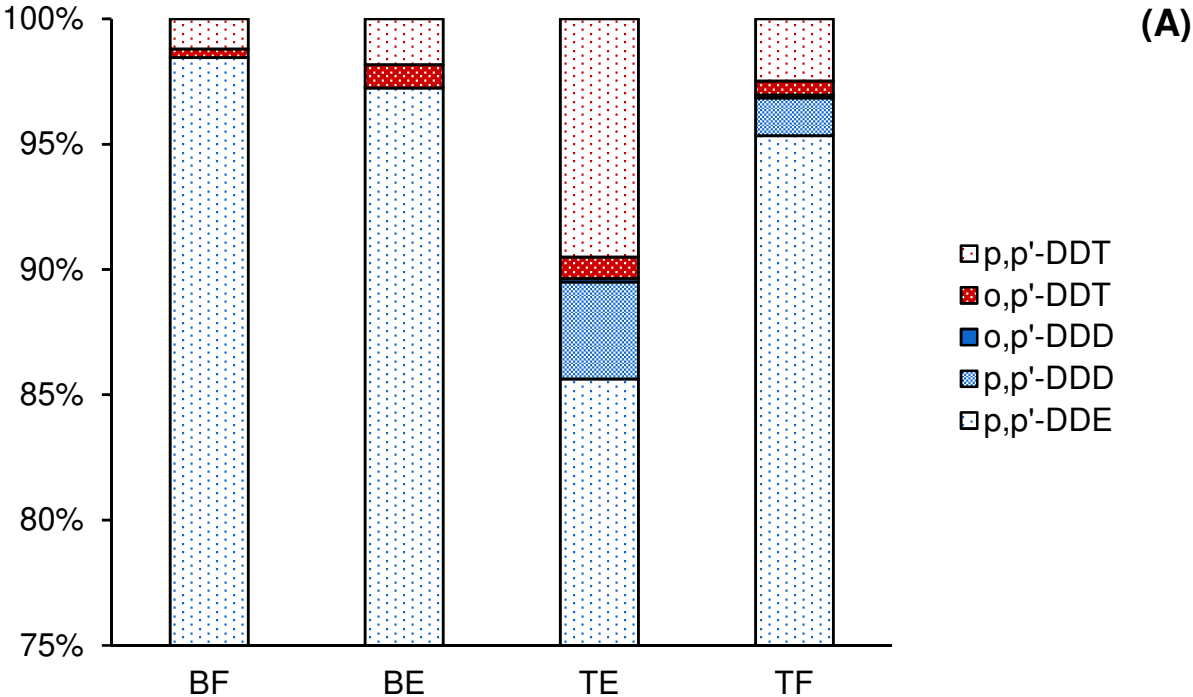
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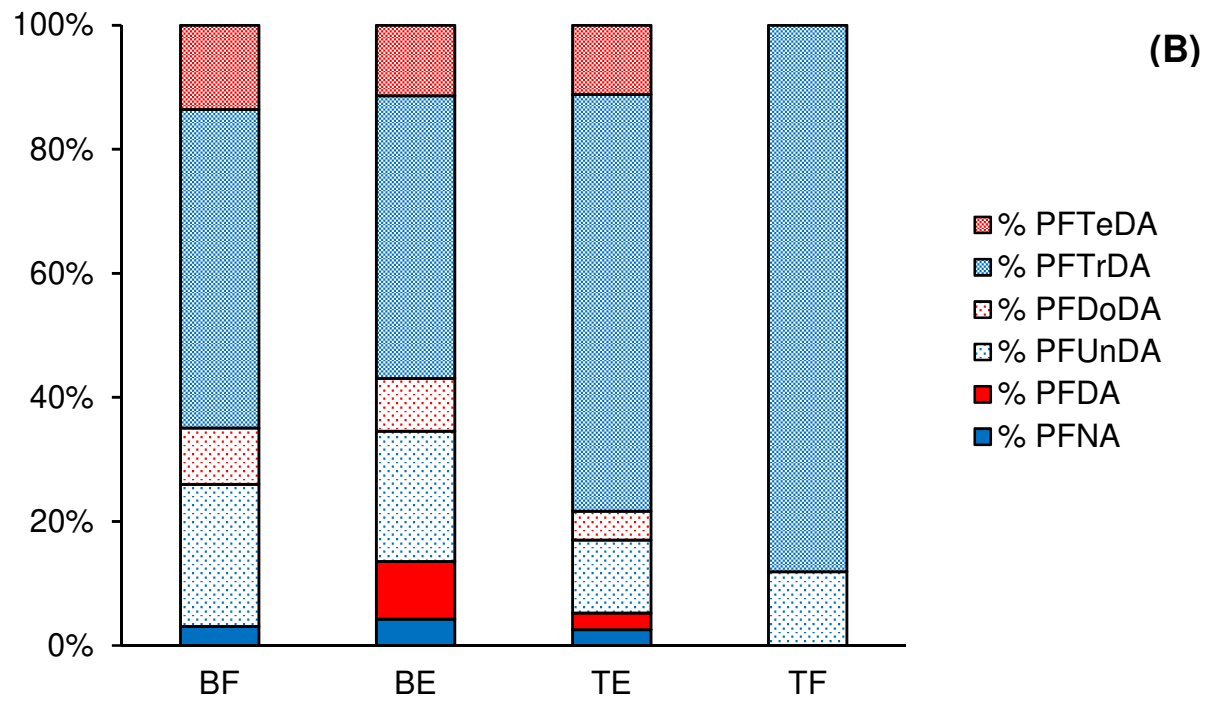
Figure 5: Partition ratios of OCPs and PCBs ( $\text{pg.g}^{-1}$  lipid weight) plotted against log octanol / water partition coefficient ( $\log K_{ow}$ ; Hawker and Connell, 1988; Shen and Wania, 2005) in bull sharks (red circles, A) and tiger sharks (green triangles, B). Partition ratios were calculated 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.





1 column

Table 1 : Biological parameters (total length: TL (cm); body mass (kg); Fulton's condition factor: K ( $\text{g cm}^{-3}$ ); 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  $\pm$  standard deviation (minimum-maximum).

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

\*: 4 pools of 2 individuals

\*\* : 4 pools of 2 individuals and 2 individuals

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



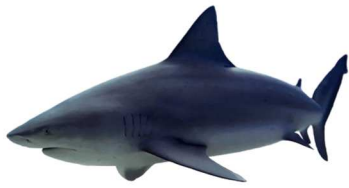
Table 1: PCB ( $\Sigma$  30), DDT ( $\Sigma$  5), mirex, dieldrin-endrin, PFCA ( $\Sigma$  6) and PFOS concentrations ( $\mu\text{g g}^{-1}$  ww) in the white muscle of bull and tiger shark embryos and their mother collected along the West coast of Reunion Island in 2018-2019. Data are expressed as mean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

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

\*: 4 pools of 2 individuals

\*\* : 4 pools of 2 individuals and 2 individuals

## Bull sharks



*Coastal*

++++

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## Tiger sharks



*Neritic*

+

++

+

PCBs

DDTs

PFASs