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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. 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%

(α-endosulfan) to 115 ± 22% (dieldrin) for OCPs and 38 ± 10% (PFTeDA) to 85 ± 10% (PFHxA) for PFASs.

Limits of quantification (LOQs) were calculated for each target compound in each sample using a signal-to-noise ratio of 3 (peak-to-peak) on the less intensive raw data signal (qualifier ion) (Wenzl et al., 2016), taking into account injection volume, the volume of concentrated extract prior to injection and extracted sample weight. LOQs ranged from 0.02 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} ww to 19.0 pg g^{-1} ww for PFASs.

2.5. Statistical analysis

In view of the small number of studied individuals and non-homogeneous nature of dataset variances, statistical analyses were conducted using non-parametric tests. Comparisons of mean contaminant concentrations between species and between sexes of each species were conducted using the Wilcoxon-Mann-Whitney test. The correlation between biological parameters (size, mass, % lipids and K index) and organic contaminant concentrations was assessed with the Spearman rank test. POP profiles were investigated using the Mann-Whitney test and a standardized Principal Component Analysis (PCA) was additionally performed using Euclidean distances for PCBs.

The data were analysed with R software (version 3.4.4). Mean comparisons and correlation tests were conducted with the stat and Hmisc packages (Hollander et al., 2013). PCAs were performed using the FactorMiner package (Husson et al., 2017).

3. Results and discussion

3.1. Biological parameters

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

0.01 kg and 63 ± 1 cm / 1.7 ± 0.1 kg in tiger and bull sharks, respectively. Embryo sizes corresponded to embryo ages of ca. 6 months and 10 months for tiger and bull sharks, respectively (Pirog et al., 2019a, 2020; Whitney and Crow, 2007), i.e. at the third of total gestation time for tiger shark (Pirog et al., 2020) and at the end of gestation for bull shark (Pirog et al., 2019).

As no significant differences were found in any of the four biological parameters (T_L, W, K, TLC) across sexes from either species, both sexes were considered together for morphometric and lipid content comparison between species. Tiger sharks showed a similar average total length and body mass to bull sharks. However, tiger shark maximum size (387 cm versus 327 cm) and mass (402 kg versus 299 kg) exceeded that of bull sharks. Fulton's condition factor mean value was 0.7 times lower in tiger sharks versus bull sharks ($p = 5.10^{-7}$), while TLCs were not significantly different between both species. K is often used as a global indicator of habitat quality or food availability and a positive correlation with lipid content has previously been described in juvenile white sharks (*Carcharodon carcharias*) (Logan et al., 2018). The absence of correlation between K and TLCs in the studied species (tiger sharks: p = 0.45; bull sharks: p = 0.12) may be due to age (large juveniles and adults) or differences in the physiology and dynamics of lipid storage in the two species.

Fish, cephalopods, birds and unidentified remains were found in the non-empty stomachs of bull sharks (64%) and tiger sharks (45%). These observations are coherent with data previously reported by Trystram et al. (2017) and confirm a dominant piscivorous dietary habit for both species, with tiger sharks exhibiting a more generalist foraging behaviour than bull sharks.

3.2. Influence of biological parameters on organic contaminant bioaccumulation

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

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

3.2.1. Chlorinated POPs

TLCs were not correlated with concentrations of the different contaminant families in either species (whether only mature individuals were considered or not), indicating that TLC had a limited influence on chlorinated POP bioaccumulation in white muscle. Chlorinated POP concentrations were therefore primarily compared on a ww basis.

Each PCB congener was quantified in over 60% of shark samples, with the exception of CB-31 and -209, which were detected in 45% of bull shark samples and CB-28, -31, -44 and -49, which were detected in 15%, 15%, 40% and 40% of tiger shark samples, respectively. Among OCPs, the 5 isomers of DDT, mirex and dieldrin were detected in over 90% of samples from both species; endrin was detected in 50% and 75% of bull shark and tiger shark samples, respectively. Contamination levels in adults ranged from 104 to 9885 pg g^{-1} 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 49 pg g^{-1} ww for the sum of dieldrin-endrine (Figure 1). A high inter-individual variability in contamination levels was observed in both species probably due to the generalist foraging behaviour of the two species (Trystram et al. 2017) that lead to a high variety of ingested prey with potential different degrees of contamination. Besides, a wide range of sizes / ages were considered, hence enhancing contamination variability. Organochlorinated compound mean concentrations, on a ww basis, were 3 times higher in male than in female bull sharks (Figure 1), although differences were not statistically significant for PCBs (p = 0.03 and 0.06 for OCPs and PCBs respectively) due to the high variability of contamination levels across individuals (OCP concentrations were 1455.5 ± 1359.7 pg g^{-1} ww in males and 419.6 ± 388.5 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 sharks, combined with their more coastal habitat in RUN (Trystram et al., 2017), indicates exposure of these coastal environments to more significant PCB sources. Conversely, the similar levels of DDT accumulation found in both species suggests similar sources in both coastal (bull sharks) and offshore (tiger sharks) feeding habitats. Among OCPs, mirex concentrations were 2.5 times higher in tiger sharks than in bull sharks (tiger shark: 403 ± 431 pg g^{-1} ww; bull shark: 180 ± 334 pg g^{-1} ww, $p = 0.028$), while dieldrin concentrations were 1.5 times higher in tiger sharks than in bull sharks (tiger shark: 36 ± 14 pg g^{-1} ww; bull shark: 24 ± 14 pg g^{-1} ww, $p = 0.016$). As tiger sharks migrate long distances across the Indian Ocean (Pirog et al., 2019a), these differences suggest that certain insecticides, such as mirex and dieldrin, may be subject to different patterns of use on the Indian Ocean scale, despite the fact that these compounds have been banned in most countries. Although mirex has been detected in air and various matrices in the Indian Ocean, no specific source has been clearly identified (Bouwman et al., 2012; Qiu et al., 2020; Srimurali et al., 2015), while dieldrin may still be in use in Kenya (Barasa et al., 2008).

Only PCBs were significantly correlated to individual size in bull sharks ($p = 0.038$), while PCB, DDT and mirex concentrations showed positive correlations with tiger shark size ($p = 0.0081$, 0.00016 and 0.001 respectively). Contaminant accumulation over fish lifetime reflects a combination of various factors, such as fish growth rate, contaminant accumulation and elimination rates, and reproduction. In sharks, previously-reported results showed species-related relationships between contaminant accumulation and size (Lyons et al., 2019).

3.2.2. PFASs

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

31.9 ± 34.1 pg g^{-1} ww, one male bull shark exhibiting a higher concentration of 555 pg g^{-1} ww was excluded), while ∑ PFCA concentrations were in the 69.0 - 353.5 pg g^{-1} ww range, with mean concentrations of 135.0 ± 38.2 pg g^{-1} ww in tiger sharks and 212.0 ± 97.8 pg g^{-1} ww in bull sharks.

PFOS and ∑ PFCA concentrations were not statistically different between sexes in either species ($p = 0.12$ and 0.24 respectively for bull shark; PFOS not tested for tiger sharks and $p = 0.39$ for ∑ PFCAs). Unlike organochlorine compounds, PFASs did not show higher concentrations in male bull sharks ($p = 0.32$); this could indicate that female contamination by these compounds is less influenced by reproduction and mode of gestation versus PCBs and OCPs. PFAS concentrations were significantly higher in bull sharks versus tiger sharks and differences were significant for each individual compound ($p = 0.00051$ to 0.02), with the exception of PFNA and PFTrDA ($p = 0.051$ and 1 respectively). The higher PFAS levels found in bull sharks suggest that their sources may be local rather than distant. High inter-individual variability was observed (a factor of 5 between minimum and maximum PFAS concentrations), as expected in view of the wide range of sizes / ages. However, no significant correlation was found between PFAS concentrations and fish sizes in the two species.

3.3. Contaminant profiles and ratios as tracers of contamination sources

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

3.3.1. PCBs

PCBs can be classified according to the number and position of chlorine atoms, which affect their hydrophobicity (Hawker and Connell, 1988) and propensity to be metabolized. Chlorine atom position defines five structure-activity groups (SAGs), characterizing congener metabolization capability (Boon et al., 1997; Yunker et al., 2011). Groups I and II are not metabolized, group III is metabolized in some mammals but not in fish, and groups IV and V are metabolized in some fish (Buckman et al., 2007).

To assess contamination profiles, a standard PCA (Figure 2) was performed on all samples except the young-of-the-year bull shark, which was included for information purposes only (additional individual). In this analysis, the two first components representing 90.9% of total variance were used according to the Kaiser criterion (component eigenvalue > 1, Figure 2). Additional variables were groups poorly-represented in the factorial plan of the first two components (inertia of the variable in a component < mean inertia of that component).

PCB congeners predominating in bull sharks were higher chlorinated (7 - 8 chlorine atoms) and non-metabolizable (SAG I), while tiger sharks were characterized by a higher variability in their profiles (Figure 2), corresponding to a higher diversity of tiger shark prey in RUN (Trystram et al., 2017). Univariate analyses confirmed that the relative abundance of these compounds differed significantly between the two species ($p = 0.00064$ and $p = 0.012$ for 7 and 8 chlorine PCBs respectively). The PCB contamination profile of the young-of-the-year bull shark was completely different to that of the other bull shark individuals.

PCBs exhibited a contamination profile dominated by higher chlorinated and non-metabolizable congeners in bull sharks. As the heavier congeners are less mobile and bind to organic matter near the coast, leading to preferential accumulation of these compounds in food webs close to their sources (Salvadó et al., 2013), bull sharks would appear to be more highly-impacted by local, coastal PCB sources. This surprising result, in view of the island's low level of industrialization, is corroborated by the study of Dirtu et al. (2016), which showed higher PCB levels in coastal Indo-Pacific bottlenose dolphins (*Tursiops aduncus*) than in spinner dolphins (*Stenella longirostris)* living further offshore in RUN. Conversely, tiger sharks were predominantly impacted by lower chlorinated PCBs, which can be transported 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 ($C \geq 8$), 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.

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

3.4.2. Contamination profiles

PCB profiles were similar in the tiger shark mother and embryos (Figure 2), while higher relative concentrations of *p,p'*-DDE were observed in the mother (mother: 95%; embryos: 86 ± 0.5 %; Figure 6A). Conversely, hexachlorinated congeners belonging to SAG III were less-prevalent in the bull shark mother than in embryos (Figure 2), while similar DDT profiles were observed (Figure 6A). The (*p,p'*-DDE + *p,p'*-DDD) / *p,p'*-DDT ratio was respectively 2 and 4 times higher in bull and tiger shark mothers than in their embryos, suggesting that DDT transfer and accumulation in embryo muscle was isomer-specific. Similar PFAS profiles were found in the bull shark mother and embryos, with the exception of PFOS, which showed higher relative concentrations in embryos (9% in the mother and 31 ± 4% in her embryos; Figure 6B). Only PFUnDA and PFTrDA were detected in the tiger shark mother, whereas PFASs were detected in 100% of embryos, with the exception of PFOS and PFDoDA detected in 33% and 83% of embryos, respectively. Our results show that the different molecule families have different fates in the two studied species, suggesting that pollutant family and mode of gestation (known to be different in the two species) are important factors in the maternal transfer of organic contaminants.

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

Large inter-individual variations in organic contaminant concentrations in sharks have been found in previous studies worldwide (Table S2 and S3). Various factors, such as size, sex and lipid levels of studied individuals, which are not always reported, the number of compounds considered, the decrease in legacy POP concentrations and potential increase in CECs over time (Tanabe and Ramu, 2012) could explain these variations.

Sharks sampled in various locations in the Southern Hemisphere have shown PCB concentrations between 2 and 100 times higher than sharks from RUN (Cagnazzi et al., 2019; Marsili et al., 2016). A pregnant bull shark and blacktip reef sharks (*Carcharhinus melanopterus*) sampled in Australia and on the east coast of South Africa showed concentrations similar to those of RUN bull sharks, but 10 times lower than those of tigers sharks (Beaudry, 2014; Cagnazzi et al., 2019). PCB and DDT concentrations reported in another top predator species (albacore tuna *Thunnus alalunga*) from the coast of South Africa were lower than those found in RUN due to higher industrialization and urbanization in South Africa (Dirtu et al., 2016; Munschy et al., 2016; Mwevura et al., 2010). DDT concentrations were 5 to 100 times higher in sharks from the Southern Hemisphere oceans than RUN (Beaudry, 2014; Cagnazzi et al., 2019; Marsili et al., 2016; Schlenk et al., 2005). 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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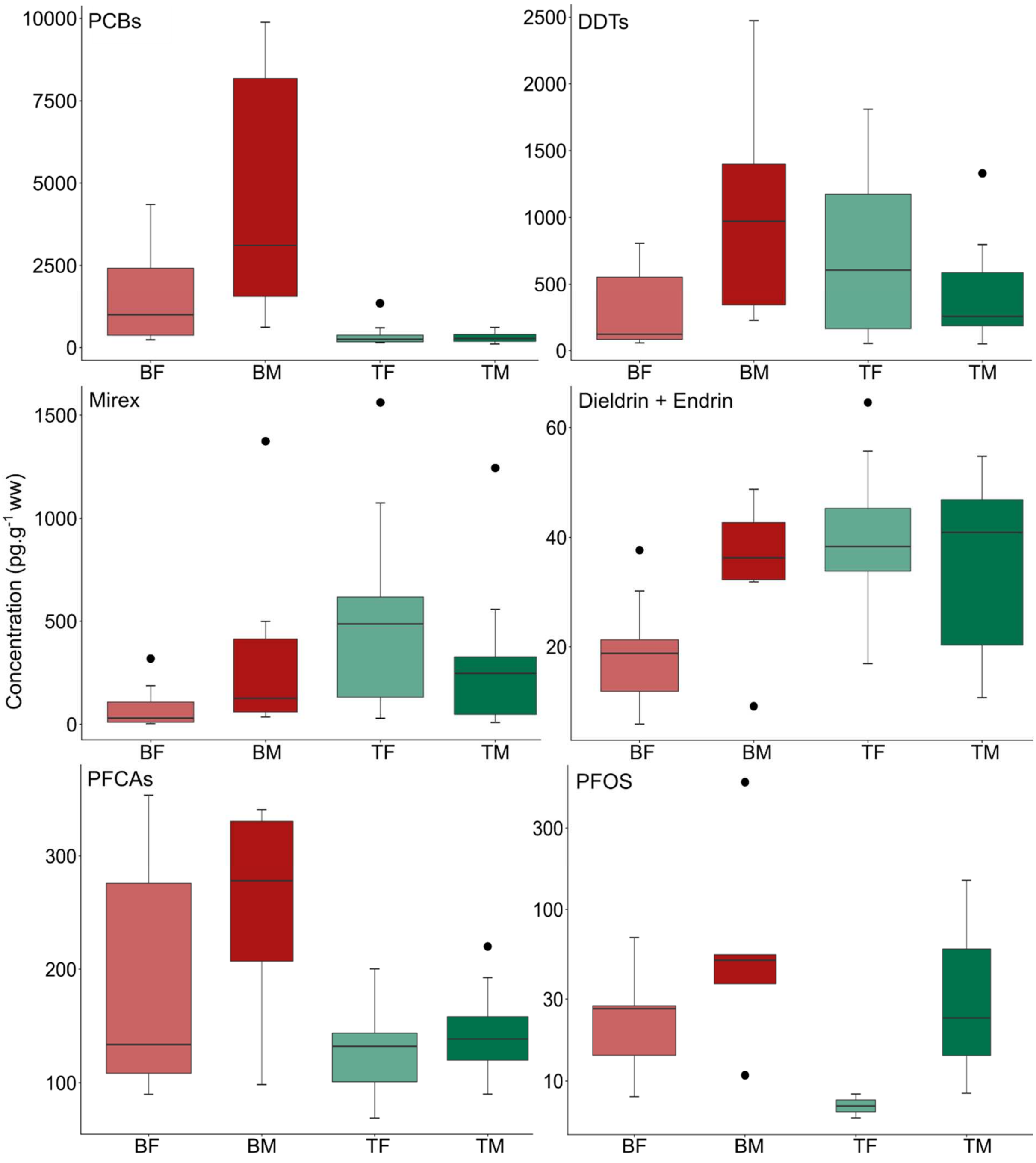
Figure 1: PCB (Σ 30 congeners), DDT (Σ 5 isomers), mirex, dieldrin+endrin, PFCA (Σ 6 compounds) and PFOS concentrations (pg g^{-1} ww) in female bull sharks (BF, n = 11), male bull sharks (BM, n = 6), female tiger sharks (TF, n = 11) and male tiger sharks (TM, n = 10) collected along the West coast of Reunion Island in 2018-2019. Median values (horizontal solid line inside the box), 25th and 75th 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
10000
7500
5000
2500
0
DDTs
2500
2000
1500
1000
500
0
Mirex
1500
1000
500
0
Dieldrin + Endrin
60
40
20
PFCAs
300
200
100
PFOS
300
100
30
10
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.

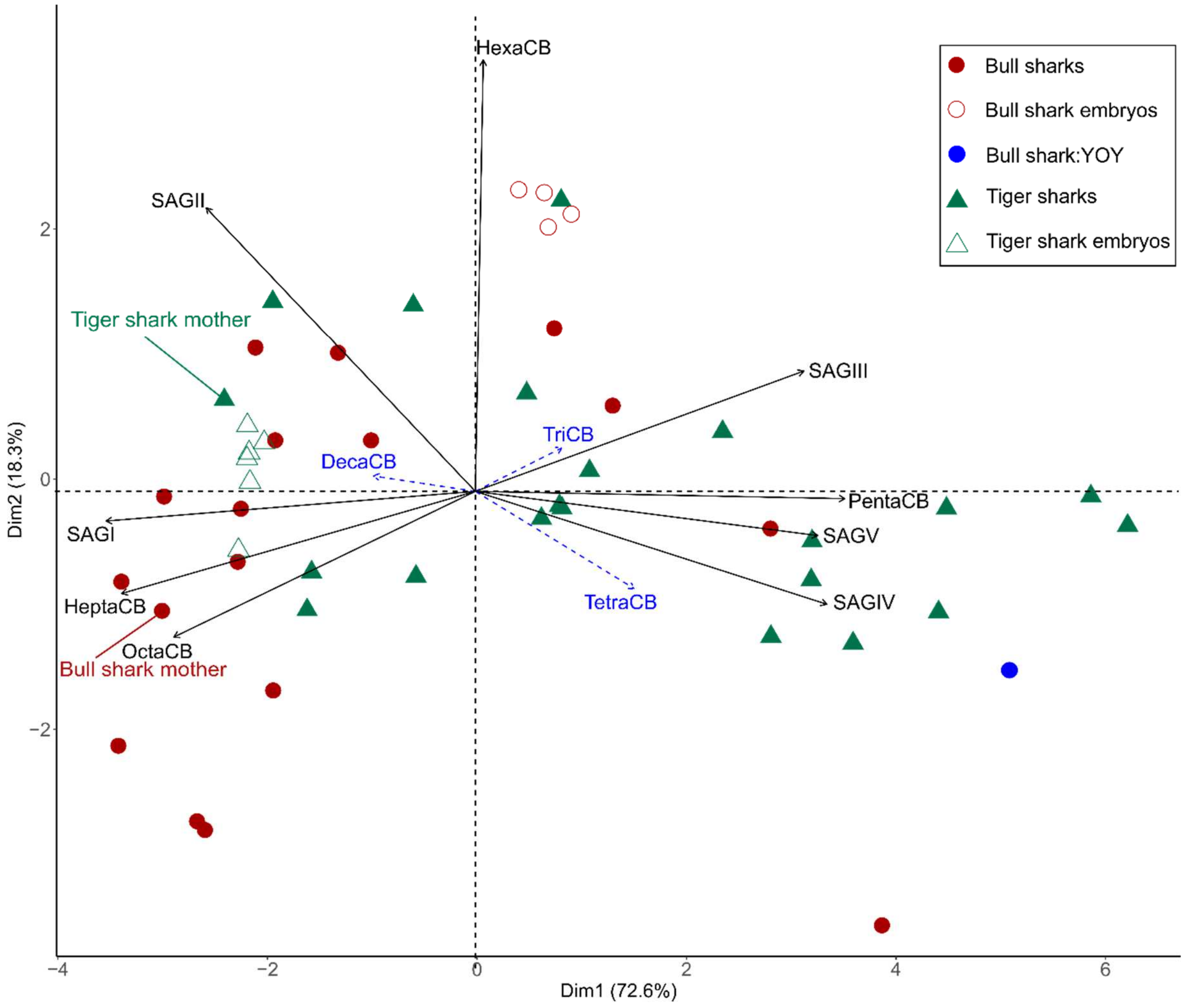


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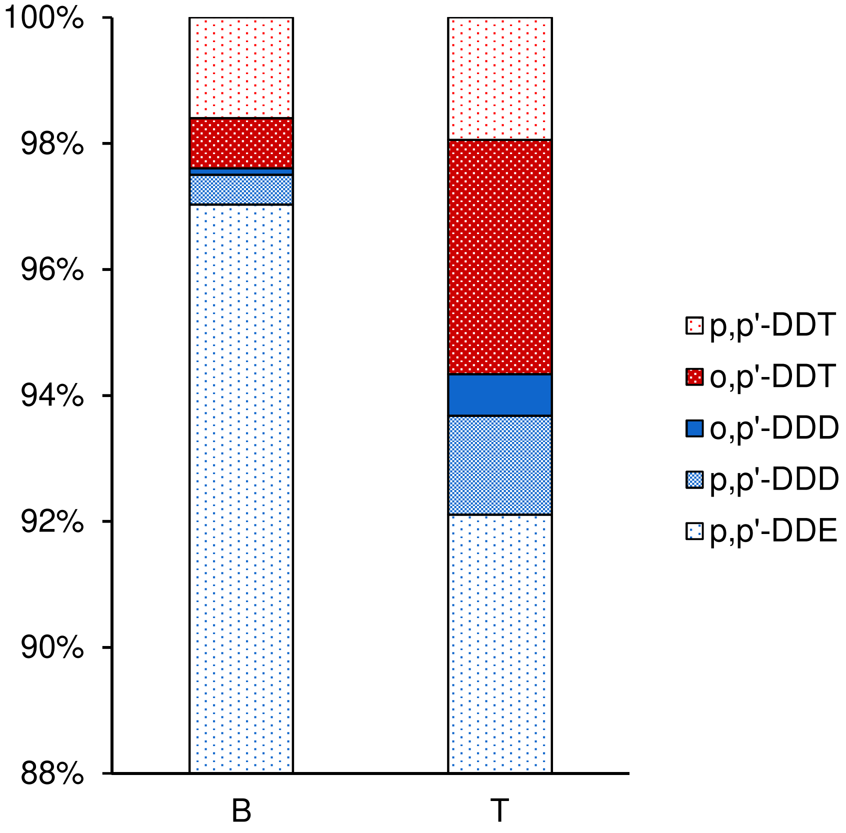
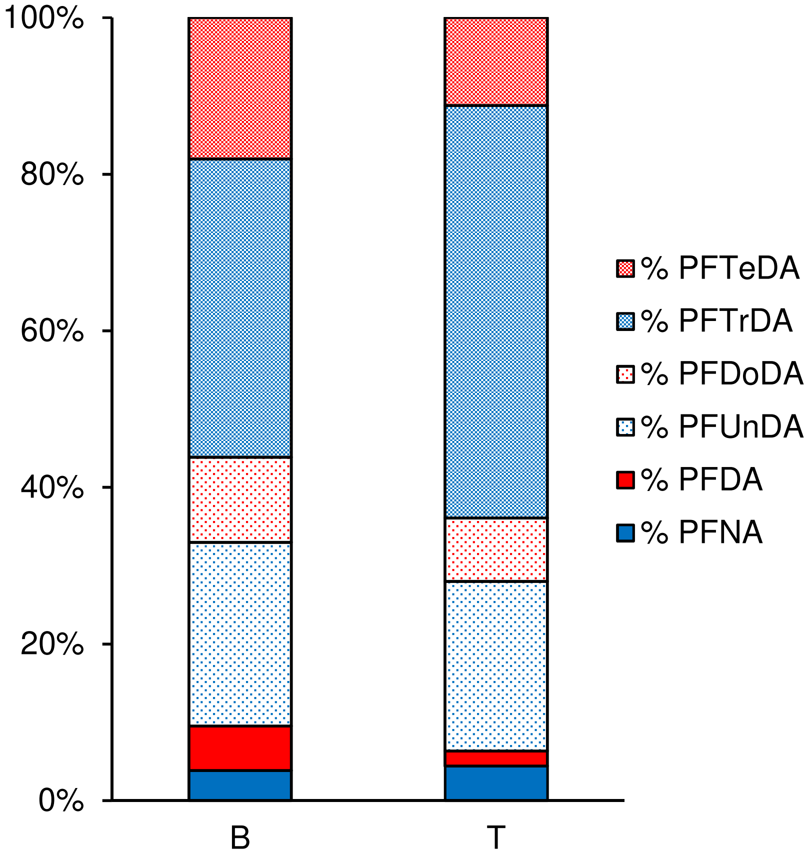
Figure 1: Mean DDT contamination profiles (% of the ∑ 5 isomers) in bull sharks (B, n = 17) and tiger sharks (T, n = 21) collected in the West coast of Reunion Island in 2018-2019.



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.

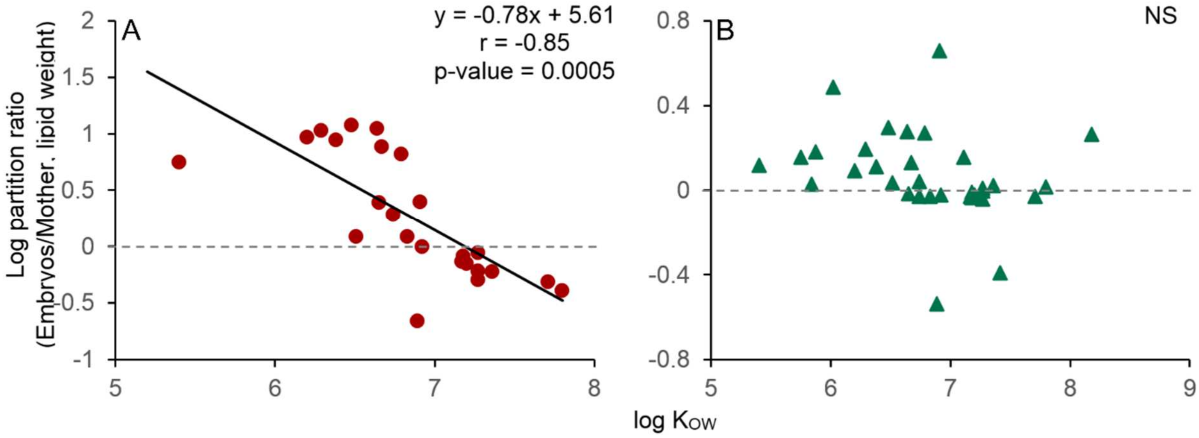


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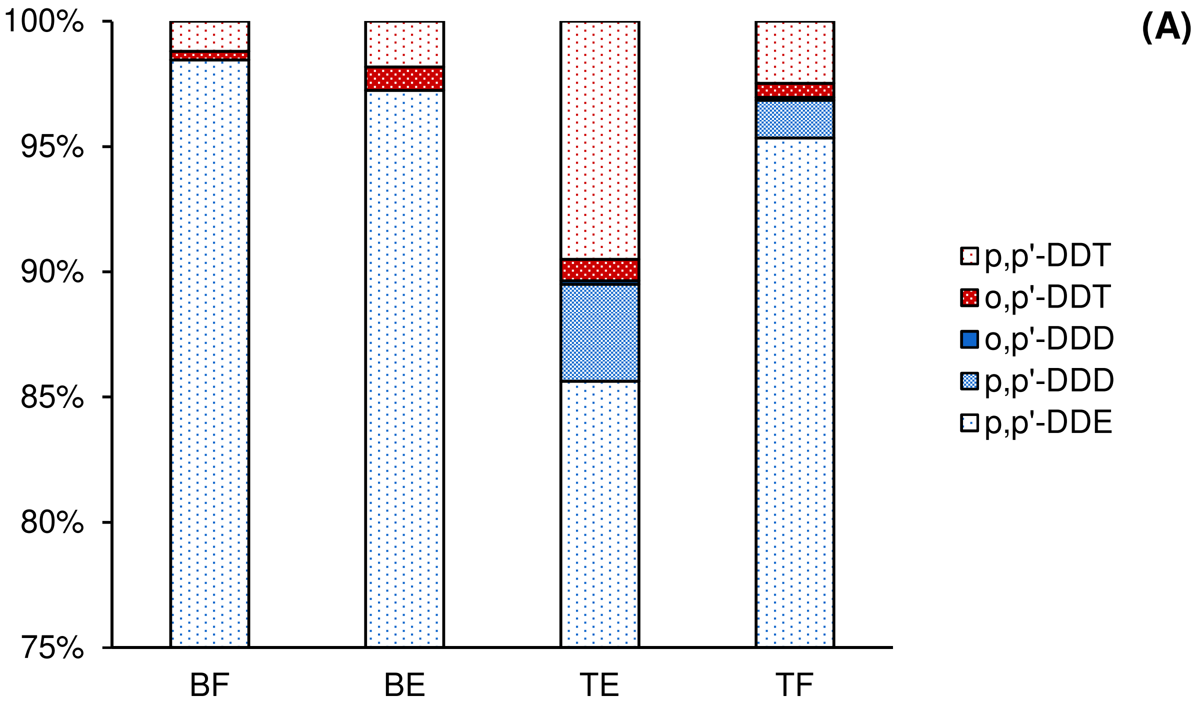
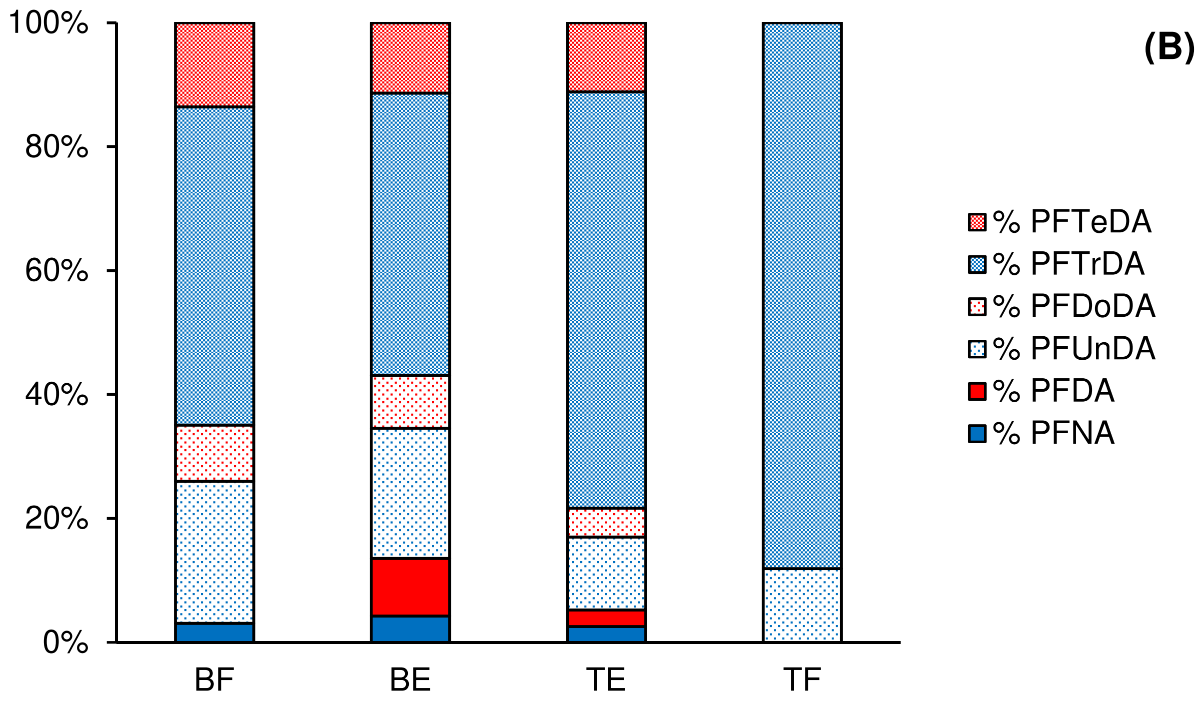
Figure 6: Mean contamination profiles of DDT (A) and PFCAs (B) in bull shark mother (BF, n = 1), bull shark embryos (BE, n = 4), tiger shark embryos (TE, n = 6) and tiger shark mother (TF, n = 1) collected along the West coast of Reunion Island in 2018-2019. Odd PFCAs are shown in blue and even PFCAs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




100%
80%
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BE
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(B)
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% 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

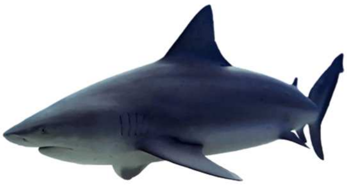
**: 4 pools of 2 individuals and 2 individuals

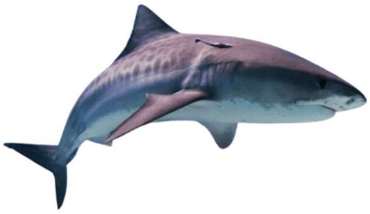
Table 1: PCB (Σ 30), DDT (Σ 5), mirex, dieldrin-endrin, PFCA (Σ 6) and PFOS concentrations (pg g^{-1} ww) in the white muscle of bull and tiger shark embryos and their mother collected along the West coast of Reunion Island in 2018-2019. Data are expressed as mean ± standard deviation (minimum - maximum).

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

*: 4 pools of 2 individuals

**: 4 pools of 2 individuals and 2 individuals





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