Legacy and emerging organic contaminants: Levels and profiles in top predator fish from the western Indian Ocean in relation to their trophic ecology

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1. Introduction

Apical marine fish predators represent a major marine resource and play an essential role in marine ecosystems. In addition, they are of high economic value to industrial and artisanal fisheries and local populations due to their nutritional benefits (proteins, essential minerals, vitamins and polyunsaturated fatty acids) (Sioen et al., 2009). However, due to their position at the top of trophic webs, coupled with a long lifespan, they are prone to accumulating high levels of various contaminants, including toxic ones such as Persistent Organic Pollutants (POPs). POPs are defined by their persistent, bioaccumulative, toxic properties and propensity to travel far from their emission sources, hence leading to global distribution (Wania and Mackay, 1996; Lohmann et al., 2007; Rigét et al., 2016). They are listed under the United Nations Stockholm Convention, which came into force in 2004 and aims to protect humans and the environment from chemicals. The POPs reported in this study include polychlorinated biphenyls (PCBs), organochlorinated pesticides (OCPs) (namely, dichlorodiphenyl-trichloroethane –DDT and its isomers, hexachlorobenzene –HCB, hexachlorocyclohexanes –HCHs, mirex, dieldrin, aldrin and endosulfans) and perfluorooctane sulfonate (PFOS). Although these POPs have been phased-out in many countries, especially in the Northern Hemisphere (Kalantzi et al., 2001; Zitko, 2003; Barber et al., 2005; Breivik et al., 2007; Vijgen et al., 2011; Land et al., 2018), their use is still reported in developing countries such as Asia, Africa and South America and secondary emissions persist (Breivik et al., 2011; Ali et al., 2014; Bouwman et al., 2015), emphasizing the need to investigate further their occurrence in the marine environment. Besides, long-chain perfluorocarboxylic acids (PFCAs) have also been investigated as contaminants of emerging concern: they share similar properties to POPs and show an increasing production in Asia together with their precursors (Wang et al.,

2014) ; in addition, oceans are recognized as PFAS main reservoir (Johansson et al., 2019), which overall raise concern on their impact on marine ecosystems.

The tropical tuna species, namely, skipjack -SKJ- (*Katsuwonus pelamis*), yellowfin -YFT- (*Thunnus albacares*), bigeye -BET- (*Thunnus obesus*) tunas, and the billfish swordfish -SWO- (*Xiphias gladius*), were investigated for organic contamination. These four large pelagic top predator fish occur in all oceans but do not share similar trophic niches, as they feed at different depths of the water column (Ménard et al., 2007; Sardenne et al., 2016). They are known to be highly-migratory species that travel long distances. However, during the foraging and spawning season, they demonstrate seasonal resident behaviour at the temporal scale of their muscle turnover rate (i.e. months, Madigan et al., 2012; Pethybridge et al., 2018). For several years, POPs in top predator fish have been used as time-integrated tracers of marine environmental contamination (Ueno et al., 2005; Hart et al., 2008). Used in conjunction with trophic and ecological parameters such as stable isotopes of carbon ($\delta^{13}C$) and nitrogen ($\delta^{15}N$), POPs have proved to be suitable tracers of foraging habitats and trophic transfers in marine ecosystems (Dickhut et al., 2009; Hisamishi et al., 2010; Deshpande et al., 2016; Dirtu et al., 2016; Chouvelon et al., 2017).

The Indian Ocean is the world's third largest water body and a major production area for tuna and billfish fisheries, in particular in the Western part (Pons et al., 2017; Chassot et al., 2019). However, to date, very few studies have focused on the contamination of its ecosystems by organic pollutants (Bouwman et al., 2012; Munschy et al., 2016). The Western Indian Ocean (WIO) is characterized by asymmetric oceanic currents (North or South of 10°S latitude) and a seasonal effect strongly influenced by the monsoon regime. These specific water circulation regimes could, in turn, influence contaminant origins and transport in the WIO.

The main objectives of this study were to i) determine the contamination levels of selected legacy and emerging organic contaminants in four large pelagic top predator fish representing key species of the WIO ecosystem, ii) evaluate the influence of biological (size / age, sex) and trophic factors on organic contaminant bioaccumulation and iii) investigate differences in spatial contamination patterns (based on specific ratios of chemical tracers) at a regional scale that could be linked to different contamination sources. To the best of our knowledge, the data presented in this report are among the first obtained for various organic contaminant families in various large pelagic top predator fish species with such a large geographical distribution.

2. Materials and Methods

2.1. Sampling

Ethical approval was not required for this study, as all fish were collected as part of routine professional fishing practises.

The investigated areas (Fig. 1) in the WIO included the Seychelles exclusive economic zone (SEY), off the coasts of Somalia (SOM), the Mozambique Channel (MOZ) and an area located offshore between SEY and the Chagos archipelago (hereafter CHAG). SWO were only collected in SEY.

In total, 27 BET, 24 SKJ, 24 YFT and 18 SWO collected in 2013-2014 were analysed for organic contaminants. The white muscle of male individuals was analysed in all species from MOZ, SOM and CHAG, and the white muscle of both male and female individuals was analysed in all species collected from SEY. Although the fish were collected in different seasons and years, which could lead to variations in contaminant concentrations, the effect of collection time on results was not studied due to the limited number of samples available in each season.

Fish size (L in cm), corresponding to tuna fork length (from the tip of the snout to the fork of the tail) and swordfish lower jaw fork length (from the lower jaw to the fork of the tail), mass (W in kg) and sex (indeterminate, I; female, F; male, M) were recorded for each individual. Individuals were selected to cover a large size range. Fulton's Condition Factor (K), as a proxy of relative fish fitness, was calculated as: $K = W \times 100{,}000) / L^3$, whereby L is fish size in cm and W is fish mass in kg.

Sub-samples of frozen fish of around 5 cm^3 were taken from the front dorsal white muscle (sampled under the dorsal spine on the left side) of each fish, and stored in amber glassware at -20°C prior to freeze-drying and further analysis of organic contaminants and total lipid content.

2.2. Sample treatment and analysis

Chemicals and sample analytical procedures for PCBs, OCPs and PFASs have been described previously (Munschy et al., 2016; Munschy et al., 2019). Details on the chemicals used and additional information are provided in the Supplementary Material (SM).

2.2.1. Stable isotopes

Stable isotope analyses were performed according to Bodin et al. (2009). Briefly, 1-2 mg of freeze-dried samples were ground and extracted with dichloromethane. Lipid-free samples were dried under a fume hood for 48h and packed in tin-capsules. Around 0.4 ± 0.08 mg of each lipid-free sample was then analyzed for stable isotopes using a Delta V Advantage isotope ratio mass spectrometer interfaced to a Flash EA 1112 elemental analyzer with a Conflo IV interface (Sardenne et al., 2015). The results were reported in the δ unit notation and expressed per mil (‰) relative to international standards (Vienna- Pee Dee Belemnite for

carbon and atmospheric N2 for nitrogen). Estimates using replicate measurements of internal laboratory standards (USGS-24, IAEA-CH6,-600 for carbon; IAEA-N2,-NO-3, -600 for nitrogen) gave an analytical variability of less than 0.15‰ for both $\delta^{13}C$ and $\delta^{15}N$.

2.2.2. Total lipid content (TLC) analysis

An aliquot of each freeze-dried sample (0.5 g dw) used for organic contaminant analysis was extracted with a mixture of hexane / acetone (80:20) at 100 °C under 100 bars using accelerated solvent extraction (ASE) with Dionex ASE 200 (ASE, Dionex Corp., USA). The extracts were evaporated to dryness at 105°C for 12 hours to determine total lipid content (TLC) and expressed in % of wet weight (ww).

2.2.3. Organic contaminant analysis

Contaminant concentrations were calculated in pg g^{-1} or ng g^{-1} wet weight (ww), with a mean humidity percentage of 72% (calculated on all samples). Concentrations were converted into ng g^{-1} lipid weight (lw) using the TLC determined in each sample.

PCB and OCP analysis

Two to ten grams of the freeze-dried muscle samples were extracted with DCM using accelerated solvent extraction (ASE) with a Dionex ASE 300 (ASE, Dionex Corp., USA). 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 column, and a 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). The samples were analyzed for 34 PCBs, ranging from trichlorinated to decachlorinated congeners and including the 12 dl-PCBs (CB-77; -81; -105; -114; -118; -123; -126; -156; -157; -167; -169; -189) and the 6 i-PCBs (CB-28; -52; -101; -138; -153; -180), 5 DDTs (*p,p'*-DDT, *o,p'*-DDT, *o,p'*-DDD, *p,p'*-DDD, *p,p'*-DDE; hereafter, the term "DDTs" refers to the sum of these 5 compounds), hexachlorobenzene (HCB) and hexachlorocyclohexanes (HCHs). In addition, aldrin, dieldrin, endrin, isodrin, mirex and endosulfans (α-, β-endosulfan isomers and endosulfan-sulfate) were analysed in tuna samples.

PCB and OCP analyses were performed by gas chromatography coupled to 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 a Micromass AutoSpec Ultima mass spectrometer. 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-labelled CB-70, -111 and -170, d_8-labelled *p,p'*-DDD and *o,p'*-DDT was added to the final purified extracts for signal correction.

PFAS analysis

PFAS analysis was conducted according to Munschy et al. (2019). 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, Couderc et al., 2015), evaporated to dryness and reconstituted in 200 µL of a mixture of MeOH:H_2O (50:50, v/v).

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 C_{18} reversed-phase column eluted with ammonium acetate in water (20 mM) (A) and methanol (B).

2.4. QA/QC

QA / QC procedures were followed during each sequence analysis, i.e. 10 samples for PCBs and OCPs and up to 20 samples for PFASs, to which one procedural blank and one quality control (as described in the SM QA/QC section) were added. In order to minimize external and cross-contaminations, all samples were processed in a clean laboratory (low dust and positive pressure) under a fume hood. QA / QC procedures included the analysis of certified material, in-house control samples, blanks and participation in interlaboratory comparison tests for the marine environment. LOQ values were determined for each target compound in each analyzed sample according to Wenzl et al. (2016), i.e., using a signal-to-noise ratio of 3 (peak-to-peak) for the less intensive raw data signal (qualifier ion). Detailed information can be found in the SM (Table S1).

2.5. Statistical analysis

Statistical analyses were performed using StatSoft Statistica software v 7.1.

Correlations (e.g. between total lipid content and organic contaminant concentrations) were tested using simple linear regression coefficients. In view of the small sample size per group, data comparisons (biological and trophic parameters, organic contaminant concentrations and ratios) across fish groups were preferably performed using non-parametric tests (Mann–Whitney test for comparison of two independent samples, or one-way ANOVA Kruskal-Wallis's test), with a significance level (α) of 0.05. Results were considered to be significant only when both tests gave significant results. Statistical analyses were conducted on summed or individual compounds detected in at least 50% of the samples.

3. Results and discussion

3.1. Biometric parameters

Out of all the studied fish species (BET from MOZ excluded, see below), SKJ was the smallest in size, with a mean FL of 57.6 ± 7.6 cm. This species is classified as a small tuna (≤ 100 cm). The largest fish was SWO (166.8 ± 28.6 cm), which is classified as one of the largest pelagic fish species (Vanpouille et al., 2001). BET (with the exception of those from MOZ) and YFT average FLs across all areas were > 100 cm (i.e. 131.0 ± 26.3 cm and 121.5 ± 28.8 cm, respectively); these species are considered as large tunas (Sardenne et al., 2016). All BET from MOZ were of indeterminate sex with lower fish size (52.9 ± 3.5 cm) and mass (3.4 ± 0.6 kg) versus other areas (Table 1). Age was estimated using growth curves from Dortel et al. (2015) for YFT, from Eveson et al. (2015) for BET and SKJ, and from Farley et al. (2016, 2019) for SWO. Calculations gave estimated ages ranging from under 1 year to 10 years for tropical tuna species (mean values of 3.2 ± 2.5 years, 5.1 ± 3.2 years and 4.8 ± 3.1 years for SKJ, BET and YFT, respectively) and from 2 to 20 years (mean value of 6.5 ± 4.6 years) for SWO (Table 1). Although inter-species differences in fish sizes were significant (except between BET and YFT), only SKJ and SWO showed significant ($p < 0.05$) age differences (SKJ being younger than SWO). Regardless of species, no differences in fish size or age were found between male and female individuals sampled in SEY. Regarding tuna species, significant differences in fish size and age were observed across the sampling areas for BET and YFT, with smaller and younger individuals of both species in MOZ versus other areas.

3.2. TLC

TLCs were low and similar in the three tropical tuna species, ranging from 0.13 to 1.11 % ww (Table 1), with values similar to those reported in Sardenne et al. (2016), i.e. from 2.0 ± 1.0 % dw to 3.1 ± 3.5 % dw versus 1.3 ± 0.7 % dw to 1.8 ± 1.1 % dw in our study. SWO and SKJ

showed the highest (9.4 ± 5.7% ww) and lowest (0.37 ± 0.19% ww) TLCs of all species, respectively. No TLC differences were identified between males and females of any species collected in SEY, or between tuna species across the study areas.

3.3. Stable isotopes

$\delta^{15}N$ and $\delta^{13}C$ values (means and standard deviations, Table 1) are shown in Figure 2 for the various fish species and sampling areas.

$\delta^{13}C$ values were significantly higher in SWO than in tunas overall, although the difference between SWO and YFT was not significant ($p = 0.09$). SWO hence appears to rely on specific carbon sources, different to those of the three tropical tuna species. The only significant difference found across areas was between fish from MOZ (lower $\delta^{13}C$) and SEY. $\delta^{15}N$ values were significantly higher in SWO and BET versus SKJ and YFT, in agreement with previously-reported data (Olson et al., 2016). Globally, tunas from MOZ were characterized by lower $\delta^{15}N$, confirming the reports of Sardenne et al. (2016), while fish from the other studied areas had similar levels.

3.4. Organic contaminant levels and profiles

3.4.1. Organic contaminant levels in tropical tunas and swordfish

Concentrations of the various families of studied organic contaminants in tunas and swordfish are presented in Fig. 3 and detailed results are given in Table 2.

Globally, organic contaminant concentrations in tropical tuna species ranked in the order PFAS > OCP > PCB, while in SWO, they ranked in the order DDT > PCB = PFAS (Figure 3). The variations in organic contaminant distribution across species could be explained by species biochemical composition and organic contaminant affinities. As a lipid-rich species,

SWO accumulates lipophilic contaminants, such as PCBs and OCPs. Conversely, the studied tropical tunas are lean species with higher protein contents (Sardenne et al., 2016) for which PFASs have a greater affinity.

The effect of fish age on POP concentrations was investigated in each species and generally showed that contaminant concentrations were not significantly correlated to fish age. The only significant correlations (positive) were found for PFCAs in both SKJ and YFT and for PCBs in YFT. A lack of significantly higher contaminant levels with increasing size / age has already been reported previously and might reflect complex interactions between fish growth rate and contaminant accumulation rates (Nakata et al., 2002; Baptista et al., 2013).

The effect of sex on contaminant concentrations was evaluated in each species collected in SEY and revealed no significant difference, except for PFCAs and mirex, which showed higher concentrations in YFT males. Previous studies have reported differences in contaminant levels between female and male mature fish related to a contaminant loss following female spawning (Madenjian, 2011). The lack of gender influence on contaminant levels in tunas and swordfish in SEY is, however, expected owing to their reproductive strategy (i.e. multiple spawning, income breeders), which requires a high energy income from food intake and may, in turn, increase contaminant inputs (Grande et al., 2016; Zudaire et al., 2015; Poisson and Fauvel, 2009).

Most chlorinated contaminants were positively intercorrelated, with high correlation coefficients (0.56-0.91), whereas correlations between the various chlorinated and perfluorinated contaminants were weaker (0.24-0.46). These correlations indicate strong similarities in sources and bioaccumulation propensities of the various organochlorine families, in contrast to the perfluorinated ones. Indeed, the various studied organic contaminant families differ in terms of physico-chemical properties and behave differently in the environment and in fish.

PCBs and OCPs are lipophilic compounds, which bioaccumulate in the fat of living organisms, while PFASs have a stronger affinity with proteins, lipoproteins, and phospholipids (Dassuncao et al., 2019; Wen et al., 2019).

Out of all the studied species, SWO was the most-contaminated by chlorinated POPs, which is consistent with the higher trophic position and higher TLC of SWO. TLC showed high correlation with PCB and DDT concentrations (r =0.86 and 0.93, respectively) in SWO. Conversely, and for similar reasons (low trophic position and low TLC), SKJ exhibited the lowest chlorinated POP concentrations. When normalized to TLC, PCBs in SWO were similar to those in tuna species, while DDTs remained significantly higher. These results suggest that the differences in DDT concentrations across species were due to parameters other than TLC and that PCB and DDT concentrations displayed varying bioaccumulation propensities and affinities with TLCs. Among tunas, BET was the most-contaminated species (significantly higher HCB and PFASs than in YFT and SKJ) but displayed similar PFAS levels to SWO.

When all species were considered, $\delta^{15}N$ was significantly positively correlated with ∑ 18 PCB ($r = 0.24$, $p < 0.05$) and ∑ DDT ($r = 0.52$, $p < 0.05$) concentrations (in lw), but not with HCB or HCHs. However, the correlations were not significant when tunas alone were considered. Similarly, PFAS concentrations (ww) showed a significant correlation with $\delta^{15}N$ when all species were considered, and the correlation was significant in SKJ and YFT when tunas alone were considered.

3.4.2. PCB contamination levels and profiles

Among the 6 i-PCBs, congeners CB-180, CB-153 and CB-138 showed the highest detection frequencies (> 80%), with concentrations varying between 203.7-3117.3 (mean of 773.7) pg g^{-1} ww in SWO and 2.6-153.5 (mean of 26.4) pg g^{-1} ww in tropical tunas. Among dl-PCBs,

congeners CB-169, -167 and -189 were the most-frequently detected (> 80%). CB-28, CB-31, CB-81 and CB-114 were seldom detected (13%, 11%, 12% and 5% of samples, respectively). On average, i-PCB concentrations were 8 times higher than DL-PCB concentrations (i-PCB / dl-PCB ratio of 8.4 ± 5.9), ranging from 34.7 to 599.1 (mean of 154.6) pg g^{-1} ww in SWO and from 0.2 to 29.8 (mean of 4.5) pg g^{-1} ww in tunas. No significant differences in PCB concentrations were detected in tunas across the studied areas (with the exception of BET from MOZ: these smaller / younger individuals showed significantly lower contamination levels).

i-PCB profiles showed a predominance of hexa- and hepta-chlorinated congeners CB-153 > CB-138 > CB-180. These highly-bioaccumulable congeners are recalcitrant to degradation due to their chlorine atoms in positions 2,4,5 in one or both rings and often prevail in fish, including tunas (Gómara et al., 2005; Corsolini et al., 2007; Vizzini et al., 2010; Munschy et al., 2016). dl-PCB profiles showed predominant CB-118, followed by CB-105, CB-156 and CB-167. Similar profiles have already been reported in marine, lake and river fish from various regions around the globe (Ueno et al., 2005; Bhavsar et al., 2007; Munschy et al., 2016). Globally, all congeners showed significant correlations, suggesting similar sources and bioaccumulation in fish.

PCB profiles were examined in terms of number of chlorine atoms and structure activity groups (SAG I to V). SAG I, II and most SAG III congeners are non-metabolizable in fish, while SAG IV and V have been shown to metabolize (Boon et al., 1997). Although PCB biotransformation in fish is expected to be low in comparison to mammals, several studies have shown that fish were able to metabolize PCBs (Buckman et al., 2006 and references therein). This process may be enhanced in warmer climates such as those encountered in tropical areas (Buckman et al., 2007). Among the various studied species, the SWO PCB

profiles differed most, with significantly higher (17 ± 5%) and lower (29 ± 2%) contributions of 5 and 7 chlorine atom congeners respectively versus tunas (9 ± 8% and 39 ± 10%, respectively). SWO profiles were also characterized by lower SAG I and higher SAG IV contributions (although SAG IV was not significantly different in SWO and BET).

3.4.3. OCP concentrations and profiles

DDTs were the most-detected OCPs (91-100% depending on the isomer), together with mirex (detected in 100% of tuna samples). DDT concentrations ranged from 244.4 to 16836.1 pg g^{-1} ww (mean of 7269.1 pg g^{-1} ww) in SWO and 13.9 to 777 pg g^{-1} ww (mean of 112.0 pg g^{-1} ww) in tunas. Mirex was in the 0.4-34.1 pg g^{-1} ww range (mean of 3.7 pg g^{-1} ww) in tunas (not analysed in SWO). Among HCHs, ß-HCH was the most-prevalent isomer (84% of samples) and displayed the highest levels, at concentrations of 1.6-101.1 pg g^{-1} ww (mean of 58.0 pg g^{-1} ww) in SWO and 0.32-19.7 pg g^{-1} ww (mean of 2.5 pg g^{-1} ww) in tunas. α-HCH was quantified in 25% of samples. δ- and γ-HCH isomers were seldom detected in tunas (8% and 4% of samples, not analysed in SWO). HCB was detected in 65% of samples, at levels ranging from 37.3 to 533.2 pg g^{-1} ww in SWO (mean of 314.0 pg g^{-1} ww) and 4.1-37.2 pg g^{-1} ww in tunas (mean of 10.1 pg g^{-1} ww). Dieldrin (not analysed in SWO) was identified in 59% of tuna samples at concentrations ranging from 1.3 to 20.7 pg g^{-1} ww (4.3 pg g^{-1} ww mean value). Aldrin and endrin were detected in 30% and 16% of tuna samples, at concentrations of 0.2-3.3 pg g^{-1} ww (mean 0.9 pg g^{-1} ww) and 0.3-2.1 pg g^{-1} ww (mean 0.8 pg g^{-1} ww), respectively. Isodrin and endosulfans were below LOQs in all tuna samples.

Among DDT isomers, *p,p'*-DDE exhibited by far the highest contributions in all species (71 ± 9%), followed by *p,p'*-DDT (16 ± 7%), *p,p'*-DDD (7 ± 6%), *o,p'*-DDT (5 ± 3%) and *o,p'*-DDD (2 ± 2 %). In view of its longer half-life (7 years in fish, Binelli and Provini, 2003a) and higher

accumulation propensity, *p,p'*-DDE is generally identified as a predominant DDT isomer in fish, while *p,p'*-DDD ranks second (Covaci et al., 2006; Jürgens et al., 2015). In our samples, the high *p,p'*-DDT abundance (second-most abundant isomer) suggests potential inputs of technical DDT to this oceanic region from surrounding countries. Indeed, several publications have reported the continued use of DDT for various purposes in Southern Asia, Africa, India and China (Chakraborty et al., 2010; Cheng et al., 2011; Gioa et al., 2012; Bogdal et al., 2013; Ali et al., 2014; Bouwman et al., 2015). In particular, various nations in South-Eastern Africa were still listed as using DDT against malaria in 2012-2014 (van den Berg et al., 2017) and DDT was reported being sprayed in the Northeast part of South Africa (Bouwman et al., 2015). Similar contamination profiles as those found in our study have also been encountered in fish from coastal areas of Southern China and explained by DDT use in antifouling paints containing *p,p'*-DDT > *o,p'*-DDT > *p,p'*-DDE (Guo et al., 2008).

3.4.4. Organochlorine contaminant specific ratios

Various ratios using DDT isomers and PCB concentrations can be used to distinguish contamination sources in fish. The *o,p'*-DDT / *p,p'*-DDT concentration ratio, which is used to track the origin of DDT, was lower in SWO and BET (0.23 ± 0.16 and 0.27 ± 0.11, respectively) versus YFT and SKJ (0.47 ± 0.18 and 0.56 ± 0.31, respectively) (Fig. 4). This ratio is reported to be in the 0.2-0.3 range in technical DDT (Kalantzi et al., 2001), whereas a ratio above 0.34 is observed when DDT originates from dicofol acaricide impurities (Suarez et al., 2013), which is used mainly in Asia (Li et al., 2015). Our results suggest that SWO and BET sources of DDT contamination are both similar and closer to technical DDT than those of YFT and SKJ. In addition, SWO and BET exhibited significantly higher ∑ DDT / ∑ i-PCB concentration ratios (12.2 ± 3.3 and 9.1 ± 4.3, respectively) than SKJ and YFT (4.0 ± 2.1 and

4.6 ± 1.9, respectively) (Fig. 4), indicating SWO and BET exposure to different sources versus SKJ and YFT. This ratio is commonly used to distinguish agricultural (DDTs) from industrial and urban (PCBs) sources and indicates that tropical tunas and swordfish from the WIO were globally more exposed to DDTs over PCBs, reflecting the past or recent use of DDT by several countries in the Southern Hemisphere for vector-borne disease control purposes (Kalantzi et al., 2001; Chakraborty et al., 2010; Cheng et al., 2011; Bogdal et al., 2013; Ali et al., 2014; Bouwman et al., 2015). Hence, although no significant differences were found in $\delta^{13}C$ values across the three tuna species, both *o,p'*-DDT / *p,p'*-DDT and DDT / PCB concentration ratios suggested that BET was exposed to similar trophic sources as SWO. Indeed, BET and SWO both feed on deep water mesopelagic cephalopods and fish, while YFT feed on surface and subsurface (< 100 m) epipelagic species (Ménard et al., 2007; Potier et al., 2007; Olson et al., 2016). No statistically significant differences in *o,p'*-DDT / *p,p'*-DDT or ∑ DDT / ∑ i-PCB ratios were observed across tropical tuna sampling areas.

The (*p,p'*-DDE + *p,p'*-DDD) / ∑ DDTs ratio, which can be used to distinguish new and old DDT sources, was similar in all species (0.77 ± 0.08, on average), indicating that all fish were exposed to old DDT inputs (Suarez et al., 2013). This ratio was significantly lower in tunas from MOZ (0.70 ± 0.08) than in those from the other sampling areas (0.79 ± 0.07, on average) (Fig.4). These results suggest that tunas from MOZ have been exposed to different DDT sources versus other areas, coinciding with the reported use of DDT in various Southern African countries (Bouwman et al., 2015; van den Berg et al., 2017). They are also consistent with the recent work of Chassot et al. (2019), showing an extended residency time of tropical tunas in the northern Mozambique Channel, which would be reflected in their contamination profiles.

3.4.5. PFAS concentrations and profiles

Among the analysed PFASs, PFUnDA, PFTrDA, PFDoDA, PFOS, PFDA, PFTeDA and PFNA were detected in 100%, 93%, 90%, 88%, 85%, 74% and 54% of samples, respectively. Most of these PFASs were significantly inter-correlated, indicating similar sources and bioaccumulation (i.e. accumulation, elimination) across all species. PFNA showed the lowest correlations (no significant correlation with PFOS and PFDoDA and lower significant correlation coefficients with other PFASs), underlining a singular behaviour and/or source for this compound. In addition, PFOA and PFHxA were seldom detected (12% and 7% of samples, respectively) and PFHpA was below LOQ in all samples. Consequently, these 3 PFASs were not considered further in data evaluation. PFCA individual mean concentrations were between 12.4 ± 8.6 pg g^{-1} ww (PFTeDA) and 95.4 ± 61.9 pg g^{-1} ww (PFUnDA), while PFOS was in the 4.5-85.9 pg g^{-1} ww range (mean concentration of 37.9 ± 17.3 pg g^{-1} ww). ∑ PFCA concentrations were, on average, 6 times higher than PFOS concentrations in all samples. Interestingly, the ∑ PFCA / PFOS ratios were significantly higher in SEY and CHAG (6.9 ± 2.9 and 5.7 ± 1.3, respectively) than in MOZ and SOM (4.1 ± 1.2 and 4.5 ± 1.7, respectively) which are situated the closest to African continental coasts. Oceans are recognized as the main reservoirs of PFASs (Johansson et al., 2019) and PFCAs have been shown to accumulate in central ocean gyres including the Indian Ocean (Gonzalez-Gaya et al., 2014). In addition, the particular regional monsoon-driven circulation pattern occurring in the WIO (Olson et al., 2016) could have a major influence upon the spatial distribution of PFCAs, and, in turn, upon swordfish and tuna exposure in open waters. SWO and BET showed significantly higher PFOS and ∑ PFCA concentrations than SKJ and YFT, in line with their higher trophic position and with the higher biomagnification propensity of PFASs of higher carbon-chain length in trophic webs (Conder et al., 2008; Loi et al., 2011). Individually, higher

correlations were found for PFDA (r = 0.67) and PFUnDA (r = 0.58). The ∑ PFCA / PFOS concentration ratios showed significant differences across species (all sampling areas considered), with SKJ exhibiting significantly lower values (3.8 ± 1.4) than the 3 other species (6.4 ± 2.5).

The majority of PFASs identified in samples were long-chain PFCAs (C ≥ 8), consistent with the higher bioaccumulative abilities of these compounds versus their short-chain counterparts (Martin et al., 2003; Kelly et al., 2009; Pan et al., 2014). In marine biota, the atmospheric degradation of neutral precursors is the main source of PFCAs (Ellis et al., 2004; Li et al., 2019). PFUnDA and other long-chain perfluorocarboxylates could also originate from the degradation of 10:2 FTOH (Hart et al., 2008). More specifically, PFCA profiles showed a predominance of odd-chain length compounds (PFUnDA and PFTrDA) versus even-chain (PFDA, PFDoA and PFTeDA), which has been previously observed in fish and partially explained by the degradation of fluorotelomer alcohols (FTOHs) (Ellis et al., 2004; Martin et al., 2004). However, as FTOHs degrade in both odd and even numbered PFCAs, this alone does not explain the predominance of odd-numbered PFCAs. Interestingly, odd-numbered PFCAs, particularly PFUnDA, were also identified as the predominant components in human serum samples from various East Asian countries (Harada et al., 2011), thought to originate from the industrial production of fluoropolymers (Surflon S-111). Indeed, odd-chain PFCAs such as PFUnDA and PFTrDA have been reported as impurities in PFNA produced from the oxidation of fluorotelomer olefins (Prevedouros et al., 2006; Rotander et al., 2012). PFUnDA has been identified as the predominant PFCA in various fish species from East Asia (Hart et al., 2008; Loi et al., 2011). Neutral precursors such as 10:2 perfluorodecanol (FTOH) could also be at the origin of PFUnDA in marine biota, together with PFDA (Ellis et al., 2004; Hart et al., 2008). PFUnDA was found in higher concentrations than PFDA, due to its higher

propensity to bioaccumulate and biomagnify in food chains (Loi et al., 2011; Pan et al., 2018). One remarkable finding was the PFUnDA / PFDA ratio, which showed significantly lower values in SWO (3.2 ± 0.8) than in tunas (5.3 ± 1.1). This finding could be the result of complex interrelated effects between the biochemical composition of muscle in the various studied species and different PFCA binding affinities (Verreault et al., 2005).

3.4.6. PCB, DDT and PFAS levels in tropical tunas and swordfish versus other areas worldwide

A comparison of the levels of PCBs and DDTs (the two most-reported POP families in tunas and swordfish) determined in this study versus those reported in the literature in various top predator species collected from around the world is presented in Table 3. By far the most widely-studied species is BFT, with the majority of studies pertaining to the Mediterranean Sea. Variations in sampling times, size/age and sex of fish studied in the literature make comparisons difficult. In addition, some of the available data are up to 20 years old and does not therefore take into account the global decrease in environmental PCB and OCP levels brought about by effective regulations. However, the concentrations determined in our study are globally low in comparison to those reported in other oceans, in particular in the Northern Hemisphere, which is subject to higher inputs from industrialized and urban areas (Aguilar et al., 2002; Tanabe and Ramu, 2012). Indeed, distance from continental industrial and urban sources is the main factor influencing POP levels in oceans (Gioa et al., 2012). Similarly to us, Nicklish et al. (2017) reported lower POP levels in YFT collected from various locations in the Southern Hemisphere and North-West Pacific Ocean versus YFT from the Northern Hemisphere.

More specifically, in terms of the Indian Ocean, our results are comparable to those reported for i-PCBs and dl-PCBs in albacore tuna (*Thunnus alalunga*) collected in 2013 from Reunion island (10.2 ± 7.6 and 1.8 ± 1.3 ng g^{-1} lw, respectively, Munschy et al., 2016). Ueno et al. (2005) published dl-PCB concentrations of 4.2 ng g^{-1} lw in the muscle of SKJ collected off the Seychelles in 1997-2001, i.e. above those determined in our study (mean values of 0.8 ± 1.6 ng g^{-1} lw, Table 3). According to the authors, PCB, DDT, HCH and HCB concentrations in SKJ liver collected off the Seychelles were the lowest out of all the studied areas, including the North Pacific (mainly), Indian Ocean (off-Seychelles and the Bay of Bengal) and Atlantic Ocean (off-Brazil) (Ueno et al., 2003). ∑ 7 PCBs were in the 21.6-226.7 ng g^{-1} lw range in YFT collected in 2007 from Reunion island (Torres et al., 2009), i.e. higher than that determined in our samples (8.0 ± 8.3 ng g^{-1} lw). PCB levels determined in our study were comparable to those found in remote areas. For example, Lana et al. (2014) reported PCB concentrations of 11.1-63.1 ng g^{-1} lw in demersal Nototheniidae fish from the South Shetland Islands (Antarctica), i.e. close to the mean concentrations determined in our samples (5.3 ± 6.5 ng g^{-1} lw in BET to 9.3 ± 12.8 ng g^{-1} lw in SWO). Dl-PCB concentrations in top predators from WIO are also comparable to those reported in various freshwater fish from Africa (for a review, see Ssebugere et al., 2019).

With regards to DDTs, the levels (in lw) determined in this study are comparable to those reported in YFT from WIO (Torres et al., 2009) and in albacore tuna (*Thunnus alalunga*) from Reunion island and South East Atlantic Ocean (Munschy et al., 2016) and in a similar range in SWO to those reported in YFT from Brazil (South West Atlantic Ocean) (Pizzochero et al., 2011). Higher DDT levels were reported in YFT from Australia-New Zealand (South West Pacific Ocean) (Endo et al., 2016). DDT levels reported in various species from the Northern Hemisphere (bearing in mind that some data were collected 20 years ago) were generally

higher than those determined in our study, but the differences between hemispheres were lower than for PCBs.

Less data are available for PFASs and liver is the most widely-studied fish tissue. Hart et al. (2008) reported PFAS concentrations in the liver of SKJ collected from various locations in the Pacific and Indian Oceans. The lowest PFAS levels were determined in samples from open-ocean sites, with Mauritius in the Indian Ocean demonstrating the lowest concentrations (1.04 ng g^{-1} ww), while samples collected off Japan were 10 to 80 times higher. PFOS and PFUnDA were the most-detected compounds (100%) and exhibited the highest concentrations, while PFNA was the least-detected compound (57%), mostly below LOQs. PFOS, PFDA and PFNA below limits of detection were reported in the muscle of YFT from the Indian Ocean (Zabaleta et al., 2015) and in the muscle and liver of SWO from the Mediterranean Sea (Corsolini et al., 2008). PFOS ranged from 21 to 87 (mean of 47) ng g^{-1} ww in the liver of Atlantic bluefin tuna (*Thunnus thynnus*) from the Italian coast, while PFOA and PFHxS were < LOQs (Kannan et al., 2002). PFOS concentrations were between < LOQ and 13 ng g^{-1} ww in SWO livers from the same area. In Atlantic bluefin tuna and SWO consumed in France (no reported geographical origin), ∑ PFASs were in the 0.86-1.38 ng g^{-1} ww and 0.28-1.11 ng g^{-1} ww ranges, respectively (Yamada et al., 2014).

3.4.7. POP levels versus environmental and human health regulations

The ubiquity, persistence, accumulation in food chains and toxicity of POPs has raised concern as to their harmfulness to humans. Food and, in particular, seafood, is one of the major routes of human exposure to these pollutants (Törnkvist et al., 2011; Shin et al., 2015). Maximum residue levels in foodstuffs have therefore been established for selected POPs. The European Commission has introduced maximum levels for PCBs (dl-PCBs and i-PCBs) in

fishery products (EU, 2011). Other countries, such as the USA, Canada, Japan and Australia, have also defined maximum or guideline values. Environmental quality standards (EQSs) have been established within the EU Water Framework Directive (WFD) for certain priority substances, in the aim of protecting humans and wildlife. EQSs for HCB, PFOS and dl-PCBs are defined according to human health considerations (contaminated fish consumption). Globally, the levels of all studied POPs were far below admissible levels (several orders of magnitude) in the white muscle of the studied fish species; their consumption is therefore of no risk to humans. i-PCBs were far below (24 to 29000 times, mean of 5200 times) the maximum levels set by the European Commission for non dl-PCBs in foodstuffs (EU, 2011), i.e. 75 ng g^{-1} ww in muscle, hence below the maximum values set by the governments of Japan (500 ng g^{-1} ww), Australia (500 ng g^{-1} ww) and the US (2000 ng g^{-1} ww) for total PCBs (Vizzini et al., 2010). dl-PCB concentrations calculated in toxic equivalents (TEQs) using the toxic equivalent factors (TEFs) set by the World Health Organization in 2005 (Van den Berg et al., 2006) showed average levels of 0.016 ± 0.011 pg TEQ g^{-1} ww in tunas and 0.375 ± 0.314 pg TEQ g^{-1} ww in SWO, i.e. far below the maximum level of 3 pg TEQ g^{-1} ww set by the European Commission for dl-PCBs in foodstuffs (EU, 2011). They were also below the guideline value of 0.79 pg TEQ g^{-1} ww set by Canada to protect wildlife consumers of aquatic biota (CCME, 2001). Non-ortho PCBs and, in particular, CB-126 (78% of dl-PCB TEQs) contributed the most to TEQ values. Lindane and other HCH concentrations were below EU and China maximum limits. HCB concentrations were far below (20 to 7000 times) the environmental quality standards (EQS) of 10 µg kg^{-1} set to protect human health. Regarding DDTs, the maximum levels found in our study were below the value of 3000 ng g^{-1} ww and 1000 ng g^{-1} ww, respectively set by Japan and Australia for *p,p'*-DDE (Vizzini et al., 2010). DDT concentrations were below the maximum admissible concentrations set by the EU (50 ng g^{-1} ww, Binelli and

Provini, 2003b) for human consumption. Three SWO samples (2 females and one male at 15.6, 16.8 and 16.6 ng g^{-1} ww) exceeded the maximum levels of 14 ng g^{-1} ww set by Canada (CCME, 1999). PFOS concentrations were below the EQS of 9.1 ng g^{-1} ww in biota in all samples.

In addition, PCB levels were compared to OSPAR assessment criteria, i.e., EACs (Environmental Assessment Criterias) and BACs (Background Assessment Criterias) defined for congeners CB-28, -52, -101, -118, -138, -153 and -180. All PCB concentrations were below BACs in tunas, but exceeded BACs for CB-101, -118, -138, 153 and -180 in 44%, 22%, 55%, 72% and 50% of SWO samples. EACs were never exceeded.

4. Conclusions

The results of this study emphasize the widespread contamination of various top predator fish species from the WIO by legacy POPs and organic contaminants of emerging concern, although at low levels in comparison to oceans worldwide, representing no risk to human health in reference to current regulations. DDTs and PFASs were identified as the predominant families of studied organic contaminants in SWO and tunas, respectively.

Organic contaminant accumulation profiles were influenced by multiple factors (both biological and ecological) and were shown to be useful tracers for differentiating fish populations exploiting different trophic habitats when used in combination with trophic ecology parameters (C and N stable isotopes). SWO showed the highest organic contamination levels, in relation to their higher lipid content and trophic position, while SKJ was the least-contaminated species. SWO and BET exhibited the most organic contaminant profile similarities, due to their common feeding habitat in deeper layers versus SKJ and YFT. More differences in contaminant profiles were found across species than areas. However, tunas from the

Mozambique Channel showed unique DDT profiles, confirming that tunas tend to reside longer in this area and possibly reflecting the use of DDT by various SE African countries. The PFCA / PFOS concentration ratio also proved to be a useful chemical tracer of fish populations from more-oceanic regions. Our results show that organic contaminant signatures are discriminant in long-lived migratory species.

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Fig. 1. Map of tropical tuna and swordfish fishing positions in the western Indian Ocean in 2013-2014. Dot size is proportional to the number of individuals (for details, see Table 1). MOZ = Mozambique Channel, SOM = off the Somalian coast, SEY = Seychelles, CHAG = Chagos. YFT = yellowfin tuna *(Thunnus albacares)*, SKJ = skipjack tuna *(Katsuwonus pelamis)*, BET = bigeye tuna (*Thunnus obesus*), SWO = swordfish (*Xiphias gladius).*

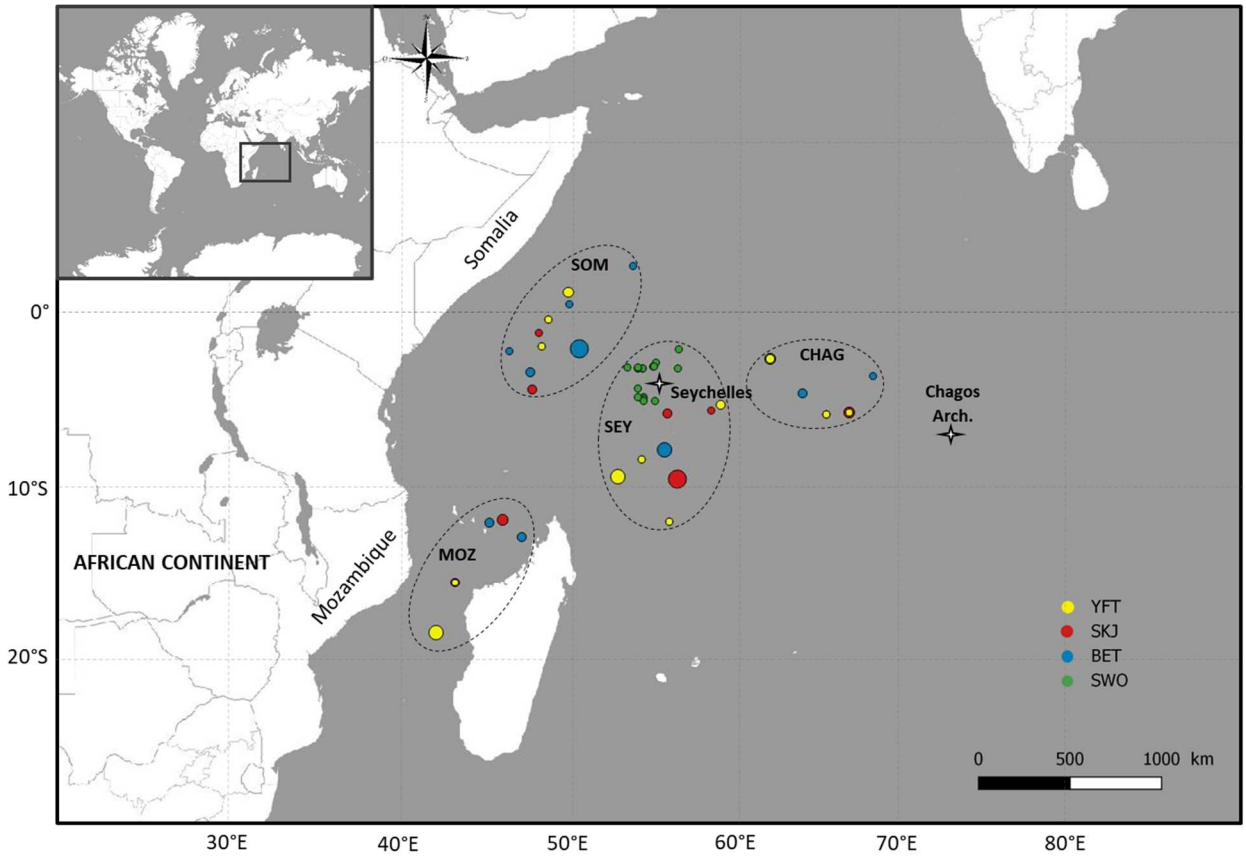

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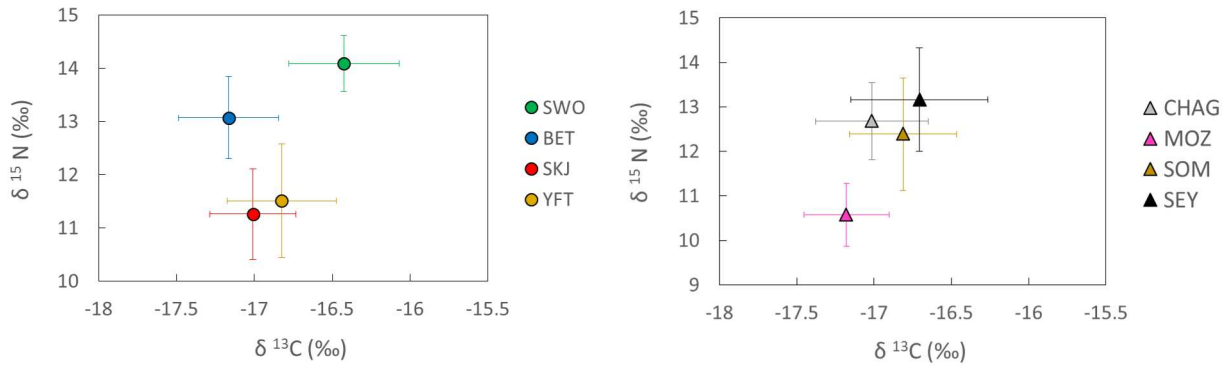
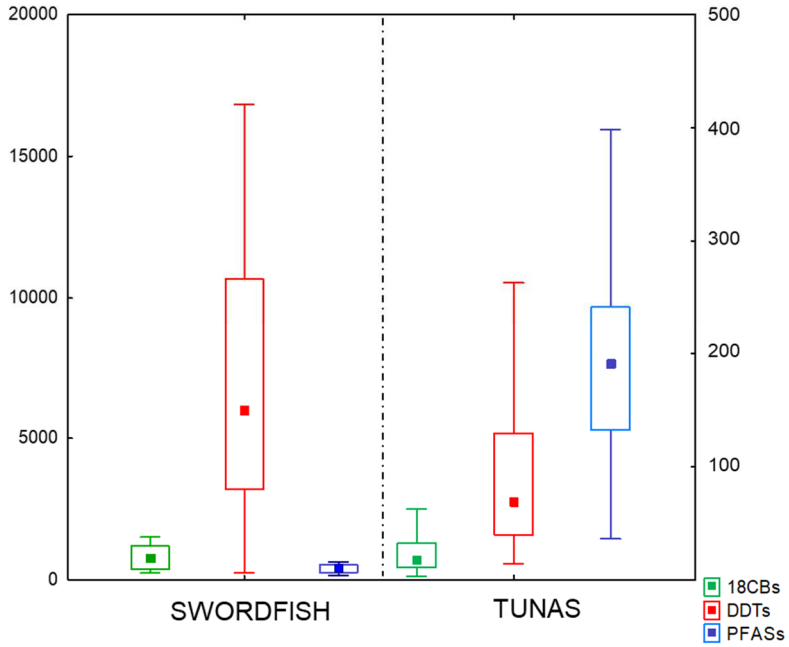
Fig. 2. Stable isotope signatures ($\delta^{13}C$ and $\delta^{15}N$ in ‰) in SWO (swordfish), BET (bigeye tuna), SKJ (skipjack tuna) and YFT (yellowfin tuna) collected in the western Indian Ocean in 2013-2014 and analysed for organic contaminants according to species and sampling areas. CHAG, MOZ, SOM and SEY are for Chagos, Mozambique Channel, off the Somalian coast and Seychelles, respectively.



2 columns

Fig. 3. PCB (∑ 18 PCBs), DDT (∑ 5 isomers) and PFAS (∑ PFSAs + PFCAs) concentrations (pg g^{-1} ww) in the white muscle of swordfish (left y-axis) and tropical tunas (right y-axis) collected in the western Indian Ocean in 2013-2014.



Single column

Fig. 4. Two dimensional plot of (*p,p'*-DDD + *p,p'*-DDE) / ∑ DDT and *o,p*-DDT / *p,p'*-DDT (a and c) and ∑ DDT / ∑ PCB and *o,p*-DDT / *p,p'*-DDT (b and d) concentration ratios in tropical tuna species and SWO collected in the western Indian Ocean 2013-2014. YFT = yellowfin tuna *(Thunnus albacares)*, SKJ = skipjack tuna *(Katsuwonus pelamis)*, BET = bigeye tuna (*Thunnus obesus*), SWO = swordfish (*Xiphias gladius)*. CHAG, MOZ, SOM and SEY are for Chagos, Mozambique Channel, off Somalian coast and Seychelles, respectively.

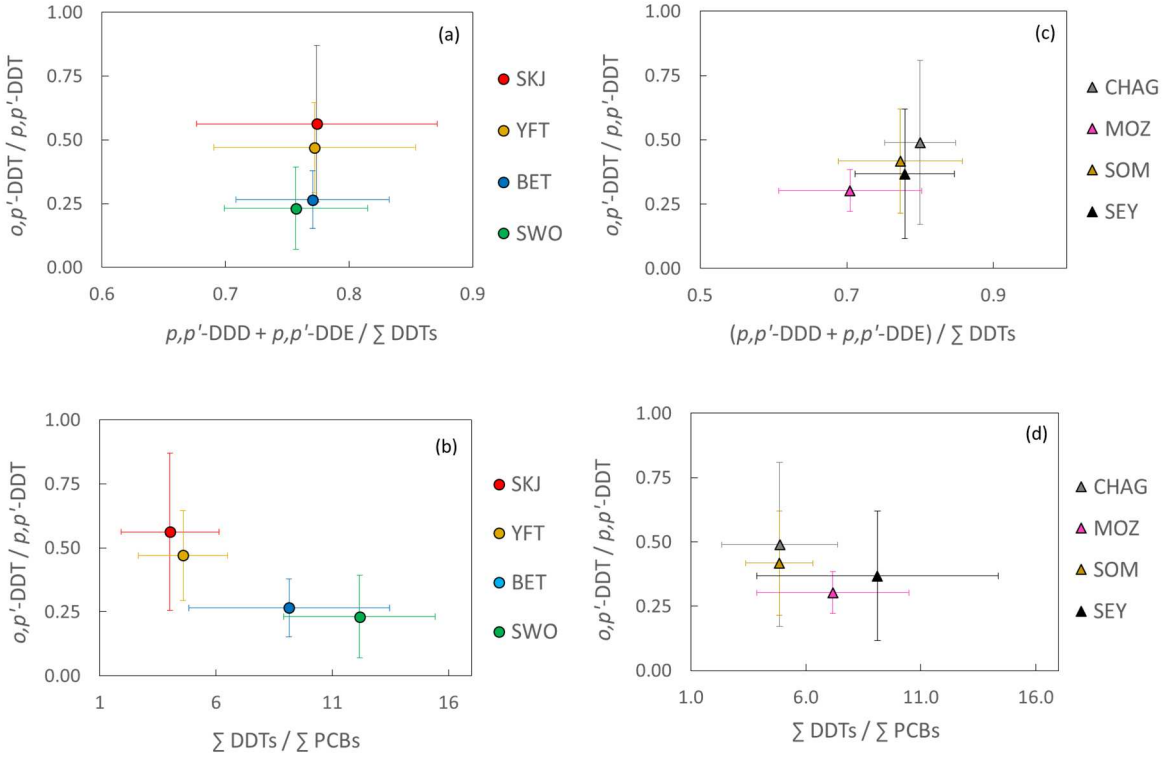


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Table 1

Biometric parameters (size in cm, mass in kg), condition index (K), TLC (% ww) and stable isotope signatures ($\delta^{13}C$ and $\delta^{15}N$ in ‰) of fish analysed for POPs. MOZ = Mozambique Channel, SOM = off the Somalian coast, SEY = Seychelles, CHAG = Chagos. YFT = yellowfin tuna *(Thunnus albacares)*, SKJ = skipjack tuna *(Katsuwonus pelamis)*, BET = bigeye tuna (*Thunnus obesus*), SWO = swordfish (*Xiphias gladius).* M = male, F = female, I = immature.

Species	Location	Sex	n	Size (cm)	Age (year)	Mass (kg)	K	TLC %	$\delta^{13}C$ ‰	$\delta^{15}N$ ‰
BET	All	All	27	116.5 ± 38.9 (47.1-166.5)	5.1 ± 3.2 (0.8-10.0)	48.5 ± 34.0 (2.6-108.7)	2.3 ± 0.2 (2.1-2.8)	0.45 ± 0.17 (0.14-0.88)	-17.2 ± 0.3 (-17.9 - -16.7)	13.1 ± 0.8 (11.3-14.0)
BET	CHAG	M	6	144.7 ± 15.6 (115.6-160.0)	7.8 ± 2.4 (3.7-10.0)	71.6 ± 20.5 35.5-94.7)	2.3 ± 0.1 (2.2-2.5)	0.37 ± 0.13 (0.25-0.56)	-17.2 ± 0.4 (-17.9 - -16.8)	13.5 ± 0.3 (13.1-13.9)
	MOZ	I	5	52.9 ± 3.5 (47.1-55.5)	1.2 ± 0.2 (0.8-1.3)	3.4 ± 0.6 (2.6-3.9)	2.3 ± 0.1 (2.1-2.5)	0.42 ± 0.11 (0.32-0.59)	-17.5 ± 0.1 (-17.5 - -17.4)	11.6 ± 0.3 (11.3-11.8)
	SEY	M	5	116.6 ± 34.3 (82.0-166.5)	4.7 ± 3.2 (2.2-10.0)	46.5 ± 39.4 (12.0-108.7)	2.4 ± 0.3 (2.2-2.8)	0.43 ± 0.16 (0.34-0.71)	-17.3 ± 0.3 (-17.6 - -17.0)	13.0 ± 0.6 (12.5-13.5)
	SEY	F	5	114.5 ± 25.8 (85.5-137.5)	4.1 ± 1.6 (2.4-5.7)	40.9 ± 24.1 (14.3-61.4)	2.4 ± 0.1 (2.3-2.6)	0.55 ± 0.22 (0.34-0.88)	-17.1 ± 0.1 (-17.2 - -17.0)	13.2 ± 0.3 (12.9-13.6)
	SOM	M	6	142.9 ± 17.5 (114.1-160.8)	7.1 ± 2.6 (3.6-10.0)	71.1 ± 22.7 (36.1-96.0)	2.4 ± 0.2 (2.2-2.7)	0.47 ± 0.23 (0.14-0.81)	-16.9 ± 0.2 (-17.1 - -16.7)	13.4 ± 0.6 (12.5-14.0)
SKJ	All	All	24	57.6 ± 7.6 (44.3-71.6)	3.2 ± 2.5 (0.7-10.0)	4.7 ± 2.0 (1.8-8.5)	2.3 ± 0.2 (1.9-2.7)	0.37 ± 0.19 (0.17-0.85)	-17.0 ± 0.3 (-17.5 - -16.4)	11.3 ± 0.8 (9.8-12.6)
SKJ	CHAG	M	4	64.6 ± 9.4 (51.0-71.6)	6.6 ± 4.2 (1.2-10.0)	6.7 ± 2.7 (2.7-8.5)	2.4 ± 0.3 (2.0-2.7)	0.37 ± 0.25 (0.20-0.73)	-17.0 ± 0.3 (-17.5 - -16.8)	11.9 ± 0.5 (11.4-12.6)
	MOZ	M	5	53.1 ± 6.5 (47.0-62.2)	1.8 ± 1.2 (0.8-3.7)	3.3 ± 1.5 (2.1-5.5)	2.1 ± 0.1 (1.9-2.3)	0.33 ± 0.09 (0.24-0.47)	-17.2 ± 0.1 (-17.4 - -17.1)	10.4 ± 0.6 (9.8-11.2)
	SEY	M	5	58.6 ± 8.1 (44.3-63.9)	3.1 ± 1.4 (0.7-4.3)	4.9 ± 1.8 (1.8-6.4)	2.3 ± 0.2 (2.1-2.5)	0.28 ± 0.10 (0.17-0.43)	-16.8 ± 0.2 (-17.0 - -17.0)	11.9 ± 0.6 (11.1-12.3)

	SEY	F	5	59.3 ± 3.3 (54.9-62.4)	2.9 ± 0.8 (1.9-3.6)	4.9 ± 0.9 (3.7-6.0)	2.3 ± 0.0 (2.3-2.3)	0.54 ± 0.23 (0.26-0.85)	-17.0 ± 0.02 (-17.0 - -17.0)	11.9 ± 0.00 (11.9-11.9)
	SOM	M	5	54.0 ± 7.3 (48.0-66.1)	2.1 ± 1.9 (0.9-5.5)	3.9 ± 2.1 (2.3-7.4)	2.3 ± 0.3 (2.0-2.7)	0.35 ± 0.18 (0.23-0.66)	-17.0 ± 0.02 (-17.0 - -17.0)	11.9 ± 0.00 (11.9-11.9)
YFT	All	All	24	121.5 ± 28.8 (66.4-161.5)	4.8 ± 3.1 (2.0-10.0)	41.6 ± 23.3 (5.5-80.3)	2.1 ± 0.1 (1.8-2.4)	0.52 ± 0.30 (0.13-1.11)	-16.8 ± 0.3 (-17.7 - -16.3)	11.5 ± 1.1 (9.7-13.2)
YFT	CHAG	M	4	125.3 ± 37.8 (72.2-155.5)	6.4 ± 4.2 (2.1-10.0)	50.6 ± 32.3 (9.1-80.3)	2.2 ± 0.1 (2.1-2.4)	0.56 ± 0.31 (0.34-0.99)	-16.8 ± 0.2 (-17.1 - -16.7)	12.2 ± 0.7 (11.6-13.2)
	MOZ	M	6	108.4 ± 8.7 (95.0-118.3)	2.8 ± 0.2 (2.4-3.0)	26.1 ± 6.1 (18.2-34.8)	2.0 ± 0.1 (1.9-2.2)	0.58 ± 0.32 (0.29-1.05)	-17.0 ± 0.3 (-17.5 - -16.8)	10.2 ± 0.4 (9.7-10.7)
	SEY	M	4	139.0 ± 17.0 (123.0-161.5)	6.7 ± 3.8 (3.3-10.0)	57.3 ± 18.2 (39.8-79.6)	2.1 ± 0.1 (1.9-2.2)	0.40 ± 0.07 (0.32-0.50)	-16.7 ± 0.2 (-16.8 - -16.6)	11.5 ± 0.1 (11.4-11.5)
	SEY	F	5	130.2 ± 4.8 (126.3-138.0)	3.9 ± 0.8 (3.4-5.4)	43.7 ± 3.5 (40.4-48.3)	2.0 ± 0.1 (1.9-2.1)	0.52 ± 0.36 (0.15-1.11)	-17.0 ± 0.6 (-17.7 - -16.6)	11.7 ± 0.6 (10.9-12.1)
	SOM	M	5	111.7 ± 40.6 (66.4-157.0)	5.4 ± 4.2 (2.0-10.0)	38.4 ± 36.4 (5.5-78.2)	2.0 ± 0.1 (2.0-2.2)	0.50 ± 0.40 (0.13-1.05)	-16.6 ± 0.3 (-17.1 - -16.3)	12.4 ± 0.8 (11.2-13.2)
SWO	All	All	18	166.8 ± 28.6 (110.4-204.0)	6.5 ± 4.6 (2.0-20.0)	nd	nd	9.4 ± 5.7 (0.63-18.8)	-16.4 ± 0.4 (-17.1 - -15.9)	14.1 ± 0.5 (13.1-14.9)
SWO	SEY	M	9	159.6 ± 31.6 (110.4-204.0)	7.3 ± 6.2 (2.0-20.0)	nd	nd	11.3 ± 5.5 (4.8-18.8)	-16.3 ± 0.4 (-17.0 - -15.9)	14.2 ± 0.6 (13.1-14.9)
		F	9	174.0 ± 25.0 (124.2-203.3)	5.6 ± 2.1 (2.3-8.6)	nd	nd	7.4± 5.4 (0.63-16.2)	-16.5 ± 0.3 (-17.1 - -15.9)	14.0 ± 0.4 (13.4-14.7)

nd: not determined

Table 2

Major POP concentrations (pg g^{-1} ww) in the white muscle of tropical tunas and swordfish collected in the western Indian Ocean in 2013-2014. MOZ = Mozambique Channel, SOM = off the Somalian coast, SEY = Seychelles, CHAG = Chagos. YFT = yellowfin tuna *(Thunnus albacares)*, SKJ = skipjack tuna *(Katsuwonus pelamis)*, BET = bigeye tuna (*Thunnus obesus*), SWO = swordfish (*Xiphias gladius).* M = male, F = female, I = immature.

Species	Location	Sex	n	i-PCBs	DDTs	PFOS	PFCAs
BET	All	All	27	26.7 ± 34.4 (3.6-153.5)	170.8 ± 167.8 (50.2-777.2)	44.0 ± 17.1 (13.6-79.9)	262.8 ± 123.4 (147.7-663.8)
	CHAG	M	6	30.7 ± 23.6 (9.1-76.0)	185.5 ± 102.3 (79.3-334.0)	44.5 ± 21.3 (21.6-79.9)	290.1 ± 189.7 (159.3-663.8)
	MOZ	I	5	7.9 ± 3.2 (5.3-12.7)	76.0 ± 25.8 (50.2-106.5)	34.2 ± 9.0 (25.1-44.6)	187.4 ± 25.4 (147.7-206.8)
	SEY	M	5	11.5 ± 8.5 (4.2-25.5)	113.2 ± 74.7 (53.5-229.4)	46.8 ± 13.9 (35.4-63.3)	303.7 ± 77.5 (193.0-402.4)
	SEY	F	5	41.7 ± 63.3 (3.6-153.5)	249.2 ± 299.6 (68.4-777.2)	40.5 ± 26.4 (13.6-72.0)	295.6 ± 174.9 (169.4-594.0)
	SOM	M	6	36.9 ± 35.4 (10.7-96.8)	227.3 ± 198.9 (70.1-544.7)	50.1 ± 13.9 (24.0-61.4)	236.6 ± 56.5 (168.4-314.7)
SKJ	All	All	24	17.4 ± 24.9 (2.6-120.0)	40.6 ± 24.5 (13.9-129.2)	27.0 ± 10.5 (9.0-49.3)	103.1 ± 47.6 (28.5-213.5)
	CHAG	M	4	12.2 ± 3.8 (6.4-14.4)	33.6 ± 7.4 (25.5-43.1)	31.2 ± 5.4 (27.6-37.4)	169.5 ± 30.1 (147.8-213.5)
	MOZ	M	5	11.7 ± 4.6 (5.3-16.9)	47.6 ± 6.2 (40.8-56.5)	25.5 ± 4.5 (20.3-30.8)	69.5 ± 12.3 (53.2-87.1)
	SEY	M	5	12.3 ± 9.9 (5.9-29.7)	30.8 ± 9.1 (20.9-39.7)	25.2 ± 4.4 (21.5-30.9)	106.3 ± 29.4 (57.4-129.7)

	SEY	F	5	(2.6-120.0)*	28.5 ± 20.5 (13.9-64.6)	11.4 ± 3.6 (9.0-15.5)	66.5 ± 43.1 (28.5-121.6)
	SOM	M	5	14.1 ± 4.1 (8.1-17.4)	61.1 ± 44.1 (16.6-129.2)	36.4 ± 11.6 (26.2-49.3)	109.6 ± 46.4 (65.2-173.0)
YFT	All	All	24	35.0 ± 29.4 (5.8-119.2)	120.0 ± 82.1 (19.6-444.8)	28.8 ± 11.7 (4.5-46.1)	152.9 ± 60.3 (31.3-276.8)
	CHAG	M	4	44.5 ± 20.2 (22.1-69.5)	151.4 ± 56.6 (103.7-224.7)	42.5 ± 2.8 (40.2-46.1)	215.1 ± 52.0 (160.9-276.8)
	MOZ	M	6	14.6 ± 6.1 (5.8-21.8)	89.9 ± 27.8 (53.1-129.3)	29.5 ± 5.3 (21.1-36.0)	120.1 ± 21.6 (85.3-145.3)
	SEY	M	4	36.4 ± 28.5 (12.1-69.7)	120.0 ± 109.6 (32.3-272.5)	34.2 ± 3.6 (30.2-36.9)	204.2 ± 40.0 (160.1-256.1)
	SEY	F	5	(8.3-13.7)*	37.7 ± 26.9 (19.6-85.2)	9.2 ± 7.9 (4.5-18.4)	97.9 ± 46.2 (31.3-142.1)
	SOM	M	5	56.4 ± 41.0 (12.1-119.2)	207.4 ± 142.8 (56.9-444.8)	23.4 ± 5.1 (17.4-26.4)	156.2 ± 55.0 (68.6-215.5)
SWO	SEY	All	18	773.7 ± 713.4 (203.7-3117.3)	7269.1 ± 5220.6 (244.4-16836.1)	51.4 ± 16.5 (27.9-85.9)	372.7 ± 202.7 (120.5-952.9)
	SEY	M	9	664.0 ± 369.5 (299.7-1255.9)	7256.4 ± 4733.0 (244.4-16633.6)	53.7 ± 16.8 (36.9-85.9)	415.9 ± 251.0 (120.5-952.9)
		F	9	883.4 ± 962.6 (203.7-3117.3)	7281.8 ± 5959.4 (353.4-16836.1)	48.8 ± 17.0 (27.9-81.6)	324.0 ± 129.8 (124.6-538.7)

*: as only two females had CB-138 and/or CB-153 and/or CB-180 above LOQs, ∑ i-PCBs was considered only for those females.

Table 3

Concentrations of PCBs and DDTs in the muscle of tropical tunas, swordfish and other top predator fish species worldwide. Albacore tuna *Thunnus alalunga* = ALB; Bluefin tuna *Thunnus thynnus* = BFT; Yellowfin tuna *Thunnus albacares* = YFT; Skipjack tuna *Katsuwonus pelamis* = SKJ; Southern bluefin tuna *Thunnus maccoyii* = SBT; Swordfish *Xiphias gladius* = SWO). ATL = Atlantic Ocean, MED = Mediterranean Sea, IO = Indian Ocean, WIO = western Indian Ocean, PAC = Pacific Ocean. Data are presented as minimum-maximum (mean ± sd) when available. Data are sorted by ocean, species and sampling date.

Species	Sampling date	Ocean / Location	PCB Congeners	PCBs ng/g ww	PCBs ng/g lw	Number of DDT isomers	DDTs ng/g ww	DDTs ng/g lw	Reference
SKJ	2013-2014	WIO	i-PCBs dl-PCBs	0.0003-0.120 (0.017 ± 0.025) 0.0002-0.021 (0.002 ± 0.004)	0.123-46.086 (5.432 ± 9.366) 0.007-8.00 (0.752 ± 1.631)	5	0.014-0.129 (0.040 ± 0.025)	2.013-55.629 (13.206 ± 10.869)	This study
YFT	2013-2014	WIO	i-PCBs dl-PCBs	0.0058-0.119 (0.035 ± 0.029) 0.0006-0.026 (0.007 ± 0.006)	0.300-27.984 (7.968 ± 8.304) 0.0437-7.796 (1.572 ± 1.902)	5	0.020-0.445 0.120 ± 0.099)	5.161-133.899 (30.873 ± 31.385)	This study
BET	2013-2014	WIO	i-PCBs dl-PCBs	0.0036-0.153 (0.027 ± 0.034) 0.0002-0.030 (0.004 ± 0.007)	0.571-27.134 (5.300 ± 6.502) 0.0708-4.697 (0.856 ± 1.251)	5	0.050-0.777 (0.171 ± 0.168)	12.866-119.283 (34.824 ± 28.492)	This study
SWO	2013-2014	WIO / Seychelles	i-PCBs dl-PCBs	0.204-3.117 (0.774 ± 0.713) 0.035-0.599 (0.155 ± 0.092)	3.017-53.776 (9.274 ± 12.797) 0.478-10.336 (1.832 ± 2.469)	5	0.244-16.84 (7.269 ± 5.221)	36.056-290.439 (81.355 ± 61.305)	This study
NORTHERN HEMISPHERE									

BFT[a]	--	MED	ndl	3.4-219.3 (84.2 ± 64.3)	209.8-6042.2 (982.1 ± 1041.0)				Barone et al., 2018
			dl	0.9-46.6 (17.4 ± 12.5)	22.7-631.3 (149.9 ± 145.3)				
Tunas	2012	MED / Spain (markets)	18	10.54					Perello et al., 2015
BFT	2005	MED / Sicily	43 PCBs		(2751.7 ± 1276.8)	1[b]		(2923.0 ± 785.9)	Vizzini et al., 2010
BFT	2004	MED/ Sicily	3 PCBs	nd-188.5	nd-2324	1[b]	3.5-515.0	nd-3480	Di Bella et al., 2006
BFT[a]	2003	MED / Italy	43 PCBs	5-1327	17-16839	1[b]	0.8-112	2.5-551	Corsolini et al., 2007
BFT	2001-2003	Spain (markets)	23 PCBs	10-176 (100.4)					Gomara et al., 2005
BFT	2001-2010	Italy (food shops)	Total PCBs	666 ± 475					Focardi, 2012
BFT	1999	MED / Sicily	35	21.39-324.54 (74.91)		6	0.95-299.58 (52.04)		Stefanelli et al., 2002
BFT	1999	MED / Italy	12	197-363		1[b]	30-67 (49)		Kannan et al., 2002
BFT	1999	MED/Sicily	57	12-229 (80 ± 86)		1[b]	5-97 (31 ± 38)		Corsolini et al., 2005
SWO	2012	MED / Spain	18	36.56					Perello et al., 2015
SWO	2005	MED / Spain (markets)	11	13.27					Bocio et al., 2007
SWO	1999	MED / Sicily	57	11-267 (89 ± 82)		1[b]	4.7-93 (38 ± 29)		Corsolini et al., 2005
SWO	1999	MED / Sicily	34	7.5-333.2 (80.4)		6	2.61-161.37 (52.11)		Stefanelli et al., 2004
SWO	1999	MED / Italy	12	258-399		1[b]	45-69 (57)		Kannan et al., 2002
BFT	2006	ATL / Spain	dl-PCBs	5.16 ± 0.88					Sprague et al., 2012
BFT	2008	ATL (north)	dl-PCBs	17.97					Mezzetta et al., 2011
SWO		ATL (north)	dl-PCBs	0.28					Mezzetta et al., 2011
YFT	2005-2007	PAC / Japan, Hawai, California	13		80 ± 70 550 ± 260	1[b]		30 ± 70 360 ± 190	Endo et al., 2016
		IO / Maldives, India	13		330 ± 420	1[b]		60 ± 100	

BFT	2003-2007	PAC / Japan (shops)	13 PCBs	0.5-46.1 (13.6 ± 17.2)	70-4370 (930 ± 600)	1[(b)]	1.0-104 (14.6 ± 17.2)	140-2860 (1040 ± 570)	Hisamichi et al., 2010
YFT			13 PCBs	0.03-7.29 (0.31 ± 0.43)	60-660 (230 ± 120)	1[(b)]	0.03-1.59 (0.24 ± 0.26)	50-480 (200 ± 100)	
ALB			13 PCBs	0.02-11.61 (1.32 ± 2.10)	30-1300 (400 ± 280)	1[(b)]	0.08-6.52 (0.97 ± 1.53)	20-1390 (290 ± 270)	
BFT	2005-2007	PAC / Korea (markets)	22 PCBs	(14.0 ± 14.3)		4	(12.5 ± 10.1)		Moon et al., 2009
SKJ	1997-2001	PAC / Various locations	dl-PCBs		6.3-62.0				Ueno et al., 2005
SKJ	1997-2001	IO / India	dl-PCBs		8.1				Ueno et al., 2005
SOUTHERN HEMISPHERE									
ALB	2013	IO / Réunion island	i-PCBs	0.02-0.52 (0.20 ± 0.14)	2.2-32.7 (10.2 ± 7.6)	5	0.08-2.18 (0.59 ± 0.48)	8.9-83.8 (28.4 ±16.1)	Munschy et al., 2016
			dl-PCBs	0.01-0.09 (0.03 ± 0.02)	0.52-5.84 (1.8 ± 1.3)				
YFT	2007	IO / Western	28 PCBs		43.8-381.7	4		4.4-43.2	Torres et al., 2009
			7 PCBs		21.6-226.7				
SBT (wild)	2004	IO / Australia	45 PCBs	0.4-0.55 (0.47)	24-200 (100)				Padula et al., 2008
SBT (farmed)	2005	IO / Australia	12 PCBs	0.08-1.58					Phua et al., 2008
SKJ	1997-2001	IO / Indonesia, Seychelles,	dl-PCBs		4.2-6.5				Ueno et al., 2005
ALB	2013	ATL / South Africa	i-PCBs	0.21-1.27 (0.58 ± 0.22)	2.7-21.8 (8.6 ± 4.7)	5	0.71-4.10 (2.09 ± 0.70)	8.9-62.8 (29.9 ±14.4)	Munschy et al., 2016
			dl-PCBs	0.05-0.33 (0.14 ± 0.06)	0.7-5.5 (2.1 ± 1.2)				
YFT	2009	ATL / Brazil	27		116.2-2556 (518.2)	6		15.8-178.7 (77.5)	Pizzochero et al., 2011
SWO	2001	ATL / Brazil	25	1.30-24.8 (6.5)	17.67-319.17 (127.2)	4	0.15-10.53 (2.47)		de Azevedo e Silva et al., 2007
SKJ	1997-2001	ATL / Brazil	dl-PCBs		14.0				Ueno et al., 2005
YFT	2005-2007	PAC / Australia, New Zealand	13		2890 ±3620 5250 ± 3380	1[(b)]		180 ± 210 540 ± 530	Endo et al ;, 2016

[(a)] Includes juveniles and adults

[(b)] *p,p'*-DDE