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Species-specific bioaccumulation of persistent organohalogen contaminants in a tropical marine ecosystem (Seychelles, western Indian Ocean)

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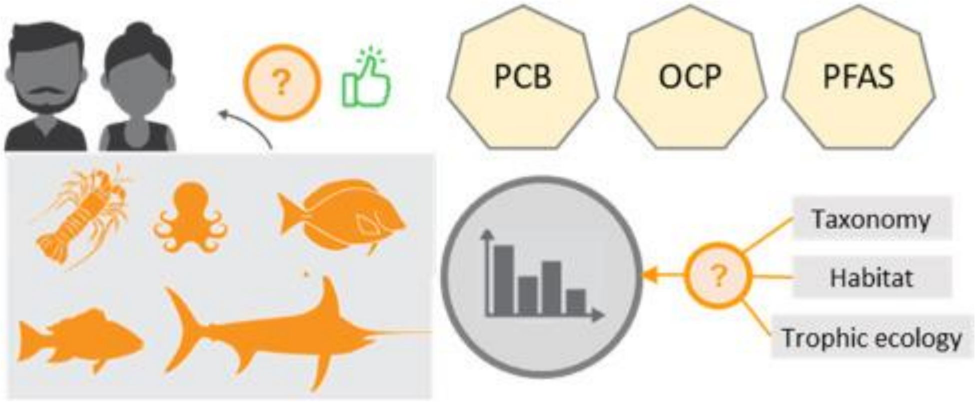
Graphical abstract



Highlights

► Numerous OHCs detected at low levels but moderate to high detection frequencies. ► Large inter-species differences in relative contributions of PCBs, OCPs and PFAS. ► Highest levels of DDTs determined in the oceanic top predator species. ► Herbivorous species characterized by a higher contribution of 3–4 Cl PCBs. ► Long-chain PFCAs (≥ C_8) by far predominant in comparison to PFOS.

1. Introduction

The Seychelles archipelago is one of the small island developing countries (SIDS) of the western Indian Ocean (WIO) characterized by a biodiverse marine ecosystem that provides high-quality seafood for the local population and sustains their local economy (Sabino et al., 2022; Jensen et al., 2023). Having a low population and no heavy industries or agricultural activities, the Seychelles waters are remote from major direct anthropogenic sources of contamination. However, coastal communities in the SIDS are recognized as particularly vulnerable to marine resource degradation in response to pollution and climate change (Landrigan et al., 2020). Global population growth and the associated development of human activities and coastal infrastructures have led to an increase in chemical pollution (Miraji et al., 2021). Besides, oceans, including those far from the sources of pollution, are final sinks for persistent substances such as organohalogen contaminants (OHCs) including the persistent organic pollutants (POPs), which are bioaccumulative, toxic and travel far from their emission sources, hence their global distribution (Dachs et al., 2002; Jones, 2021). Despite their ecological, social and economic value, extremely limited knowledge exists on the contamination of marine tropical ecosystems by POPs, especially in coastal reef species around the Seychelles. Data on the contamination of various large pelagic top predator fish, such as swordfish, tunas and sharks, in the western Indian Ocean have been published previously (Munschy et al., 2016; Munschy et al., 2020 and references therein; Chynel et al., 2021), but no data exist on the contamination of lower trophic level species such as those studied here (i.e. mollusks, crustaceans, herbivorous and planktivorous fish) by organic contaminants in the vicinity of the Seychelles archipelago. The target contaminants included the major families of POPs, namely polychlorinated biphenyls (PCBs), organochlorinated pesticides (OCPs) and perfluoroalkyl substances (PFAS), including the long-chain perfluorocarboxylic acids (PFCAs). PCBs and OCPs have been studied for decades in the marine environment; their persistence, bioaccumulation and propensity for biomagnification, and their toxicity to wildlife and humans, have been extensively proven (Jones, 2021). Despite the fact that international efforts have been made for decades to eliminate them by banning their use in most countries worldwide (Kalantzi et al., 2001; Breivik et al., 2007; Vijgen et al., 2011; Land et al., 2018), secondary emissions, contemporary uses and global distribution demonstrate the lack of success in their global management (Breivik et al., 2011; Ali et al., 2014; Bouwman et al., 2015; Jones, 2021; Melymuk et al., 2022). Given the above, POPs are still a cause for concern regarding their occurrence and impact in marine ecosystems and, in the context of increasing human activities and climate change, are likely to remain so in the coming decades. PFAS are another family of contaminants of increasing concern, in particular in oceans, which are their largest reservoir (Prevedouros et al., 2006; Johansson et al., 2019; Savvidou et al., 2023). Although PFAS present a higher diversity in their structure and have been less studied to date than the more classic POPs, they share similar properties (i.e., persistence, distribution in our global environment and especially in oceans), which has led to a growing interest from environmental scientists in recent years (Muir and Miaz, 2021; Wu et al., 2022).

The main objectives of this study were i) to gain knowledge on the contamination levels and profiles of the selected contaminants in a diverse range of tropical reef species, ii) to assess how the main biological and trophic factors (lipid content, trophic parameters, habitat) explain the observed differences in contamination, and iii) to compare the contaminant concentrations with current human consumption regulations. In the present study, a diverse range of coastal species characteristic of the Seychelles reef ecosystem were studied for their contamination in organohalogen hydrophobic compounds, in addition to various species collected offshore. To the best of our knowledge, the data presented here are among the first obtained in such a diverse range of tropical species representing various trophic levels and habitats from this part of the world. In the context of the increasing development of human activities and of climate change, the contamination status of these coastal ecosystems needs to be better assessed in order to increase knowledge on existing pressures in view of future conservation and sustainable development (Lu et al., 2018; Miraji et al., 2021). In this context, this study contributes towards achieving the Sustainable Development

Goals (SDGs) of the United Nations, in particular SDG 14 (life below water) and SDG 3 (good health and well-being) (https://sdgs.un.org/goals).

2. Material and Methods

2.1. Sampling

Ethical approval was not required for this study, as all studied organisms were bought directly from professional fishers during landing at Victoria fishing port, Seychelles. The samples were collected nearshore and offshore of the Mahé plateau in the Seychelles EEZ during 2013–2018 (Sabino et al., 2022). The majority of the reef species were collected between 2015 and 2018, while a limited number of species were collected in 2013-2014 (a total of 23 individuals out of the 115). With the very few exceptions of NXM and YFT, the replicate individuals of a given species were collected at the exact same time.

A total of 37 species (115 individuals) were selected for OHC analysis, including one species of Octopodidae, one species of Palinuridae and 35 species of fish belonging to various families (Table S1). Three individuals of each species were selected for OHC analyses, except in the case of the brown-marbled and the longspine grouper (n=2 for each) and the swordfish, bigeye tuna and yellowfin tuna (n=5 for each). The length (total length for octopus, cephalothorax length for crustaceans, lower jaw-fork length for swordfish, and fork length or total length for other fish species) and, whenever possible, the sex, were recorded for each individual. The edible part (white muscle) of each individual was sub-sampled and stored in amber glassware at -20°C prior to freeze-drying at the Seychelles Fishing Authority (SFA) laboratory. The freeze-dried samples were sent to Ifremer, Nantes, France, for organic contaminant and total lipid content analysis, and to the LIENSs laboratory, France, for stable isotope analysis.

2.2. Stable isotope analysis

Stable isotopes of carbon ($\delta^{13}C$) and nitrogen ($\delta^{15}N$) were analysed in each dry samples after removal of lipids (Sabino et al., 2022). Analyses were performed on a Thermo Scientific Flash 2000 elemental analyser coupled to a Delta V Plus interface mass spectrometer, using international isotopic standards of known $\delta^{13}C$ and $\delta^{15}N$ (USGS-61 and USGS-62). Measurement errors (SD) were <0.10 ‰ for both the nitrogen and carbon isotope measurements. For each sample, the C:N ratio was calculated, and never exceeded 3.5, proving that reserve lipids were adequately removed (Sabino et al., 2022).

2.3. Total lipid content (TLC) analysis

An aliquot of each freeze-dried sample (0.5 g dw) used for OHC analysis was extracted with a mixture of hexane/acetone (80:20) at 100 °C under 100 bars using pressurised liquid extraction (PLE) with a 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) expressed in % of wet weight (ww).

2.3. Contaminant analysis

Detailed analytical procedures for PCBs, OCPs and PFAS can be found in Munschy *et al.* (2020). Details on the chemicals and reagents (including solvents, analytical standards, cleanup materials) are provided in the Supplementary Material. Sample preparation and instrumental analysis are briefly described below and in Table S2.

Organochlorinated contaminants were analysed from 3–9 g of samples extracted by PLE with dichloromethane, followed by gel permeation chromatography, a silica and alumina adsorption chromatography column and a two-dimensional HPLC system with two columns coupled in series. Analyses were performed by gas chromatography (Agilent 6890, Palo Alto, CA, USA) coupled to a high-resolution mass spectrometer (AutoSpec Ultima, Waters Corp.). The samples were analysed for 35 PCBs ranging from tri- 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, hexachlorocyclohexanes - HCHs and hexachlorobenzene - HCB, referred to as ∑ OCPs later in the text).

PFAS analyses were conducted on 1 g of freeze-dried sample extracted using liquid-solid extraction (LSE) with a mix of MeOH/KOH and purified onto two consecutive SPE cartridges (Oasis WAX weak anion exchange stationary phase and Envicarb charcoal stationary phase). Analyses were 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 an electrospray ionization source Z-spray™ (Waters Corp.). The mass spectrometer was operated in negative ionization mode using multiple reaction monitoring (MRM) with argon as the collision gas. PFAS were analysed for five C_4- to C_{10}-perfluoroalkyl sulfonates (PFSAs) and nine C_6- to C_{14}- perfluorocarboxylic acids (PFCAs), namely: 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).

Concentrations were expressed on a wet weight (ww) basis using the moisture percentage determined after freeze-drying in each sample (mean of 76 ± 2%). PCB and OCP concentrations were also normalized to the TLCs and expressed on a lipid weight (lw) basis.

2.4. Quality assurance/quality control (QA/QC)

QA/QC procedures were followed during each sequence analysis. In order to minimize external and cross-contamination, all samples were processed in a clean laboratory (low dust and positive pressure) under a 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. Detailed analytical procedures can be found in Munschy et al. (2020) and QA/QC parameters are given in the supporting information.

2.5. Statistical analyses

All statistical analyses were performed using StatSoft Statistica software v 13.3 with a significance level (α) of < 0.05. Concentrations below the limits of quantification (LOQs) were assigned as missing values (i.e. counted as zero in the calculation of sums), and compounds quantified in less than 40% of the samples were not considered for statistical analyses. Data were tested for normality using the Shapiro-Wilk's test and parametric or non-parametric tests were performed depending on a normal distribution. Correlations (e.g. between total lipid content or $\delta^{15}N$ values and POP concentrations) were tested using simple linear regression coefficients, and Spearman's rank correlation test was used to evaluate the strength and direction of relationships. Data comparisons (biological parameters, POP concentrations and ratios) across groups were performed using non-parametric tests (Mann–Whitney and one-way ANOVA Kruskal-Wallis tests to compare independent groups) for non-normally distributed data. Results were considered significant only when both tests gave significant results.

3. Results and Discussion

3.1. Stable isotopes

The distribution of $\delta^{13}C$ and $\delta^{15}N$ values (Fig. 1) showed wide variation between species. $\delta^{13}C$ values ranged from -20.16‰ in the streamlined spinefoot (IGA) to -12.97‰ in the pronghorn spiny lobster (NUP). This wide range of values (~7‰ difference) indicates that there are different carbon sources sustaining the studied ecosystem. $\delta^{15}N$ values also showed variability, ranging from 9.90‰ in the blue-barred parrotfish (USY) and 9.94‰ in the streamlined spinefoot (IGA), respectively, to 14.75‰ in the pickhandle barracuda (BAC), i.e. a difference of more than one trophic level (following the expected mean difference of 3.4‰ per trophic level, Post, 2002). However, most studied species were in the 12.90-14.75‰ range. In addition to the herbivorous blue-barred parrotfish (USY) and streamlined spinefoot (IGA), the rough triggerfish (CNT), the pronghorn spiny lobster (NUP), the big blue octopus (OQC) and the Indian mackerel (RAG) were at the lower end of the $\delta^{15}N$ values.

3.2. Biometric parameters and total lipid content

Individuals to be analysed for OHCs were selected based on their lengths in order to minimize size variations within each species, resulting in fairly homogeneous lengths within species (most rsd < 10%); the highest length variations (12–30%) were observed in the large pelagic fish, namely swordfish (SWO), yellowfin tuna (YFT) and bigeye tuna (BET) (Table S1). Most species' biometric characteristics were within the common lengths reported in SeaLifeBase (Palomares and Pauly, 2020) and FishBase (Froese and Pauly, 2020). We can thus reasonably assume that the majority of specimens were adults.

TLC values showed wide variability between species, ranging from 0.22 ± 0.03 % ww in dogtooth tuna (DOT) to 9.4 ± 3.7% ww in swordfish (SWO) (Table S6). The inter-individual variability of TLC values within a species was also high in some cases: the streamlined spinefoot (IGA), the bigeye trevally (CXS), the bludger (NGY), the humpback red snapper (LJG), the two-spot red snapper (LJB), the tomato hind (EFT), the pickhandle barracuda (BAC), the common dolphinfish (DOL) and the striped marlin (MLS) showed relative standard deviations ≥ 50%. The highest TLC values within each fish family were determined in the bigeye trevally (CXS, Carangidae) (2.1 ± 1.7% ww), in the humpback red snapper (LJG, Lutjanidae) (4.5 ± 2.4% ww), and in the eightbar grouper (EWO, Serranidae) (1.8 ± 0.9% ww) and tomato hind (EFT, Serranidae) (1.4 ± 1.3% ww), while Lethrinidae showed less variable TLC values (0.4 ± 0.1% ww for the four studied species, i.e., the blue-lined large-eye bream -GMW, the yellowtail emperor -ICZ, the sky emperor -LTQ and the slender emperor -LHV). Notably, TLC values in the humpback red snapper (LJG) were seven times higher than in the other species of the Lutjanidae family, although no major differences in their trophic groups were noted. Another notable result was the difference in TLC values between the two herbivorous species, the blue-barred parrotfish (Scaridae) and streamlined spinefoot (Siganidae), at 0.3 ± 0.1% ww and 2.0 ± 1.4% ww, respectively. Both species feed on algae and have similar trophic positions (based on their $\delta^{15}N$ values) but are characterized by different $\delta^{13}C$ values, which suggests that they feed on different sources (i.e. rock and coral scraper versus algae grazer). Globally, the TLC values did not show any obvious relationship with either the species' trophic level nor their habitat or trophic group. More knowledge on the physiology of the studied species would be necessary to understand the high inter-species (and in some cases inter-individual) variabilities in TLCs observed in our amples.

As expected for lipophilic contaminants, the majority of PCB and OCP individual concentrations showed positive significant correlations with TLC. However, when all data were considered, the relationships were highly influenced by the swordfish samples (showing high TLCs and high PCB and OCP concentrations), which were therefore removed from the correlation study. Among the various lipophilic contaminants studied, only the lower-chlorinated PCBs (CB-18 to CB-52), α-HCH and aldrin did not show any significant correlation with TLC (Table S3).

3.3. Bioaccumulation of OHCs in tropical species from the Seychelles waters

3.3.1. OHC are moderately to highly detected in tropical reef species

The majority of the targeted OHCs were found above the LOQ, although their detection frequencies varied greatly between individual compounds (Tables S4 to S6). Among PCBs (Table S4), hexa- to octa-chlorinated congeners, and among them CB-138, CB-153 and CB-180, exhibited the highest detection frequencies (>70%), which is consistent with their higher persistence and propensity to bioaccumulate (Corsolini et al., 2007; Munschy et al., 2016, 2020). The OCPs *p,p'*-DDE and mirex showed the highest detection frequencies (94% and 93% respectively), followed by β-HCH (78%) and dieldrin (70%) (Table S5). The high persistence and long-range transport propensity of the legacy PCBs and OCPs explain their high occurrence in our samples. As for PFAS, the odd-chain length PFUnDA and PFTrDA were the most frequently quantified ones (83% and 80% respectively), while other PFCAs and PFOS were quantified in 41–50% and 42% of the samples (Table S6). The high prevalence of long-chain PFCAs compared to PFOS has already been reported in top predator pelagic fish species from the WIO (Munschy et al., 2020) and explained by PFCA accumulation in oceans (Gonzalez-Gaya et al., 2014).

3.3.2. OHCs showed highly variable inter-species relative contributions

Large differences in PCB, OCP and PFAS contributions were observed between the various families (Fig. 2) and species (Fig. S1). While PFAS were clearly predominant (68–96%) in the Octopodidae (big blue octopus - OQC), the Palinuridae (pronghorn spiny lobster - NUP) and the four Scombridae species (dogtooth tuna - DOT, Indian mackerel - RAG, yellowfin tuna- YFT and bigeye tuna - BET), PCBs were the predominant contaminants in the two herbivorous species, USY and IGA (74% and 65% respectively) and OCPs were predominant in the Istiophoridae (striped marlin - MLS) and the Xiphiidae (swordfish - SWO) (70% and 84% respectively) (Fig. 2). Although these differences could be due to various factors such as the species' habitats and trophic preferences, the muscles' biochemical composition, which differs between the studied species (i.e. lipids versus protein content) could also have a major influence. For example, both striped marlin and swordfish showed significantly higher TLC values (3.1% ww and 9.4% ww respectively) than the tuna species (0.4%), despite sharing a similar habitat and feeding habits, which resulted in higher contributions of OCPs in the former species and predominating PFAS in the tunas (Munschy et al., 2020). Similar PFAS contributions relative to tuna were observed in the other two Scombridae species, the Indian mackerel and dogtooth tuna, although they do not share the same feeding preferences as the large top predator species. The higher trophic level of striped marlin and swordfish combined with the lower propensity of PFAS to biomagnify compared to OCPs might also explain the observed contributions. However, no significant tendency between the different OHC families' contributions and $\delta^{15}N$ values was observed at the species level.

3.3.3. OHC concentrations were highly variable among species

PCB concentrations varied widely across families (Fig. S2) and species (Table S7). The $\sum_{34}$ PCB mean concentrations ranged from 0.57 ± 0.53 ng g^{-1} lw in the Palinuridae (pronghorn spiny lobster -NUP, n=2) to 23.22 ± 2.99 ng g^{-1} lw in the Istiophoridae (striped marlin -MLS, n=3). The highest concentrations within each multi-species family were recorded in the bludger (10.28 ± 11.41 ng g^{-1} lw in NGY, Carangidae), the slender emperor (5.10 ± 3.50 ng g^{-1} lw in LHV, Lethrinidae), the deep water longtail red snapper (9.47 ± 3.83 ng g^{-1} lw in ETC, Lutjanidae), the brownspotted grouper (39.02 ± 10.66 ng g^{-1} lw in EFH, Serranidae) and the yellowfin tuna (13.58 ± 12.33 ng g^{-1} lw in YFT, Scombridae). The $\sum_6$ indicator PCBs and the dl-PCBs accounted, on average (n=112), for 56 ± 13% and 9 ± 6%, respectively, of the total quantified congeners. On average, i-PCB concentrations were 10 times above the DL-PCB concentrations. PCB profiles (i.e. the relative contributions of the various congeners) were characterized by the predominance of the higher-chlorinated congeners in most species (79 ± 29% for the summed penta- to decachlorinated congeners). The predominance of hexa- and hepta-chlorinated congeners, the most highly bioaccumulable and recalcitrant to degradation, is commonly reported in marine fish including in large pelagic top predators (Corsolini et al., 2007; Munschy et al., 2016, 2020) and is consistent with the increasing bioamplification of higher-chlorinated PCBs in food chains. However, peculiar profiles were observed in the pronghorn spiny lobster (NUP), in both herbivorous fish species (blue-barred parrotfish (USY) and streamlined spinefoot (IGA)) and in the planktivorous rough triggerfish (CNT), which all showed the predominance of tri- to tetra-chlorinated congeners (between 63 ± 20% and 97 ± 2%, Fig. 3A). The higher contribution of tri- to tetra-chlorinated congeners could be explained by both the lower $\delta^{15}N$ values and the feeding preferences of the species. The four species have low trophic levels (Fig. 1) and, except for the pronghorn spiny lobster, feed on algae and/or zooplankton (Table S1). High relative contributions of tri- and tetra-chlorinated PCB congeners have already been reported in zooplankton and herbivorous fish in various ecosystems (Nie et al., 2005; Covaci et al., 2006). The species feeding preference (i.e. herbivorous / planktivorous versus carnivorous) was therefore a strong driver of the PCB profiles determined in the studied tropical reef ecosystem.

DDTs were below the LOQ in the Palinuridae and seldom detected in the Octopodidae and the Scaridae. In the other species, DDTs were by far the predominant OCPs. ∑ DDT mean concentrations ranged from 0.25 ± 0.13 ng g^{-1} lw in the Siganidae (streamlined spinefoot - IGA) to 78 ± 24 ng g^{-1} lw and 85 ± 28 ng g^{-1} lw in the two billfish (MLS and SWO), respectively (Table S7 and Fig. S2). These results are consistent with our previously published results showing that DDTs were the predominant OHCs in swordfish (Munschy et al., 2020), highlighting the predominance of DDT in pelagic top predators in the WIO. Carangidae, Scombridae and the Coryphaenidae showed intermediate levels (9.46 ± 6.38, 18.78 ± 16.89 and 12.64 ± 8.73 ng g^{-1} lw respectively). Interestingly, the two large pelagic yellowfin tuna and bigeye tuna were more contaminated than the more coastal Indian mackerel and dogtooth tuna (Table S7), both belonging to the Scombridae family, suggesting no coastal sources of DDT in the Seychelles. All large pelagic top predator fish species studied so far, i.e. SWO, MLS, YFT and BET, were therefore characterized by higher DDT concentrations. At the lower end, all isomers were below the LOQ in the three pronghorn spiny lobster (NUP) samples and *p,p′*-DDE was quantified in only one out of the three individuals of both big blue octopus (OQC) and blue-barred parrotfish (USY). Within families, the species with the highest concentrations were similar to those observed for PCBs (i.e. bludger - NGY, Carangidae; slender emperor - LHV, Lethrinidae; deep water longtail red snapper - ETC, Lutjanidae; brownspotted grouper - EFH, Serranidae; and yellowfin tuna - YFT, Scombridae). Among DDT isomers, *p,p′*-DDE made by far the highest contribution (85 ± 11% of the five quantified isomers), followed by *p,p′*-DDT (10 ± 6%) and *o,p′*-DDT (6 ± 5%) (Fig. 3B). As *p,p'*-DDE has a longer half-life and higher accumulation propensity (Binelli and Provini, 2003), this isomer is classically identified as predominant in fish, while *p,p'*-DDD ranks second (Jürgens et al., 2015). In our samples, *p,p′*-DDD accounted for 4 ± 2% only, while *p,p'*-DDT accounted for 10 ± 6% of ∑ 5 isomers. The relatively high proportion of *p,p'*-DDT found in our samples in comparison to those from other oceans worldwide would argue in favour of continuing inputs of DDT in the region. Various publications have reported its past use in various countries surrounding the western Indian Ocean (Chakraborty et al., 2010; Bogdal et al., 2013; Ali et al., 2014; Bouwman et al., 2015) and it is still authorized for use against malaria (Annex B of the Stockholm Convention). However, the *p,p′*-DDE + *p,p′*-DDD/Σ DDT ratio had a mean value of 0.88 ± 0.10, which is characteristic of old DDT inputs (Suarez *et al*., 2013). Dieldrin, mirex and HCHs were the other OCPs quantified in more than 70% of the samples at levels ranging from 0.10 ± 0.03 to 2.48 ± 3.98, 0.04 ± 0.02 to 1.51 ± 1.52 and 0.08 ± 0.04 to 0.57 ± 0.36 ng g^{-1} lw, respectively (mean concentrations ± standard deviation per family). Similarly to what was observed for PCBs, noticeably higher concentrations of dieldrin (but not of DDTs) were recorded in the brownspotted grouper (EFH, Serranidae) (11.81 ± 2.35 ng g^{-1} lw). This species that can be found in seagrass beds, reef slopes or mud bottoms is distinguished by a wider range of habitats than the other species of the Serranidae that inhabit rocky and coral reefs (Table S1), which might explain its peculiar contamination pattern.

As for the lipophilic OHCs, ∑ PFAS concentrations were also highly variable between the studied species and families, ranging from 0.01 ± 0.01 ng g^{-1} ww in the Scaridae and the Siganidae to 0.48 ± 0.34 ng g^{-1} ww in the Xiphiidae (swordfish – SWO) (Table S8 and Fig. S2). Interestingly, PFAS concentrations in the pronghorn spiny lobster (NUP) and the big blue octopus (OQC) were as high as in the most-contaminated fish species such as the tomato hind (EFT) and even the top predators in the Scombridae, Istiophoridae and Xiphiidae, showing that trophic levels were not strong drivers of PFAS bioaccumulation in the studied tropical reef species (see below as well). High PFAS concentrations within the range detected in various organisms from a tropical marine food web have also been reported in cephalopods from the South China Sea (Du et al., 2021). In addition to trophic preferences, the biochemical composition of biological tissues (proteins and polar lipids) is key to explaining the compound-specific affinity of PFAS (Ng and Hungerbuhler, 2014), although the latter might not have such a huge effect in view of the small differences in total lipids and their constituent fatty acids, and in total proteins and their constituent amino acids, observed between the studied species (Sardenne et al., 2020; Jensen et al., 2023). Among PFAS, long-chain PFCAs ($>=C_8$) were by far more predominant than PFOS (Fig. 3C), resulting in a median PFCAs/PFOS concentration ratio of 5, which is consistent with the higher bioaccumulative abilities of these compounds compared to the shorter-chain compounds (Evich et al., 2022). Among the long-chain PFCAs, the concentrations of the odd-numbered PFCAs (PFUnDA and PFTrDA) were on average six times higher than the even-numbered PFCAs (PFDA and PFDoDA). This relative abundance of odd-numbered PFCAs has previously been reported in marine fish (Fujii et al., 2019; Munschy et al., 2020; Aminot et al., 2023) and partly explained by the preferential accumulation of the longer chain PFCAs (i.e., $C_{11} > C_{10}$ and $C_{13} > C_{12}$) (Armitage et al., 2009).

Although PCBs and OCPs are well known to biomagnify in trophic webs (Kelly et al., 2007; Walters et al., 2016), the present study did not reveal many significant relationships between the measured concentrations (in lw) and the individuals' $\delta^{15}N$ values (used as a proxy of trophic levels). As shown in Table S9, only few high-chlorinated PCB congeners (CB-153, -167, -169, -170, -183, -189 and -201) and three DDT isomers (*p,p'*-DDE, *o,p'*-DDT and *p,p'*-DDT) showed significant increase with trophic levels. A lack of significant biomagnification has already been observed in tropical species (Haar et al., 2022) and might be due to the complexity of the studied ecosystem, which includes demersal, pelagic, coastal and offshore species. The high biodiversity of tropical ecosystems, leading to a high diversity of diets and a high tissue turnover, might also explain these results (Borga et al., 2012; Deribe et al., 2013). Similarly, the wide variation in all OHC concentrations across species and families most probably reflects the high diversity in their habitats, diet and feeding preferences (Table S1). Similar results showed extremely high variability between PCB concentrations in fish species of various trophic groups in the Marshall Islands, equatorial Pacific, with contrasting results across trophic groups (Nalley et al., 2023). Similarly to the lipophilic OHCs, most PFAS were not significantly correlated with $\delta^{15}N$ values (Table S9). Although PFOS and long-chain PFCAs are bioaccumulative, their biomagnification is less than for lipophilic contaminants, especially in tropical ecosystems (Loi et al., 2011 ; Pan et al., 2021). Because of their relative water solubility and preferential distribution in blood, PFAS could be eliminated via respiration and show biodilution in aquatic food webs including in tropical ones (Kelly et al., 2009 ; Miranda et al., 2021).

3.3.4. OHC concentrations showed no evidence of risk for human health

Although limited data exist on organic contamination of tropical marine ecosystems, especially in the Indian Ocean, a comparison of the chlorinated OHCs determined in the present study with previously published results was undertaken (Table 1, Table 2). In order to minimize the influence of lipid content on the comparison of concentrations, only normalized concentrations in muscle (either reported or recalculated) were considered for PCBs and DDTs. The results showed that the levels of PCBs and DDTs determined in fish from the Seychelles were globally in the range of those reported in other marine tropical areas in the Indian Ocean (Zanzibar - Tanzania, La Reunion Island, or other regions in the western Indian Ocean), although higher levels were reported in the silver biddy in Zanzibar (Haar et al., 2021; 2022) (Table 1). In the majority of cases, PCB and DDT concentrations in (sub)-tropical fish from the South China Sea stood at the upper end or above the concentrations determined in our study (Hao et al., 2014; Zhang and Kelly, 2018). As for PFAS (Table 2), PFOS concentrations in tropical organisms from the China Sea and the western Atlantic Ocean were far above those determined in our study (Du et al., 2021; Miranda et al., 2021; Pan et al., 2021), likely because of the remote location of the Seychelles relative to direct sources. PFCAs in the Seychelles' ecosystem were in the lower range of those reported elsewhere, although differences were less pronounced than for PFOS, which might emphasize PFCAs' accumulation in oceanic waters due to long-range transport (Prevedouros et al., 2006).

The contamination of the studied tropical species was also compared with existing maximum residue levels in foodstuffs. At the European level, the European Commission has introduced maximum levels for PCBs (dl-PCBs and i-PCBs) in fishery products (EU, 2011). i-PCB mean concentrations were between 140 times (in the Xiphiidae) to 18,000 times (in the Palinuridae) below the maximum levels set by the European Commission for the six priority PCBs in foodstuffs (EU, 2011), i.e. 75 ng g^{-1} ww in muscle. Total PCB concentrations were several orders of magnitude (up to 60,000 times in Lethrinidae) below the maximum values set by the governments of Japan and Australia (500 ng g^{-1} ww) and the US (2,000 ng g^{-1} ww) (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 values eleven times (in the Xiphiidae) to 657,000 times (in the Balistidae) below the maximum level set by the European Commission for dl-PCBs in foodstuffs (EU, 2011), i.e. 3 pg TEQ g^{-1} ww in muscle, and also below (3 to 173,000 times) the guideline value of 0.79 pg TEQ g^{-1} ww set by Canada to protect wildlife consumers of aquatic biota (CCME, 2001). For DDTs, the maximum levels found in our study were three to six orders of magnitude below the values of 3,000 ng g^{-1} ww and 1,000 ng g^{-1} ww, respectively set by Japan and Australia for *p,p'*-DDE (Vizzini et al., 2010). ∑ DDT mean concentrations were 3 to 7 orders of magnitude below the maximum admissible concentrations of 5 µg g^{-1} ww set by the US FDA for human consumption. Globally, these results suggest that no risk to humans from consumption of these organisms (muscle for fish), including the most contaminated ones (SWO), was expected.

In addition to regulations on fishery products, environmental quality standards (EQSs) have been established within the EU Water Framework Directive (WFD) for HCB and PFOS to protect humans (via contaminated fish consumption). In our samples, mean HCB concentrations were between 34 times (in the Xiphiidae) and 1,843 times (in the Lethrinidae) below the EQS of 10 µg kg^{-1} set to protect human health. PFOS was also below the EQS of 9.1 ng g^{-1} ww in the Xiphiidae and the Siganidae (154 to 2,191 times, respectively).

4. Conclusion

This study provides the first data on the contamination of coastal and offshore tropical species of different trophic levels and habitats from the western Indian Ocean by major OHCs. By studying a large diversity of species belonging to major trophic groups and habitats of the Seychelles, our results provide a large overview of the contamination of this ecosystem by major OHCs. Despite being far from major direct contamination sources, the targeted OHCs showed moderate to high detection frequencies, although very low levels were found. Due to the specificity of tropical ecosystems (high biodiversity, diversity in diet, quick tissue turnover), wide variations in concentrations were found across species and families. The species trophic levels were not found to be major determinants of OHC concentrations. OHC profiles were strongly influenced by species trophic groups, with the herbivorous species showing the predominance of lower-chlorinated PCBs while oceanic large top predators were characterized by a higher contribution of DDTs. The results showed levels of contamination that were several orders of magnitude lower than regulations for food consumption in all species, indicating no health risk from human consumption of these resources. These data are of the utmost importance for SIDS communities such as the Seychelles, relying mainly on ocean food resources. Besides, our results provide reference values for future studies and will help in protecting marine ecosystems and ensuring food security in the context of the current development of human activities and associated increasing contaminant inputs both locally and globally.

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Table 1. Total lipid content (TLC, % ww), Σ PCB and Σ DDT concentrations (ng.g^{-1} lw) in the muscle of (sub)/tropical marine fish from various oceanic regions around the world. Ranges (min/max) or ranges of mean values ± standard deviations are given.

Sampling location / year	**English name**	**Lipids% ww**	**Σ PCBs (ng g^{-1} lw)**	**Σ DDTs (ng g^{-1} lw)**	**Reference**
INDIAN OCEAN (IO)					
Seychelles / 2013-2018	Coastal reef fish (see Table S1)	0.22±0.03/4.52±2.43	0.53±0.17/39.02±10.66	0.25±0.13/15.11±11.24	This study
	Swordfish, blue marlin, bigeye tuna, yellowfin tuna	0.39±0.07/9.39±3.69	3.85±1.63/23.22±2.99	25.05±11.17/84.79±27.78	This study
Western IO / 2013-2014	Swordfish, bigeye tuna, yellowfin tuna, skipjack tuna	0.37±0.19/9.4±5.7	5.30±6.50/9.27±12.80	13.21±10.87/81.36±61.31	Munschy et al., 2020
Reunion Island / 2013	Albacore tuna	1.9±1.6/2.3±1.2	10.2±7.6	28.4±16.1	Munschy et al., 2016
Zanzibar / 2018	Silver-stripe round herring	1.32/1.97	2.33/19.3	10.4/89.0	Haar et al., 2021
	Indian Mackerel	1.24/1.95	3.34/129	15.8/68.5	Haar et al., 2021
	Pickhandle Barracuda	1.50	17.8	31.8	Haar et al., 2021
	Mackerel Tuna	3.62	7.71	27.6	Haar et al., 2021
	Silver biddy	0.41/0.89	0.88/2.80	24.1/194	Haar et al., 2021
	Thumbprint emperor	0.25/0.36	5.68/38.6	8.56/85.0	Haar et al., 2021
Zanzibar / 2019	Silver biddy	1.28	671	417	Haar et al., 2022
	Indian Mackerel	1.27	27.0	37.2	Haar et al., 2022
CHINA SEA (CS)					
East CS / 2011	Omnivorous fish	0.41-14.40	3.02-27.95	--	Shang et al., 2016
	Carnivorous fish	0.34-16.70	6.62-51.53	--	Shang et al., 2016
Singapore Strait / 2011-2012	Pike conger eel	0.3±0.4	232.25	83.4	Zhang and Kelly, 2018
	Marine catfish	0.6±0.5	84.27	54.58	Zhang and Kelly, 2018
	Bamboo shark	0.2±0.02	87.02	32.48	Zhang and Kelly, 2018
	Stingray	0.5	84.78	<	Zhang and Kelly, 2018
	Snapper	0.2±0.01	77.73	36.77	Zhang and Kelly, 2018
	Grunter	0.3±0.2	120.38	80.2	Zhang and Kelly, 2018
South CS / 2013	Brushtooth lizardfish	0.90±0.10	26.4±4.12	16.7±2.96	Hao et al., 2014
	Russel’s mackerel-scad	0.72±0.02	31.9±3.14	24.4±6.21	Hao et al., 2014
	Striped fin goatfish	1.65±0.28	14.3±3.36	10.8±2.81	Hao et al., 2014
	Snakefish	0.46±0.11	48.1±6.83	40.3±4.48	Hao et al., 2014

Truncatetail bigeye	0.66±0.06	24.9±4.53	7.99±2.06	Hao et al., 2014

Table 2. PFAS concentrations (ng.g^{-1} ww) in the muscle of tropical or equatorial marine fish from various oceanic regions around the world. Ranges (min/max), or ranges of mean values ± standard deviations, are given.

Sampling location / year	English name	PFOS	Σ PFCAs (C>8)	Reference
INDIAN OCEAN (IO)	Big blue octopus	0.005/<LOQ-0.005	0.214/0.158-0.310	This study
Seychelles / 2013-2018	Pronghorn spiny lobster	<LOQ	0.275/<LOQ-0.286	This study
	Coastal reef fish (see Table S1)	0.011/0.003-0.037	0.048/0.005-0.168	This study
	Swordfish, blue marlin, bigeye tuna, yellowfin tuna	0.045/<LOQ-0.059	0.268/0.168-0.417	This study
Western IO / 2013-2014	Swordfish, bigeye tuna, yellowfin tuna, skipjack tuna	0.027±0.011/0.051±0.017	0.153±0.060/0.373±0.203	Munschy et al., 2020
CHINA SEA (CS)				
Beibu Gulf, south SC / 2018	Cephalopods	0.666/0.19-1.09	0.544/0.44-0.76	Pan et al., 2021
	Crustaceans	0.658/0.25-1.25	0.943/0.37-1.44	Pan et al., 2021
	Fishes	0.489/nd-1.53	0.396/0.06-1.09	Pan et al., 2021
Quinzhou Bay, south CS / 2018	Cephalopod	1.146	1.229	Du et al., 2021
	Crustaceans	0.547-1.854	0.932-2.276	Du et al., 2021
	Fishes	0.149-2.082	0.088-0.674	Du et al., 2021
ATLANTIC OCEAN (AO)				
Brazil, western AO /	Fish	0.569/0.24-1.20	0.109/0.01-0.18	Miranda et al., 2021

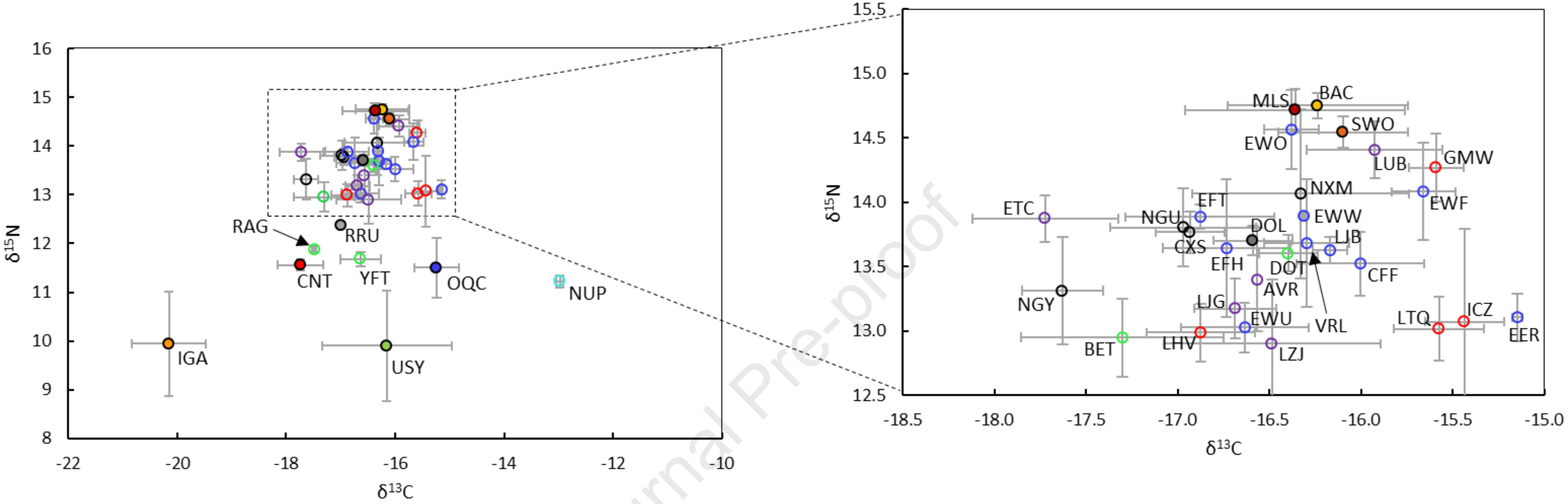

Fig. 1. Food web structure assessed by stable isotope signatures ($\delta^{13}C$ and $\delta^{15}N$ in ‰) in various tropical species collected from the Seychelles waters. Mean values and standard deviations calculated from replicates are indicated. Different colours were used to distinguish the various families. Species names are abbreviated according to the following (according to Table S1 and in alphabetical order): AVR: green jobfish, BAC: pickhandle barracuda, BET: bigeye tuna, CFF: peacock hind, CNT: rough triggerfish, CXS: bigeye trevally, DOL: common dolphinfish, DOT: dogtooth tuna, EER: honeycomb grouper, EFH: brownspotted grouper, EFT: tomato hind, ETC: deepwater longtail red snapper, EWF: brown-marbled grouper, EWO: eightbar grouper, EWU: white-blotched grouper, EWW: longspine grouper, GMW: blue-lined large-eye bream, ICZ: yellowtail emperor, IGA: streamlined spinefoot, LHV: slender emperor, LJB: two-spot red snapper, LJG: humpback red snapper, LTQ: sky emperor, LUB: emperor red snapper, LZJ: humphead snapper, MLS: striped marlin, NGU: yellowspotted trevally, NGY: bludger, NUP: pronghorn spiny lobster, NXM: bluefin trevally, OQC: big blue octopus, RAG: indian mackerel, RRU: rainbow runner, SWO: swordfish, USY: blue-barred parrotfish, VRL: yellow-edged lyretail, YFT: yellowfin tuna.

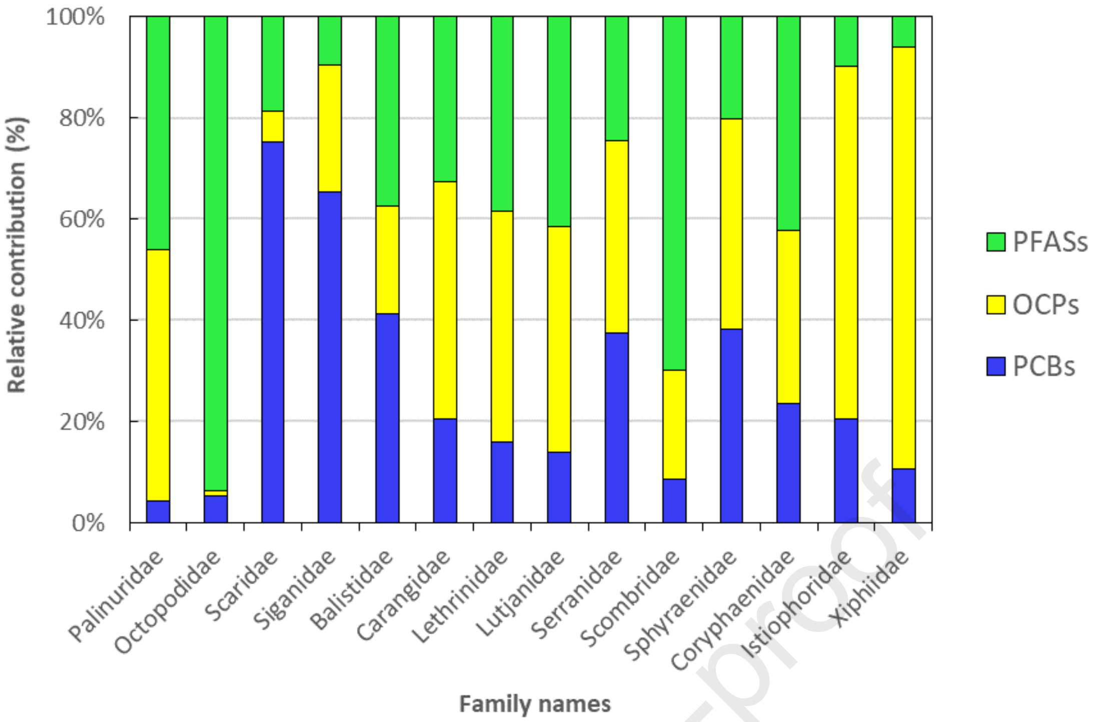

Fig. 2. Relative contribution (%) of OHC (PCBs, OCPs, PFASs) in tropical species (grouped by family) collected from the Seychelles waters.

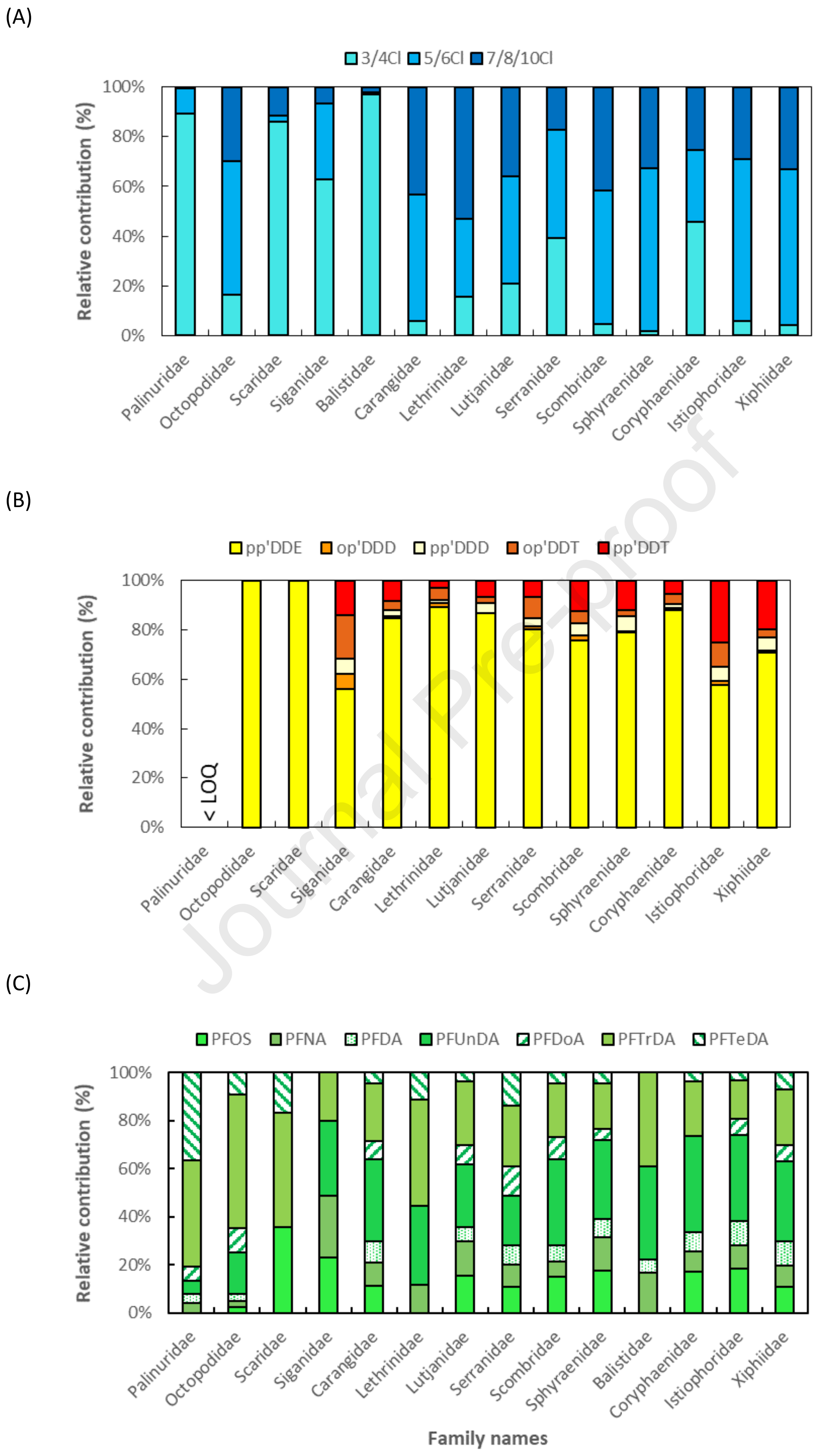
(A)
3/4Cl
5/6Cl
7/8/10Cl
Relative contribution (%)
100%
80%
60%
40%
20%
0%
Palinuridae
Octopodidae
Scaridae
Siganidae
Balistidae
Carangidae
Lethrinidae
Lutjanidae
Serranidae
Scombridae
Sphyraenidae
Coryphaenidae
Istiophoridae
Xiphiidae
(B)
pp'DDE
op'DDD
pp'DDD
op'DDT
pp'DDT
Relative contribution (%)
100%
80%
60%
40%
20%
0%
< LOQ
Palinuridae
Octopodidae
Scaridae
Siganidae
Carangidae
Lethrinidae
Lutjanidae
Serranidae
Scombridae
Sphyraenidae
Coryphaenidae
Istiophoridae
Xiphiidae
(C)
PFOS
PFNA
PFDA
PFUnDA
PFDoA
PFTrDA
PFTeDA
Relative contribution (%)
100%
80%
60%
40%
20%
0%
Palinuridae
Octopodidae
Scaridae
Siganidae
Carangidae
Lethrinidae
Lutjanidae
Serranidae
Scombridae
Sphyraenidae
Balistidae
Coryphaenidae
Istiophoridae
Xiphiidae
Family names

Fig. 3. OHC relative contributions (%) in tropical species (grouped by family) collected in coastal and offshore Seychelles waters. (A) PCBs (% of summed congeners according to their number of Cl atoms), (B) DDTs (% of 5 summed isomers), (C) PFAS (% of summed PFOS and >C_8 PFCAs).

Highlights

- Numerous OHCs detected at low levels but moderate to high detection frequencies
- Large inter-species differences in relative contributions of PCBs, OCPs and PFAS
- Highest levels of DDTs determined in the oceanic top predator species
- Herbivorous species characterized by a higher contribution of 3-4 Cl PCBs
- Long-chain PFCAs (>=C_8) by far predominant in comparison to PFOS

Declaration of interests

☒ The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

☐ The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: