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Seasonal rainfall affects occurrence of organohalogen contaminants in tropical marine fishes and prawns from Zanzibar, Tanzania

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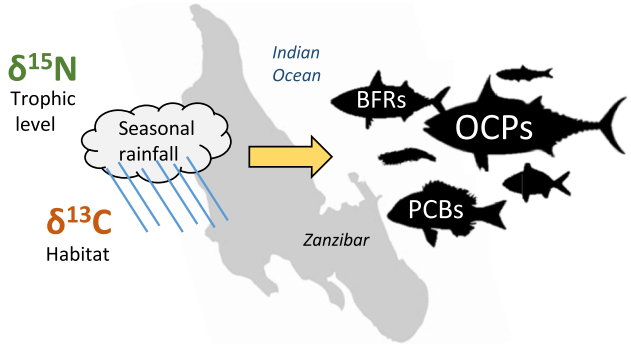
HIGHLIGHTS

- Assessing organic pollutants in a tropical, marine ecosystem where little data exist
- Occurrence of PCBs, BFRs and OCPs varied with trophic level, habitat and season
- Higher relative concentration increase of more mobile congeners after the rainfall
- Seasonal variation in contaminant occurrence suggests a signal of terrestrial runoff.

GRAPHICAL ABSTRACT



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ABSTRACT

Seasonal differences in precipitation may affect contaminant dynamics in tropical coastal regions due to terrestrial runoff of contaminants to the marine environment after the rainy seasons. To assess the effect of seasonal rainfall on occurrence of organohalogen contaminants in a coastal ecosystem, marine fishes and prawns were collected off the coast of Zanzibar, Tanzania in January and August 2018, representing pre- and post-rainy season, respectively. Samples were analyzed for organochlorine pesticides (OCPs), polychlorinated biphenyls (PCBs), brominated flame retardants (BFRs), including polybrominated diphenyl ethers (PBDEs) and emerging BFRs, as well as the dietary descriptors stable isotopes of carbon ($\delta^{13}C$) and nitrogen ($\delta^{15}N$). Across all species and seasons, mean contaminant concentrations ranged from below limit of detection (LOD) to 129 ng/g lipid weight (lw) ΣPCBs; 5.6–336 ng/g lw ΣOCPs; and < LOD –22.1 ng/g lw ΣPBDEs. Most of the emerging BFRs were below LOD. Contaminant concentrations generally increased with higher pelagic carbon signal ($\delta^{13}C$) and higher relative trophic position ($\delta^{15}N$). The ratio of DDE/ΣDDTs in fishes and prawns was lower in August than in January, suggesting runoff of non-degraded DDT into the marine system during or after the seasonal rainfall. Contaminant patterns of OCPs and PCBs, and concentrations of BFRs, differed between seasons in all species. A higher relative concentration-

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increase in lower halogenated, more mobile PCB and PBDE congeners, compared to higher halogenated congeners with lower mobility, between January and August aligns with a signal and effect of terrestrial runoff following the rainy season.

1. Introduction

Environmental fate, food web dynamics and spatial, and temporal variations of persistent organic pollutants (POPs) have been relatively understudied in tropical ecosystems (Kidd et al., 2001; Verhaert et al., 2013). High ambient temperatures and low organic matter content in environmental compartments in tropical regions increase volatilization and reduce the environmental storage capacity of semi-volatile POPs, in contrast to high latitude regions that typically function as contaminant sinks due to long range atmospheric transport and cold condensation (Wania and Mackay, 1993). However, POPs may also be transported via trade of products and waste from developed to developing regions of the world where a lack of appropriate waste management practices may result in environmental pollution (Abbasi et al., 2019; Asante et al., 2011; Breivik et al., 2015; Gioia et al., 2011). In Africa, local sources of pollution including discharge from landfills and waste incineration plants, sewage, wastewater, industry and agriculture play an increasingly important role in environmental pollution pressure (Gioia et al., 2014; Mmochi and Francis, 2003; UNEP/Nairobi Convention Secretariate, 2009).

The Zanzibar islands of Tanzania are representative of a developing region with major identified pressures for the marine environment in the Western Indian Ocean region, including increasing anthropogenic activity and urbanization, overexploitation of fish stocks, destruction of sensitive ecosystems and habitats, and pollution (Johnstone et al., 1998; Mmochi and Francis, 2003; UNEP et al., 1998). Commercial fisheries, small-scale artisanal fishing and an increasing aquaculture industry are important for the economy and food security in Tanzania (Mwakalapa et al., 2018; Wetengere et al., 2008). On Zanzibar, fisheries mostly consist of artisanal fishers using traditional canoes and small vessels, and local fisheries provide an income for around 20% of the population (Feidi, 2005). Dietary intake of fish and seafood represents a major source of contaminants to humans (Darnerud et al., 2006; Djien Liem et al., 2000). Only few studies have addressed POPs in freshwater fish from Tanzanian lakes (Henry and Kishimba, 2006; Mahugija et al., 2018; Mdegela et al., 2009; Polder et al., 2014), and even fewer results are available on POPs in marine fish from coastal Tanzania and Zanzibar (Pratap et al., 2008; Mwakalapa et al., 2018). The occurrence of organochlorine pesticides in the Tanzanian environment has been more extensively documented (Elibariki and Maguta, 2017; Kishimba et al., 2004; Lema et al., 2014; Machiwa, 2010; Mahugija et al., 2017, 2018; Mmochi and Mberek, 1998; Mwakalapa et al., 2018; Mwevura et al., 2020; Müller et al., 2017; Nonga et al., 2011; Polder et al., 2016; Polder et al., 2014), and is still considered a significant pollution issue due to indoor residual spraying, unregulated use and large pesticide stockpiles still residing in the country.

The effect of episodic heavy rain on occurrence of organic contaminants was studied in a temperate food web, resulting in a shift in dichlorodiphenyltrichloroethane (DDT) composition to higher contribution of non-degraded DDT relative to degradation products in post-flood sediments, as well as increased concentrations of total DDTs and total polychlorinated biphenyls (PCBs) in fishes after the flooding event (Stewart et al., 2003). However, little is known about the effects of seasonally heavy rain on contaminant dynamics in tropical food webs (Fu and Wu, 2006; Verhaert et al., 2017). Seasonal rainfall can represent a significant driver of contaminant dynamics and mobility, as atmospheric wet deposition, soil erosion and runoff from land, as well as increased discharge of untreated wastewater may introduce pollutants to the marine environment where they can cause environmental and human health concerns (Fu and Wu, 2006; Gerber et al., 2015; Sorensen et al., 2015).

The present study aimed to quantify concentrations and patterns of legacy and emerging brominated flame retardants (BFRs), PCBs and organochlorine pesticides (OCPs) in tropical marine fishes and prawns, and to assess the effect of seasonal rainfall on contaminant dynamics. We further explore how different feeding habitats and trophic position may affect the contaminant occurrence in the study species. Samples of commercially important fishes and prawns representing different ecological niches were collected from Zanzibar in January and August 2018, representing pre- and post-rainy season, respectively.

2. Materials and methods

2.1. Study area and field sampling

The Zanzibar archipelago is located 25–50 km off the coast of the Tanzanian mainland. The climate in coastal Tanzania is tropical and humid, and the main rainy season usually occurs from March through May. In 2018, the precipitation peak at Unguja Island (6.1357°S, 39.3621°E), Zanzibar, was registered in April and May with 610.8 mm and 528.2 mm average monthly precipitation, respectively, compared to a yearly average of 156.4 mm in 2017 and 166.5 mm in 2018 (Table A1 (OCGS, 2018). Stone Town is located on the west coast of Unguja Island and is the center of the capital, Zanzibar City. In January and August 2018, prawn and six fish species were purchased from small-scale artisanal anglers near Stone Town Harbor in Zanzibar City. Muscle samples from 4 to 8 individuals per fish species and 3–4 pooled samples of approximately 100 g prawn muscle and whole herring were analyzed for organic contaminants, including polychlorinated biphenyls (PCBs), legacy and emerging brominated flame retardants (BFRs), organochlorine pesticides (OCPs), lipid content, and the dietary descriptors stable isotopes of carbon ($\delta^{13}C$) and nitrogen ($\delta^{15}N$). The collected species included silver-stripe round herring (*Spratelloides gracilis*), Indian mackerel (*Rastrelliger kanagurta*), pickhandle barracuda (*Spyraena jello*), and mackerel tuna (*Euthynnus affinis*), representing pelagic, offshore feeders; and prawn (*Penaeus spp.*), silver biddy (*Gerres oyena*) and thumbprint emperor (*Lethrinus harak*), representing demersal, inshore feeders (Richmond, 2011). Herring and prawn were sampled to represent lower trophic level species, pelagic and demersal, respectively. Silver biddy, thumbprint emperor (both demersal species) and mackerel (pelagic) were sampled to represent mid-trophic levels. Barracuda and tuna were selected as pelagic predatory species.

2.2. Ethical clearance and research permission

Research permit was granted by the Office of Chief Government Statistician, Zanzibar. Permission to export samples from Tanzania was granted by the Ministry of Agriculture, Livestock and Fisheries (Tanzania), and permission to import samples to Norway was granted by the Norwegian Food Safety Authority.

2.3. Analyses of stable isotopes

Stable isotopes of carbon and nitrogen can be used as dietary descriptors to characterize an organism's dietary habits integrated over time, and are useful tools providing ecological linkages to contaminant

occurrence and distribution in organisms and food webs (Borgå et al., 2004; Kidd et al., 2001). The heavier nitrogen isotope (^{15}N) is enriched relative to the lighter nitrogen isotope (^{14}N) from prey to predator by 3–5‰, and thus $\delta^{15}N$ reflect relative trophic status. $\delta^{13}C$ reflects the carbon source and change with different photosynthetic pathways in primary producers (e.g. C3 and C4 plants) (Layman et al., 2012). In marine systems, $\delta^{13}C$ can be used to assess gradients of pelagic/benthic and marine/terrestrial carbon sources.

Muscle tissue of fish, prawn and whole, homogenized herring were freeze dried overnight and ground into a fine powder using a mortar and pestle. Pre-weighed samples (1 mg) were sealed in tin capsules and analyzed for carbon ($\delta^{13}C$) and nitrogen ($\delta^{15}N$) isotopes using a Thermo Fisher Scientific EA IsoLink IRMS System (consisting of Flash Elemental Analyses and DeltaV Isotope Ratio Mass Spectrometer) at the Stable Isotope Laboratory of the University of Oslo (UiO:CLIPT). With each sample batch, two calibrated internal laboratory reference materials (GLUT, POPPGLY, Fisher Scientific) were analyzed and used to normalize the data. In addition, calibrated quality control material (JALA, Fisher Scientific) were analyzed for each sample run to assess precision and accuracy of the measurement. For $\delta^{13}C$, reference/control materials were calibrated to the VPDB scale using LSVEC (lithium carbonate, $\delta^{13}C = -46.6‰$) and NBS- 19 (calcium carbonate, $\delta^{13}C = 1.95‰$) (International Atomic Energy Agency, Vienna, Austria). For $\delta^{15}N$, reference/control material were calibrated to the AIR scale using USGS40 (L-glutamic acid, $\delta^{15}N = -4.52‰$) and USGS41 (L-glutamic acid, $\delta^{15}N = 47.57‰$) (United States Geological Survey, Reston, VA, USA). Isotope measurements are reported in delta notations (δ) and in parts per thousand (‰). Stable isotopes were analyzed at the University of Oslo Stable Isotope Laboratory (CLIPT).

2.4. Analyses of organic contaminants

A total of 50 chemicals were analyzed, including 16 organochlorine pesticides: *p,p'* - and *o,p'* -congeners of DDD, DDE and DDT, hexachlorobenzene (HCB), α-, β-, and γ- HCH, heptachlor, oxychlordane, *cis*- and *trans*-chlordane/nonachlor, and mirex; 16 PCBs: CB-28, -52, -74, -99, -101, -105, -118, -128, -136, -138, -153, -156, -170, -180, -183, and -187; 13 polybrominated diphenyl ethers (PBDEs): BDE-28, -47, -99, -100, -153, -154, -183, -196, -202, -206, -207, -208, and -209; and five non-PBDE BFRs: hexabromocyclododecane (HBCDD), hexabromobenzene (HBB), pentabromotoluene (PBT), 2,3-dibromopropyl-2,4,6-tribromophenyl ether (DPTE), and pentabromoethylbenzene (PBEB). The analyses of organic contaminants were conducted at the Laboratory of Environmental Toxicology at the Norwegian University of Life Sciences (NMBU).

2.4.1. Sample extraction and clean-up

Fish and prawn muscle and whole herring were homogenized, and 5 g of pre-weighed sample was used for analyses. Internal standards were added to all samples: 25 μL PCB-29, -112 and -207 (1000 μg/mL) (Ultra-Scientific, RI, USA); 20 μL BDE-77, −119, −181, and $^{13}C_{12}$–209, (Cambridge Isotope Laboratories, Inc., MA, USA). For the first round of extraction, 10 mL distilled water, 2 mL 6% sodium chloride (NaCl), 15 mL acetone and 20 mL cyclohexane were used. For the second round of extraction, 5 mL acetone and 10 mL cyclohexane were used. All samples were homogenized with an Ultra Turrax homogenizer (IKA UltraTurrax T25, IKA Laboratory Technology, Staufen, Germany) followed by ultrasonic sonication (Cole Parmer CPX 750, Vernon Hills IL, USA). The lipid extract was concentrated to 5 mL using a Zymark Turbo Vap II evaporator (Zymark Cooperation, Hopkinton, MA, USA) at 40 °C. Lipid content was determined gravimetrically using a 1 mL aliquot of the lipid extract. Cleanup was done using 96% sulfuric acid (H_2SO_4) (Fluka Analytika, Sigma-Aldrich, St. Louis, USA). The final extracts were put on a sand bath at 40 °C followed by gentle evaporation using N_2, concentrated to a final volume of 0.4 mL and transferred to 2 mL vials for storage until gas chromatography (GC) analysis.

2.4.2. Instrumental analyses

Detailed description of the instrumental analyses can be found in Mwakalapa et al. (2018, and references therein). In short, separation and detection of organochlorine compounds were performed on a HRGC (Agilent 6890 Series) coupled to a MS detector (Agilent 5975C Agilent Technologies) operated in negative chemical ionization (NCI) mode with selected ion monitoring (SIM), and configured with a programmable temperature vaporization (PTV) injector (Agilent Technologies). The compounds were separated on a DB-5 MS column (60 m, 0.25 mm i.d., 0.25 mm film thickness; J&W Scientific). The carrier gas was helium (He) at a 1.3 mL/min constant flow. Detection of tri- through hepta-PBDEs, BDE-28, -47, -99, -100, -153, -154, -183 and non-PBDE BFRs was performed on a HRGC (Agilent 6890 Series; Agilent Technologies), equipped with an auto-sampler (Agilent 7683 Series; Agilent Technologies) and coupled to a MS detector (Agilent 5973; Agilent Technologies). Separation and identification of the compounds were performed on a DB-5 MS column (30 m, 0.25 mm i.d., 0.25 mm film thickness; J&W Scientific). The carrier gas was He at a 1.6 mL/min constant flow. For detection of octa-, nona-, and deca-BDE congeners, extracts (10 μL) were injected on a GCMS (Agilent 6890 Series/5973 Network) configured with a programmable temperature vaporization (PTV) injector (Agilent Technologies). The separation and identification of BDE-209 were performed on a 10 m DB-5-MS column (J&W Scientific, Agilent Technologies). The carrier gas was He at a 1.8 mL/min constant flow.

2.4.3. Quality assurance and quality control

The analytical quality of the laboratory is approved by routinely analyzing different Certified Reference Materials (CRMs), and participation in the Arctic Monitoring and Assessment Program (AMAP) ring test. For every analytical series (17 samples), one blind sample of non-spiked Atlantic cod muscle (*Gadus morhua*) two samples of spiked cod muscle for recovery, three procedural blanks of only solvents and an internal reference sample of harp seal blubber (*Pagophilus groenlandicus*) were included. The recoveries for OCPs were between 86% and 135%, for PCBs between 97% and 107% and for BFRs between 81% and 116%. Results under and over the recovery limit (80%–120%) were corrected for recovery. The limit of detection (LOD) was individually defined as 3 times the signal noise for each analyte. The LODs ranged 0.001–0.017 ng/g wet weight (ww) for OCPs, 0.001–0.07 ng/g ww for PCBs and 0.001–0.016 ng/g ww for BFRs. Individual contaminants below LOD in >60% of the samples per species were removed from the data analyses. For contaminants detected in 60% or more of the samples per species, missing values < LOD were replaced with a random number between 0.5 × LOD and LOD, and further used in statistical analyses and calculations of sums. Detection frequency for each chemical is given in Table A2.

2.5. Data treatment

Multivariate analyses were conducted using the vegan package in R (Jari Oksanen et al., 2019). Principal component analysis (PCA) was run initially to explore trends and tendencies in the dataset (Sparks et al., 1999). In short, PCA is an indirect ordination method for dimension reduction into principal component axes, where each extracted axis reflects as much variation as possible. Individual samples are assigned scores representing linear combinations of the response variables. PCA results are presented as biplots with two uncorrelated axes, where axis one (PC1) accounts for the largest part of the variance of the samples and axis two (PC2) accounts for the largest part of the remaining variance. Response variable loadings (here: contaminant concentrations or relative proportion), are plotted as arrows where direction and length of arrows indicate intercorrelation and variance, respectively.

Arrows with similar orientation are positively correlated, whereas orthogonal arrows are uncorrelated. Long arrows indicate that the parameter shows large variation among the samples. Redundancy analysis (RDA) is a direct ordination method where sample scores are additionally constrained to be linear combinations of a set of explanatory variables. In the present study, RDA was run to assess which explanatory variables (lipid, length, body mass, $\delta^{15}N$, $\delta^{13}C$, season and species) could account for the observed structure and variation in the response variables, i.e. concentrations (ng/g wet weight) or patterns (relative contribution to total concentration). Significance at α-level 0.05 was tested with forward permutation tests. When significant, lipid was included as a covariable in the analyses of concentrations to remove effect of lipid content (lipid% explained up to 16% of the constrained variation in initial analyses of contaminant concentrations). After initial PCA analysis using individual contaminants (not shown), several contaminants were found to be strongly intercorrelated. PCB and PBDE congeners were therefore grouped in homologue groups according to degree of halogenation. OCPs were grouped according to functional groups (ΣHCH, ΣChlordanes, DDE, DDD, DDT).

3. Results and discussion

In total, 65 samples of fish and prawn were analyzed for chlorinated and brominated contaminants, and dietary descriptors. Of the 50 contaminants analyzed for, 18 were below LOD in all samples (Table 1), whereas 12 OCPs, 13 PCBs and 7 BFRs were above LOD in sufficient samples to be included in further data analyses. For the contaminants included in the data analyses, 65 substitutions were made for values below LOD, representing around 3% of the whole dataset. Lipid content in the muscle samples varied between January and August for some species, including the silver biddy (0.89% lipid content in January and 0.41% in August), thumbprint emperor (0.36% in January and 0.25% in August) and herring (1.32% in January and 1.97% in August) (Table 1). Thus the effect of variation in lipid content on contaminant concentration was removed by including lipid as a covariable in the multivariate analyses. Some individuals were characterized as juveniles due to small relative body size (Table 2). Other potential explanatory factors such as sex and age were not included in further data analyses due to an incomplete dataset and resources, respectively. Although sampling was selective to obtain individuals with similar body size (length and weight), differences in size were observed for some species between the seasons, e.g. the mackerel was larger in August compared to January and the silver biddy was larger in January compared to August. However, this was assumed a negligible explanatory factor for contaminant accumulation relative to lipid content, diet and trophic position, which was supported by regression analyses (Fig. A2).

3.1. Dietary descriptors: food web characteristics and seasonal variation

Due to overall low lipid% (<5%), low C/N ratio (<3.6) in all species, and low interspecies variability (Table 2), the $\delta^{13}C$ values were not corrected for lipid content (Post et al., 2007). The spread of the species along the $\delta^{13}C$ gradient indicate a shift in carbon source around −16‰ from pelagic/offshore feeding species, such as the mackerel, herring, tuna and barracuda, to the more demersal/inshore feeders, such as the silver biddy and thumbprint emperor (Fig. 1). Prawn (*Penaeus* sp.) is located in the middle of the $\delta^{13}C$ gradient, with no clear pelagic or demersal signal. The $\delta^{15}N$ gradient represents trophic status signal, with top predators, i.e. tuna and barracuda, having the highest $\delta^{15}N$ values. The prawn has similar $\delta^{15}N$ values as the thumbprint emperor and silver biddy, which might reflect an opportunistic feeding behavior, e.g. scavenging, resulting in elevated $\delta^{15}N$ values. The mackerel can be characterized as a mid-trophic level species, while the rest are characterized as low trophic level species, with some overlap with the mackerel. The tuna and barracuda were not available for sampling in both seasons. However, due to a similar trophic niche in the two predator species, as supported by stable isotopes results, they were included in the multivariate analyses as representatives of a pelagic top predator in January and August, respectively.

In addition to food web characterization, stable isotopes of nitrogen and carbon can indicate eutrophication in coastal ecosystems. While an increase in $\delta^{15}N$ can indicate an increased anthropogenic input of nitrogen into a system, $\delta^{13}C$ may increase in response to increased primary production during eutrophication events (Oczkowski et al., 2014). For prawn, the demersal thumbprint emperor and silver biddy, there was no difference in $\delta^{13}C$ or $\delta^{15}N$ between the seasons. For the pelagic mackerel, $\delta^{13}C$ and $\delta^{15}N$ were higher in August compared to January, while the pelagic herring showed an opposite trend. Due to these contrasting results, no conclusions on eutrophication can be made. However, the factors governing isotope uptake and fractionation in food webs are complex and might challenge the data interpretation.

3.2. Contaminant concentrations and patterns

Mean contaminant concentrations in fishes and prawn were relatively low throughout, with organochlorine pesticides being the most dominant contaminant group in all species and seasons, except for mackerel in August, where ΣPCB was the dominating contaminant

Table 1
Mean (range) contaminant concentrations (ng/g lipid weight (lw)) in fishes and prawn from Zanzibar in January and August 2018. Concentrations in wet weight (Table A3) and concentrations of individual congeners (Table A4) are presented in Appendix.

Species	Season	n	Lipid%	ΣDDT[a]	ΣOCP[b]	DDE/ΣDDT	ΣPCB[c]	ΣPBDE[d]	Σ(PCB/(PCB + PBDE))
Herring (S.gracilis)	January	4*	1.32 (1.10–1.54)	89.0 (76.5–102)	90.4 (77.5–104)	0.79	19.3 (16.8–22.7)	4.09 (3.70–4.50)	0.83
	August	3*	1.97 (1.91–2.02)	10.4 (9.76–10.9)	12.0 (11.4–12.3)	0.66	2.33 (2.29–2.43)	3.28 (2.65–4.43)	0.42
Mackerel (R.kanagurta)	January	6	1.95 (1.08–3.35)	15.8 (4.49–30.6)	26.9 (14.7–36.6)	0.68	3.34 (1.93–4.43)	0.79 (0.33–1.14)	0.82
	August	8	1.24 (0.25–3.89)	68.5 (2.59–262)	73.3 (3.24–277)	0.60	129 (0.56–562)	6.73 (2.61–9.94)	0.75
Barracuda (*S. jello*)	August	6	1.50 (0.91–2.93)	31.8 (28.3–37.6)	34.6 (31.4–41.0)	0.70	17.8 (12.1–31.6)	4.10 (2.32–6.31)	0.81
Tuna (E.affinis)	January	6	3.62 (0.91–10.3)	27.6 (11.1–42.3)	30.9 (13.4–46.5)	0.71	7.71 (3.28–15.6)	7.38 (1.11–23.8)	0.59
Prawn (Penaeus sp.)	January	3*	0.55 (0.45–0.65)	5.60 (3.92–8.76)	5.60 (3.92–8.76)	1	nd	4.15 (1.68–6.82)	0
	August	3*	0.59 (0.37–0.79)	331 (233–457)	336 (237–464)	0.9	116 (87.4–160)	22.1 (17.2–27.8)	0.84
Silver biddy (G.oyena),	January	4	0.89 (0.48–1.12)	194 (3.35–724)	195 (4.07–725)	0.80	0.88 (0.36–1.82)	7.54 (3.06–12.2)	0.13
	August	8	0.41 (0.31–0.59)	24.1 (15.7–41.8)	27.3 (18.1–44.6)	0.66	2.80 (1.43–6.36)	18.8 (1.10–34.4)	0.24
Thumbprint emperor (L.harak)	January	6	0.36 (0.31–0.46)	8.56 (3.15–15.7)	22.2 (17.4–32.2)	1	5.68 (3.97–7.18)	nd	1
	August	8	0.25 (0.18–0.33)	85.0 (22.4–332)	89.6 (26.0–337)	0.60	38.6 (5.76–167)	7.80 (3.63–16.0)	0.65

[a] ΣDDTs: (p,p′-, o,p′-) DDD + DDE + DDT.
[b] ΣOCP: ΣDDTs, ΣHCH, ΣChlordane, HCB. <LOD: α-HCH, oxychlordane, *trans*-chlordane, heptachlor.
[c] ΣPCB: CB-28, -74, -99, -101, -105, -118, -138, -153, -156, -170, -180, -183, -187. <LOD: CB-52, -128, -136.
[d] ΣPBDE: BDE-28, -47, -99, -100, -153, -154, -209. <LOD: BDE-28, -186, -196, -202, -206, -207, -208.
* Number of pooled samples (3-4 samples of pooled individuals of prawn and herring).

Table 2
Biometric measurements, stable isotopes of carbon and nitrogen, and C/N ratio (%C/%N) in marine fish and prawn sampled on Zanzibar in January and August 2018. Values are reported as means and ranges.

Species (common name)	Season	Length (cm) (mean, range)	Weight (g) (mean, range)	$\delta^{13}C$ (mean, range)	$\delta^{15}N$ (mean, range)	C/N	Trophic niche
Silver-stripe herring *Spratelloides gracilis* (dagaa lumbunga)	January	NA	100[a]	−18.6, -(18.7–18.3)	9.7, 9.4–10.4	3.2 3.1–3.2	Low, pelagic
	August	NA	100[a]	−19.8[b]	9.12[b]	3.5 3.5–3.5	
Indian mackerel *Rastrelliger kanagurta*, (kibua)	January	21.3, 19.5–22.1	110.6, 80.7–123.4	−18.9, -(20.2–18.4)	10.7, 10.4–11.0	3.5 3.3–4.1	Medium, pelagic
	August	26.6, 25.3–28.5	257.7, 227.2–298.4	−17.6, -(18.4–17.3)	11.3, 10.2–11.6	3.3 3.1–3.9	
Pickhandle barracuda Spyraena jello, (mzia)	August	44.7, 43.0–46.0	429.5, 366.3–466.9	−17.2, -(17.7–16.8)	13.3, 13.2–13.5	3.4 3.3–3.7	High, pelagic (juvenile)
Mackerel tuna *Euthynnus affinis*, (jodari)	January	56.0, 52,0–64.3	3100, 2500–3500	−17.1, -(18.5–16.3)	13.4, 13.0–13.6	3.4 3.1–3.7	High, pelagic
Prawn Penaeus sp. (kamba)	January	NA	NA	−15.0, -(16.0–14.4)	9.8, 9.6–10.3	3.2 3.3–3.3	Low, demersal (scavenging)
	August	NA	NA	−16.3, -(16.5–15.9)	10.0, 9.9–10.1	3.2 3.2–3.2	
Silver biddy *Gerres oyena*, (chaa)	January	21.5, 21.1–21.9	149.9, 127.1–159.6	−10.7, -(14.2–9.1)	9.4, 8.7–10.6	3.2 3.2–3.4	Low/medium demersal
	August	20.3, 29.0–23.0	119.4, 102.1–170.5	−9.7, -(12.6–8.9)	9.1, 8.2–10.4	3.1 3.1–3.2	
Thumbprint emperor *Lethrinus harak*, (changu)	January	26.9, 26.2–27.3	290.0, 254.2–318.1	−12.3, -(13.6–11.2)	9.6, 9.5–9.7	3.2 3.1–3.3	Low/medium demersal
	August	27.2, 22.5–34.0	348.5, 186.0–645.0	−13.9, -(18.4–11.1)	10.4, 8.5–12.9	3.2 3.1–3.2	

[a] Number of pooled samples (3–4 samples of pooled individuals of prawn and herring).
[b] Pooled samples.

group. Although there were species specific exceptions, for all the species combined, mean concentrations increased from January to August, with January concentrations of 5.60–195 ng/g lw (0.03–1.18 ng/g ww) ΣOCPs, <LOD–19.3 ng/g lw (<LOD–0.25 ng/g ww) ΣPCBs, and <LOD–7.54 ng/g lw (<LOD–0.18 ng/g ww) ΣBFRs. In August, concentration range was 12.0–336 ng/g lw (0.11–1.66 ng/g ww) ΣOCPs, 2.80–129 ng/g lw (0.01–1.55 ng/g ww) ΣPCBs, and 3.28–22.1 ng/g lw (0.02–0.11 ng/g ww) ΣBFRs in all species combined (Table 1).

3.2.1. Organochlorine pesticides (OCPs)

ΣOCPs were higher in August than January for the mackerel, prawn, and thumbprint emperor, but the opposite trend was found for the herring and silver biddy. One individual of silver biddy from January was driving the seasonal difference in pesticide concentrations, with 652 ng/g lw DDE compared to a mean of 16 ng/g lw DDE in the remaining three individuals. This individual had relatively high $\delta^{15}N$ signal (Fig. 1), indicating that individual feeding behavior and diet specialization can lead to large intraspecific variability in contaminant accumulation. ΣDDTs (*p,p′* and *o,p′* isomers of DDD + DDE + DDT) exhibited the highest proportion in the organochlorine pesticide pattern (70% of ΣOCP) in most species, except for mackerel and thumbprint emperor in January, where HCB represented 40% and 61% of ΣOCP, respectively (Fig. 2). Concentrations of chlordanes (*cis*-, *trans*- chlordane/nonachlor), HCH (α-, β-, γ- HCH) and mirex were low (<LOD–2.70, <LOD–1.59 and <LOD–0.66 ng/g lw, respectively) across all species and seasons.

For DDTs, *p,p′* -isomers represented on average 100%, 97% and 93% of total DDE, DDD and DDT, respectively. Thus, herein DDE, DDD and DDT refer to the sum of their para-para (*p,p′*) and ortho-para (*o,p′*) isomers. Technical DDT consists of 75% *p,p′* -DDT, 15% *o,p′* -DDT, 5% *p, p′* -DDE, and <5% other (Zhou et al., 2014), and the ratio of the main DDT metabolite, DDE to ΣDDTs (DDE / (DDD + DDE + DDT)), can reflect recent or historical use of technical DDT. A high ratio (~1) indicates older sources of DDT, i.e. that DDE is the only major DDT component, while a lower ratio indicates recent input of non-degraded DDT (Kidd et al., 2001; Mwevura et al., 2002; Ssebugere et al., 2009).

DDE, DDT and DDD represented 85%, 12% and 3%, respectively, of ΣDDT concentrations in January, and 81%, 13% and 6%, respectively, in August. As DDE represented most of ΣDDT in fish and prawn from the present study, we have no indication of recent DDT use in the marine environment on Zanzibar. However, seasonal difference in composition of ΣDDTs was observed, as the DDE/ΣDDTs ratio was higher in January compared to August in all species (Table 1). The higher DDE/ΣDDTs ratios in January compared to August might indicate transport of recent-use or non-degraded DDT from the terrestrial environment (Ruus et al., 2010). The percent increase in concentration between January and August for DDE, DDT and DDD was 53%, 73% and 282%, respectively. Thus, the difference in DDE/ΣDDTs ratio is most affected by changes in DDD concentrations between seasons relative to changes in DDT concentrations. While DDT is degraded to DDE under aerobic conditions,

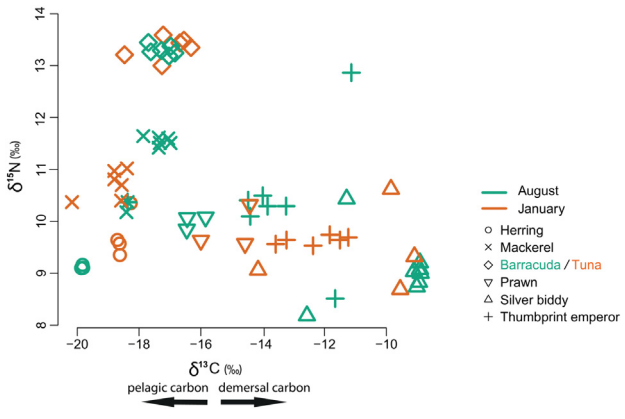

Fig. 1. Stable isotopes of carbon and nitrogen in homogenized muscle tissue, or whole body (herring), of marine fish and prawn sampled on Zanzibar in January and August 2018. The tuna and the barracuda are both representative of a pelagic predator, and different colors indicate that they are sampled from two different seasons (August: green, January: orange).

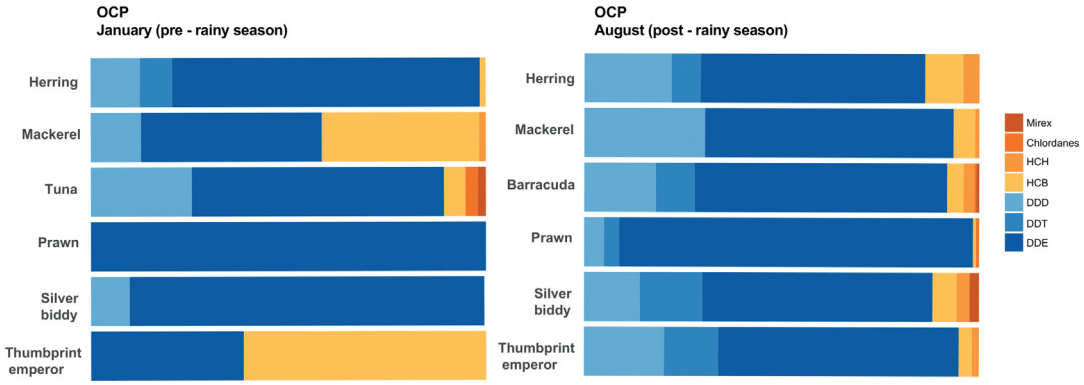

Fig. 2. Relative contribution of each compound to total organochlorine pesticide (OCP) concentration in fish and prawn from Zanzibar, Tanzania in January (pre- rainy season) and August (post- rainy season) 2018.

DDD is the main metabolite under anaerobic conditions (Connor et al., 2007). Anoxic conditions in sediments and the flooding of soils have been shown to promote the reduction of DDT to DDD, and DDD readily accumulates in soil (Castro and Yoshida, 1971; Huang et al., 2001). The seasonal difference in DDE/ΣDDTs ratios may therefore be a result of terrestrial runoff of DDD and, to a lesser degree, DDT runoff from contaminated soils and/or leaching/redistribution from coastal sediments.

3.2.2. Polychlorinated biphenyls (PCBs)

Mean ΣPCB concentrations increased in all species between January and August, except in herring, with highest concentrations in January. The mackerel in August had the highest ΣPCB concentration compared to all other species, but also showed high intra-species variability (mean ΣPCB 129 ng/g lw, range: 0.6–562 ng/g lw). While mean concentrations of hexa- and hepta-CBs increased up to 20 times between January and August in the mackerel, silver biddy, thumbprint emperor and prawn combined (2.2 and 51.7 ng/g lw in January and August, respectively), concentrations of tri-, tetra- and penta-CBs increased up to 60 times between January and August for the four species combined (0.3 and 19.8 ng/g lw in January and August, respectively). In January, PCB-138, PCB-153 (hexaCBs) and PCB-180 (hepta) dominated the overall PCB pattern in most species. The higher chlorinated congeners also dominated the PCB patterns in August, but the lower chlorinated congeners, particularly PCB-28, were found in higher concentrations compared to January, and thus made up a larger part of ΣPCB (Fig. 3). In some species, PCB-28 concentrations were similar to, or even higher than PCB-153, which is unusual as PCB-153 is a more persistent and bioaccumulative congener.

Respiratory and dietary bioaccumulation of PCBs in aquatic organisms occur faster for lower chlorinated congeners with lower octanol-water partitioning coefficient ($\log K_{ow} < 6$), compared to higher chlorinated congeners (Mackay and Fraser, 2000), as has been shown previously in cod (*Gadus morhua*) (Ruus et al., 2012). Thus, the relative concentration increase of lower chlorinated PCBs between January and August could be explained in two ways: differences in rate of dietary and/or respiratory uptake of the various PCB congeners after an increase in total PCB exposure; or, an increase in exposure to lower chlorinated, more mobile PCBs following the rainy season. In accordance with the present study, concentrations of lower chlorinated PCBs in sediment and mullet (*Liza macrolepis*) increased more relative to higher chlorinated PCBs in the estuary of Er-Jen River, Taiwan, after the seasonal rainfall (Fu and Wu, 2006). Furthermore, following the rainy season, the sediment in the estuary was covered with surface soil contaminated with un-weathered PCBs (Fu and Wu, 2006). We observed seasonal change in PCB occurrence with increasing detection of lower chlorinated, more mobile congeners in August compared to January, which could suggest atmospheric wet-deposition and/or discharge of these congeners from terrestrial pools into the marine environment after the seasonal rainfall.

3.2.3. Legacy and emerging brominated flame retardants (BFRs)

Mean ΣPBDE concentration increased between January and August in all species, except herring, where concentrations were comparable between seasons (~4 ng/g lw). HBCDD was only quantified above LOD in mackerel and tuna in January, and prawn and mackerel in August. A high blank signal for HBCDD resulted in the removal of this compound in the mackerel samples from August. None of the emerging BFRs (PTB, DPTE, PBEB, HBB) were detected above LOD. The dominating congeners in the PBDE pattern depended on species and season, with no clear overall trend (Fig. 4). Tri-, tetra penta- and hexa- BDE congeners were dominating the PBDE patterns in fish and prawn sampled in August, and in herring and silver biddy from January. BDE-209 was the

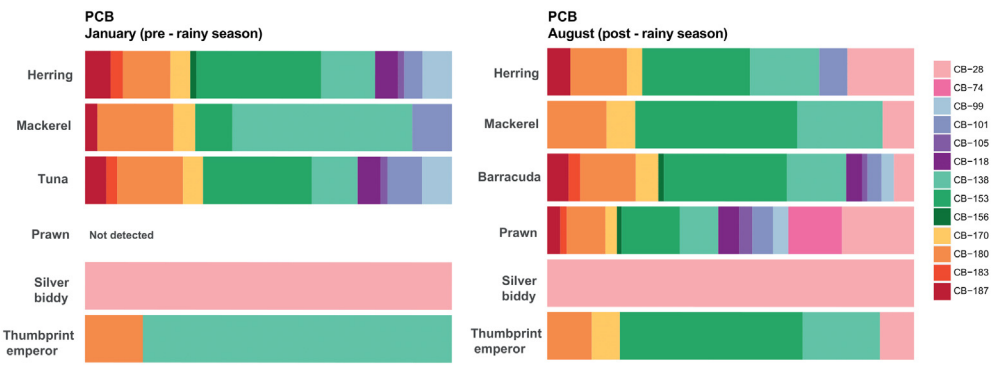

Fig. 3. Relative contribution of each polychlorinated biphenyl (PCB) congener to total PCB concentration in fish and prawn from Zanzibar, Tanzania in January (pre-rainy season) and August (post-rainy season) 2018.

dominating compound in tuna and prawn from January, and in thumbprint emperor from August. Due to its high hydrophobicity (log K_{ow} around 10) BDE-209 has low bioavailability and is known to be efficiently biotransformed by fish. Thus, the detection of BDE-209 in the present study might indicate recent exposure to the commercial mixture deca-BDE (Stapleton et al., 2004). BDE-47 was in general more prominent in August compared to January. BDE-47 is commonly detected in biota due to its effective uptake and resistance to degradation, and is the main component of the widely used penta-BDE mixture (De Wit, 2002). BDE-47 is also an important degradation product resulting from debromination of higher brominated PBDEs, including BDE-209. Species-specific differences in debromination of PBDEs could explain the observed variation in occurrence of PBDEs (Roberts et al., 2011), but is not the scope of the present study. Lower brominated PBDE congeners showed a higher relative concentration increase from January to August compared to higher brominated congeners. Concentrations of BDE-47, 99, 100, 153 and 154 combined increased by about 370% between January and August, while BDE-183, 202 and 209 combined increased by about 67%. Similar to PCBs, an increase in the detection of lower halogenated, more volatile congeners after the rainy season might indicate atmospheric wet deposition or runoff from land.

A ΣPCB/Σ(PCB + PBDE) ratio reflect the contribution of the two contaminant groups to their total, and can be used to infer possible differences in sources. For the herring from August, prawn from January and silver biddy in both seasons, the ΣPCB/Σ(PCB + PBDE) ratio was below 0.5, i.e. PBDE concentrations exceeded PCB concentrations (Table 1). Comparable concentrations of PCBs and PBDEs, or PBDE concentrations exceeding PCB concentrations, are relatively unusual in biota as PCBs are among the most persistent and ubiquitous contaminants found in the environment, despite declining levels following the international ban (Rigét et al., 2019). The dominance of PBDEs over PCBs may be explained by global production and use of PCBs that historically have occurred in industrialized regions in the northern hemisphere (Breivik et al., 2002), while PBDEs are more associated with consumer products that are being introduced to developing regions as a result of global trade and declining use in developed regions (Abbasi et al., 2019).

3.3. Contaminant occurrence in marine fishes and prawns from Zanzibar

In the multivariate analyses of contaminant occurrence, species, $\delta^{15}N$ and $\delta^{13}C$ were significant explanatory variables for OCP and PCB concentrations, but season was only a significant explanatory factor for BFR concentrations (Table 3, Fig. A1). Season was a significant explanatory factor for PCB concentrations only when including two mackerels from August with higher concentrations compared to all other individuals and therefore treated as outliers in the multivariate analyses. A positive association between $\delta^{15}N$ and DDT, chlordanes, mirex and PCBs indicates higher concentrations of these compounds with increasing trophic position. A negative association between $\delta^{13}C$ and DDT and PCBs indicates higher concentration with increasing pelagic influence (Table 3, Fig. A1). $\delta^{13}C$, $\delta^{15}N$ were also significant explanatory variables for PCB patterns. $\delta^{13}C$ was positively associated with PCB-28, suggesting increasing relative contribution of PCB-28 to ΣPCB in the demersal system. $\delta^{15}N$ was positively associated with higher chlorinated congeners (including PCB-187, -153 and -101), suggesting increasing relative contribution of these congeners to ΣPCB with increasing trophic signal (Table 3, Fig. A1). Season was a significant explanatory variable for PCB and OCP patterns, which confirms the observed change in contaminant- and congener composition from January to August.

Concentrations of chlorinated and brominated contaminants in marine fish and prawn from the present study were comparable to concentrations found in tilapia (*Oreochromis* sp.) from Tanzanian lakes (Polder et al., 2014) and in lakes and lagoons from Ghana (Asante et al., 2013). However, tilapia is a relatively low trophic level species and contaminants are likely more concentrated in freshwater systems compared to the ocean off the coast of Zanzibar. ΣDDTs concentration in fish and prawn from the present study were around two orders of magnitude lower compared to predatory fish species in Lake Malawi (Kidd et al., 2001). ΣDDTs concentration in fish and prawn from the present study (0.03–1.67 ng/g ww) were up to four orders of magnitude lower compared to concentrations in marine fish and prawn collected from markets around several coastal cities in the Pearl River Delta region in China (ΣDDTs LOD-52.2 ng/g ww in prawn and LOD-699 ng/g ww in fish) (Guo et al., 2010). ΣPBDE concentrations ranged from LOD-1.11 ng/g ww in prawn and LOD- 5.93 ng/g ww in fish (Guo et al., 2010). ΣPBDE concentrations in fish and prawn from the present study were around three orders of magnitude lower compared to ΣPBDE concentrations in marine fish sampled in Hong Kong that ranged from 0.95–60.6 ng/g ww (Cheung et al., 2008). Even higher ΣPBDE concentrations in fish have been reported in studies targeting freshwater systems connected to dismantling and recycling of electronic waste in the Pearl River Delta, which is a sub-tropical region associated with rapid urbanization and agricultural and industrial development during the last decades (Zhang et al., 2013).

Very few studies are available on organic contaminants in the marine environment of Zanzibar. Shilla (2016) and Mwevura et al. (2020) showed that marine invertebrates and sediments were more contaminated by POPs in the area around Zanzibar harbor relative to areas less affected by anthropogenic activities, suggesting that the sampling location of the present study represents a pollution hotspot on Zanzibar. Mwakalapa et al. (2018) analyzed organic contaminants in farmed and wild milkfish (*Chanos chanos*) and mullet (*Mugil cephalus*), which are relatively low trophic level species, from Zanzibar and the Tanzanian mainland. Lipid normalized liver concentrations of ΣDDTs, ΣPCBs and ΣPBDEs in those farmed milkfish sampled on Zanzibar in

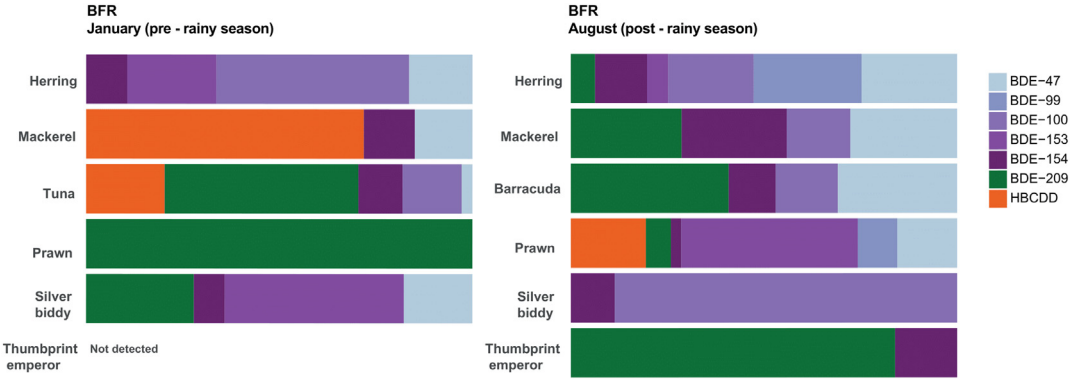

Fig. 4. Relative contribution of each compound to total brominated flame retardant (BFR) concentration in fish and prawn from Zanzibar, Tanzania in January (pre- rainy season) and August (post- rainy season) 2018.

Table 3
Multivariate redundancy analysis of contaminant concentrations (ng/g ww) and patterns (individual compound relative to its respective contaminant group, i.e. ΣOCP, ΣPCB, ΣBFR). Lipid is included as covariate in analyses of concentrations. Grouped contaminants are used as response variables for concentrations, but not for patterns. OCP: ΣHCH. HCB, Mirex, Σchlordanes, ΣDDE, ΣDDD, ΣDDT. PCB: tetra, penta, hexa, hepta-CBs. BFR: tri/tetra/penta, hexa, octa, BDE-209, ΣHBCDD.

	Concentrations				Patterns			
	Explanatory variable	Variation explained	*p*-Value	Total variation explained by RDA	Explanatory variable	Variation explained	p-Value	Total variation explained by RDA
OCP	Season	9%	0.138	38%	Season	12%	0.001	67%
	Species	35%	0.001		Species	34%	0.001	
	$\delta^{15}N$	14%	0.001		$\delta^{15}N$	7%	0.883	
	$\delta^{13}C$	9%	0.001		$\delta^{13}C$	7%	0.782	
PCB	Season	4%	0.083	44%	Season	6%	0.001	77%
	Species	32%	0.001		Species	59%	0.001	
	$\delta^{15}N$	13%	0.001		$\delta^{15}N$	19%	0.001	
	$\delta^{13}C$	9%	0.018		$\delta^{13}C$	22%	0.001	
BFR	Season	6%	0.002	36%	Season	5%	0.320	44%
	Species	24%	0.001		Species	28%	0.001	
	$\delta^{15}N$	9%	0.223		$\delta^{15}N$	4%	0.039	
	$\delta^{13}C$	4%	0.070		$\delta^{13}C$	10%	0.052	

January 2016 were comparable to lipid normalized concentrations in muscle tissue in mackerel, thumbprint emperor and prawn from the present study sampled in January 2018. However, concentrations in the same species sampled in August 2018 exceeded concentrations in the farmed milkfish, which indicates the importance of season when conducting field sampling in tropical regions. Ratios of DDE/ΣDDTs were relatively high in farmed and wild mullet and milkfish (0.8–1) at all sampling sites, and did not indicate recent use of DDT (Mwakalapa et al., 2018). In accordance with the present study, ΣPCB/Σ(PCB + PBDE) ratios were low (0.1–0.8) in farmed and wild milkfish and mullet from Zanzibar and HBCDD was only sporadically detected in wild fish, but with concentrations up to ten times higher than ΣPCBs and ΣPBDEs (Mwakalapa et al., 2018). Higher levels of PBDEs compared to PCBs could indicate that flame retardants from electronics, other consumer products and waste may be more important contributors to pollution pressure on Zanzibar compared to older-type flame retardants and legacy industrial contaminants such as PCBs. Regular monitoring of emerging BFRs should be conducted, as these contaminant could be expected to increase in this region due to regulations and replacements of PBDEs.

4. Conclusion

In the present study, seasonal variation in occurrence of organic contaminants was found in marine fishes and prawn sampled on Zanzibar before and after the main rainy season. Species identity, dietary habitat and relative trophic position were in general dominating factors explaining contaminant concentrations and patterns in marine fish and prawn from Zanzibar. Despite only explaining a low relative proportion of the constrained variation, season explained variability in BFR concentrations, and OCP and PCB patterns. Atmospheric wet-deposition and terrestrial runoff after the rainy season are possible mechanisms resulting in increasing contaminant exposure to marine organisms, particularly exposure to lower halogenated, more mobile compounds. Coastal areas with increasing anthropogenic activities along with lack of appropriate waste management and sewage and wastewater treatment might be particularly vulnerable to seasonal variation in contaminant discharge and runoff, warranting further research on the impact of seasonality in contaminant occurrence also in tropical regions.

CRediT authorship contribution statement

Ane Haarr: Conceptualization, Investigation, Writing – original draft. **Eliezer B. Mwakalapa:** Investigation, Writing – review & editing. **Aviti J. Mmochi:** Supervision, Resources, Writing – review & editing. **Jan L. Lyche:** Supervision, Resources, Writing – review & editing. **Anders Ruus:** Supervision, Writing – review & editing. **Halima Othman:** Conceptualization, Investigation. **Martin M. Larsen:** Conceptualization, Resources, Writing – review & editing. **Katrine Borgå:** Supervision, Conceptualization, Project administration.

Declaration of competing interest

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.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2021.145652.

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