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Spatial Variation in Contaminant Occurrence in Marine Fishes and Prawns from Coastal Tanzania

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Abstract: There are limited data on organic contaminants in marine biota from coastal Tanzania, especially on the occurrence of industrial-use contaminants such as polychlorinated biphenyls (PCBs) and polybrominated diphenyl ethers (PBDEs). The present study, performed in 2018–2019 in coastal Tanzania and Zanzibar Island, aimed at assessing spatial variation in the occurrence of PCBs; brominated flame retardants (BFRs), including PBDEs; and organochlorine pesticides, including dichlorodiphenyltrichloroethane (DDT), among three locations that differ in degree of anthropogenic activity. Analyzed samples included edible tissues of marine fishes and prawns representing different trophic levels and habitats. The results indicate a mainland–island difference, with fishes and prawns collected on Zanzibar having significantly lower PCB and DDT concentrations but higher concentrations of hexachlorobenzene compared to the two mainland locations. The highest contaminant concentrations were found in fishes and prawns collected around central Dar es Salaam harbor, with median ΣPCBs ranging from 22.3 to 577 ng/g lipid weight and ΣDDTs from 22.7 to 501 ng/g lipid weight, suggesting local sources. Concentrations of PBDEs were similar among locations, suggesting more diffuse sources. None of the "newer-type" BFRs, including compounds introduced as replacements for PBDEs, were detected in the present study. Stable isotope values of carbon ($\delta^{13}C$) and nitrogen ($\delta^{15}N$) varied among locations, and the relationship between contaminants and $\delta^{15}N$ varied among locations and habitat (pelagic/demersal). Concentrations measured in the present study are below European guidelines for human consumption of fishes and prawns. However, industrial-use contaminants should be monitored in developing countries because they are contaminants of emerging concern as a result of increasing industrialization and global trade of used products and wastes. *Environ Toxicol Chem* 2022;41:321–333. © 2021 The Authors. *Environmental Toxicology and Chemistry* published by Wiley Periodicals LLC on behalf of SETAC.

Keywords: Tropical ecotoxicology; Spatial variation; Coastal ecosystems; Brominated flame retardants; Organochlorines; Polychlorinated biphenyls

INTRODUCTION

The challenge of environmental pollution is a global issue because pollutants disperse over vast distances and often far from initial production and use. Long-range transport of persistent organic pollutants (POPs) occurs mainly via atmospheric and oceanic currents (Wania & MacKay, 1996) but also via transboundary trade of goods and wastes (Breivik et al., 2015). Following international regulations on production and use of contaminants (Stockholm Convention, http://www.pops.int/), a spatial and temporal shift in contaminant sources is estimated to occur; from emissions from production and products in use to emissions from end-of-life products and waste (Abbasi et al., 2019). A decrease in environmental levels of certain POPs such as polychlorinated biphenyls (PCBs) and polybrominated diphenyls (PBDEs) in industrialized regions is a result of international regulations (United Nations Environment Programme, 2021). Further decline is also hypothesized as a

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result of the controlled and uncontrolled export of goods and wastes from industrialized regions to developing regions of the world (Breivik et al., 2011; Vaccher et al., 2020). Consumer products and wastes, for example electronic waste, contain both elements and organic contaminants and represent a potential threat to the environment and human health when subjected to insufficient waste management and crude recycling practices (Asante et al., 2010; Breivik et al., 2015). Contaminants of emerging concern thus also comprise well-known contaminants that have been in commerce or regulated for several years but are of growing concern in certain regions of the world as a result of increasing use and disposal of products containing these contaminants.

Tanzania ratified the Stockholm Convention on POPs in 2004. Although Tanzania is not a known recipient of bulk waste from developed countries, the rapid economic growth, urbanization, and industrialization render a need for understanding environmental contamination from sources of emerging concern. Despite being banned under the Stockholm Convention, organochlorine pesticides (OCPs), such as dichlorodiphenyltrichloroethane (DDT), are still considered an environmental issue in Africa (United Nations Environment Programme, 2021); and findings of DDT and its metabolites in Tanzania suggest recent use in certain regions and matrices (Mwevura et al., 2002, 2020; Polder et al., 2014). However, knowledge of the occurrence of industrial-use contaminants including PCBs and PBDEs in the Tanzanian environment is scarce, especially in biota (Haarr et al., 2021; Mwakalapa et al., 2018; Polder et al., 2014). Because diet is the main source of contaminant uptake in humans (Vaccher et al., 2020), it is important to monitor contaminant concentrations and patterns in food items. In Tanzania, fish represent an important source of nutrients, and commercial fisheries, small-scale artisanal fishing, and an increasing aquaculture industry are of great importance for the economy and food safety in the country (Wetengere et al., 2008).

The purpose of the present study was to assess spatial variation in occurrence of PCBs; brominated flame retardants (BFRs), including PBDEs; and OCPs in marine fishes and prawns at three locations in coastal Tanzania including both the mainland and the islands of Zanzibar. The locations differ in degree of anthropogenic activity and potential pollution exposure. In addition, we address variation in contaminant occurrence among the study species differing in habitat and trophic level and compare the contaminant concentrations with maximum residue limits (MRLs) and environmental quality standards (EQSs) set by the European Union for contaminant residues in fish products for protection of the environment and human health.

MATERIALS AND METHODS

Study area and field sampling

Samples were collected in January 2018 and 2019. The sampling locations in coastal mainland Tanzania and Zanzibar represent important locations for both small- and large-scale fisheries and differ in anthropogenic activity and potentially contaminant exposure (Figure 1). The East African Coastal Current flows northward along the Tanzanian coast, but the Zanzibar channel is also locally influenced by tidal currents, winds, and gyres (Jahnke et al., 2019; Richmond, 2011). Dar es Salaam is the most populated city in Tanzania and a fast-growing economic center in the region. The fish market in central Dar es Salaam is located by the harbor area, which is associated with heavy traffic from large international container ships, fishing, and public transport vessels. Additional sources of pollution to the Dar es Salaam harbor include uncontrolled disposal of solid and liquid wastes, discharge from polluted rivers and streams, and discharge of untreated industrial and municipal stormwater and sewage (Machiwa, 1992; Tanzania Ports Authority, 2016). The fish market in Kunduchi is located approximately 25 km north of the harbor in Dar es Salaam, with less influence from heavy ship traffic and anthropogenic activity. The fish market on Unguja Island, Zanzibar, is located close to the harbor area of Zanzibar City, which is also a densely populated area but with less industrial activity compared to Dar es Salaam.

Fish was purchased from small-scale artisanal anglers at Malindi fish market on Zanzibar in January 2018 (data presented in Haarr et al. [2021]) and the fish markets at the Dar es Salaam harbor and Kunduchi in January 2019. Muscle samples from four to eight individuals per species (pooled samples of prawn and herring in triplicates) were analyzed for organic contaminants, including PCBs, BFRs, OCPs, and stable isotopes of carbon and nitrogen (Table 1).

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*), thumbprint emperor (*Lethrinus harak*), and white-spotted grouper (*Epinephelus fasciatus*), representing demersal, inshore feeders (Froese & Pauly, 2020; Richmond, 2011). Herring and prawn were sampled to represent low–trophic level species, pelagic and demersal, respectively. Silver biddy, thumbprint emperor, and mackerel were sampled to represent mid-trophic levels, where the latter is a pelagic feeder. Barracuda, tuna, and grouper were

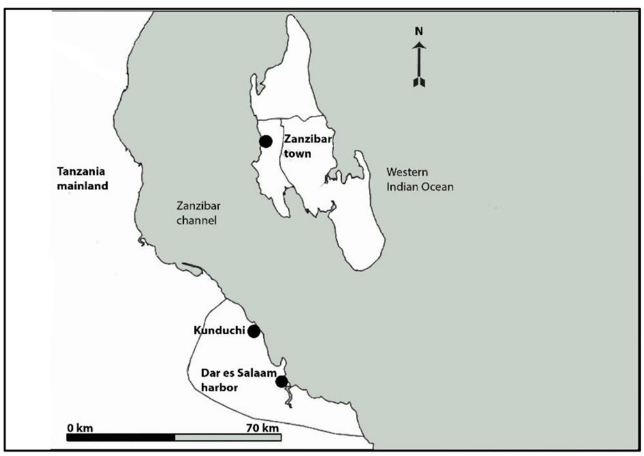

FIGURE 1: Study area and fish marked locations at mainland Tanzania (Dar es Salaam harbor and Kunduchi) and on Unguja Island, Zanzibar.

TABLE 1: Biometrics and stable isotopes in fishes and prawns collected from Dar es Salaam harbor, Kunduchi, and Zanzibar

Species (common name)	Trophic niche	Location	*n*	Length (cm) mean range	Weight (g) mean range	$\delta^{13}C$ mean range	$\delta^{15}N$ mean range	Total C/total *N*
Silver-stripe herring,	Low, pelagic	DAR	1[a]	–	100[a]	−17.9	7.94	0.29
Spratelloides gracilis	Fish	KUN	1[a]	–	100[a]	−17.9	8.13	3.3
(dagaa lumbunga)		ZNZ[2]	4[a]	–	100[a]	−18.6	9.7	3.2
						−(18.7–18.3)	9.4–10.4	3.1–3.2
Indian mackerel,	Medium, pelagic	DAR	6	29.5	267	−17.3	11.7	3.47
Rastrelliger kanagurta				27.0–31.0	197–297	−(17.7–16.9)	11.2–11.9	3.3–3.7
(kibua)	Fish	KUN	6	30.5	306	−17.2	11.8	3.4
				29.0–32.0	256–353	−(17.5–17.0)	11.7–12.0	3.3–3.6
		ZNZ[a]	6	21.3	110.6	−18.9	10.7	3.5
				19.5–22.1	80.7–123.4	−(20.2–18.4)	10.4–11.0	3.3–4.1
Pickhandle barracuda,	High, pelagic	DAR	–	–	–	–	–	–
Spyraena jello	Fish	KUN	6	70.8	1470	−15.8	13.6	3.1
(mzia)				59.0–88.0	835–2300	−(16.2–15.4)	13.2–14.5	3.1–3.2
		ZNZ[b]	–	–	–	–	–	–
Mackerel tuna,	High, pelagic	DAR	–	–	–	–	–	–
Euthynnus affinis	Fish	KUN	–	–	–	–	–	–
(jodari)		ZNZ[a]	6	56.0	3100	−17.1	13.4	3.4
				52.0–64.3	2500–3500	−(18.5 to 16.3)	13.0–13.6	3.1–3.7
Prawn,	Low,	DAR	1[a]	–	100[a]	−14.5	13.5	3.2
Penaeus sp.	demersal	KUN	1[a]	–	100[a]	−17.0	13.7	3.3
(kamba)	(scavenging)	ZNZ[b]	3[a]	–	100[a]	−15.0	9.8	3.2
						−(16.0–14.4)	9.6–10.3	3.3–3.3
Silver biddy,	Low/medium,	DAR	5	19.4	105	−14.1	14.0	2.9
Gerres oyena	demersal			17.0–21.0	69.1–130	−(14.6–13.6)	7.71–16.1	0.29–4.1
(chaa)	Fish	KUN	–	–	–	–	–	–
		ZNZ[b]	4	21.5	149.9	−10.7	9.4	3.2
				21.1–21.9	127–160	−(14.2–9.10)	8.7–10.6	3.2–3.4
Thumbprint emperor,	Low/medium,	DAR	6	23.8	224	−12.9	14.0	3.2
Lethrinus harak	demersal			22.0–25.5	172–273	−(14.1–11.9)	8.88–17.0	3.1–3.4
(changu)	Fish	KUN	6	27.8	329	−15.0	10.6	3.2
				25.5–31.0	275–451	−(18.6–12.4)	7.65–12.0	3.1–3.4
		ZNZ[b]	6	26.9	290.0	−12.3	9.6	3.2
				26.2–27.3	254.2–318.1	−(13.6–11.2)	9.5–9.7	3.1–3.3
Whitespotted grouper,	High, demersal	DAR	4	24.8	243	−13.8	14.8	3.2
Epinephelus fasciatus				17.0–33.0	73.7–499	−(14.9 to 12.9)	13.1–16.9	3.2–3.3
	Fish	KUN	–	–	–	–	–	–
		ZNZ[b]	–	–	–	–	–	–

Trophic niche is determined from the literature (FishBase, Froese & Pauly, 2020) and stable isotope data (pelagic, $\delta^{13}C < -15‰$; demersal, $\delta^{13}C > -15‰$).
DAR = Dar es Salaam harbor; KUN = Kunduchi; ZNZ = Zanzibar; – = data not analyzed.
[a]Pooled samples. Samples of herring and prawn include one to four pooled samples of 100 g homogenized muscle tissue from prawns and whole fish for herring.
[b]Data presented in Haarr et al. (2021).

selected to represent top predators, where the latter is a demersal feeder.

Ethical clearance and research permission

Ethical clearance and a research permit were granted by National Institute for Medical Research (Tanzania) and the Tanzanian Commission for Science and Technology (permit no. 2019-016-NA-2018-251). Permission to export samples from Tanzania was granted by the Ministry of Agriculture, Livestock and Fisheries; and permission to import samples to Norway was granted by the Norwegian Food Safety Authority.

Analyses of stable isotopes

Bulk stable isotope analyses of carbon and nitrogen were carried out at the University of Oslo Stable Isotope Laboratory, as described in detail in Haarr et al. (2021). In short, muscle tissue of fishes and prawns and homogenized whole herring were freeze-dried overnight and ground into a fine powder using a mortar and pestle. 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 the DeltaV Isotope Ratio Mass Spectrometer).

Analyses of organic contaminants

Analyses of organic contaminants were conducted at the Laboratory of Environmental Toxicology at the Norwegian University of Life Sciences. The chemical method used in the present study is accredited by Norwegian Accreditation for analyzing organohalogen contaminants in biological samples according to the requirements of the NS-EN SO/IEC 17025 (test 137). A total of 50 components were analyzed, including 16 OCPs: *p,p′*-/*o,p′* -dichlorodiphenyldichlorethane (DDD), -dichlorodiphenyldichloroethylene (-DDE), -DDT, hexachlorobenzene (HCB), α-/β-/γ-hexachlorocyclohexane (HCH), heptachlor, oxychlordane, *cis-*/*trans*-chlordane/nonachlor, and mirex; 16 PCBs: PCB-28, -52, -74, -99, -101, -105, -118, -128, -136, -138, -153, -156, -170, -180, -183, and -187; 13 PBDEs: BDE-28, -47, -99, -100, -153, -154, -183, -196, -202, -206, -207, -208, and -209; and seven non-PBDE BFRs: hexabromocyclododecane (HBCDD), hexabromobenzene (HBB), pentabromotoluene (PBT), 2,3-dibromopropyl-2,4,6 -tribromophenyl ether (DPTE), decabromodiphenyl ethane (DBDPE), 1,2-bis(2,4,6-tribromophenoxy)-ethane (BTBPE), and pentabromoethylbenzene (PBEB). Sample extraction and cleanup, instrumental analyses, analyte detection, and quality assurance and control are described in detail in Polder et al. (2014). In short, 5 g of fresh, homogenized muscle tissue was used for analyses. Because of the small weight, for herring the whole fish was homogenized. Internal standards were added to all samples: 25 µl PCB-29, -112, and -207 (1000 µg/ml; Ultra-Scientific); 20 µl BDE-77, -119, -181, and $^{13}C_{12}$-209 (Cambridge Isotope Laboratories); as well as solvents for extraction, followed by homogenization with an Ultra-Turrax®. Lipids were extracted twice with cyclohexane and acetone (3:2) using an ultrasonic homogenizer, followed by centrifugation and separation. Cleanup of lipids was done using 96% sulfuric acid, and quantification of lipids was done gravimetrically using 1 ml of sample extract aliquot prior to lipid cleanup. Sample extracts were run on a high-resolution gas chromatograph (Agilent 6890 Series) coupled to a mass-spectrometry detector (Agilent 5975C; Agilent Technologies).

Analytical quality. The laboratory is accredited by Norwegian Accreditation for testing the analyzed chemicals in biological material according to the requirements of the NS-EN ISO/IEC 17025 (test 137). Each analytical series of 24 samples included three blank samples (only solvents), one blind (nonspiked sample of Atlantic cod [*Gadus morhua*] muscle), two spiked samples of Atlantic cod, and the laboratory's own internal reference material of harp seal blubber (*Pagophilus groenlandicus*). The cod samples were spiked with a wide spectrum of the analytes to be analyzed in the study, in different concentrations, with the purpose of calculating the recoveries and clarifying the sensitivity range of the method. The limit of detection (LOD) was set to three times the signal noise for each analyte. Detection frequency for each chemical is given in Supporting Information, Table A1. In addition, quality assurance was obtained by analyzing several certified reference materials and participation in relevant ring tests such as QUASIMEME Laboratory Performance Studies. The results of these tests were within acceptable ranges.

Data treatment

Contaminants detected above the LOD in a minimum of 65% of individual samples per species per location were included in data presentation and statistical analyses, while contaminants detected in <65% of the samples were not included in the data treatment. Contaminant sums (e.g., ΣPCBs and ΣPBDEs) are defined as the sum of all congeners detected in a minimum of 65% of the samples (Table 2).

Statistical analyses were conducted using R (R Foundation for Statistical Computing, 2017). To account for variations in lipid content among species and potential confounding effects, all statistical analyses were conducted using lipid-normalized concentrations. Spatial variation in contaminant concentrations was assessed by combining all species (excluding the silver biddy because this species was not collected from Kunduchi), including one top predator for each location (tuna, barracuda, grouper at Zanzibar, Kunduchi, and Dar es Salaam harbor, respectively). The "ggstatsplots" package was used for statistical testing and visualization of spatial variation in contaminant concentrations (Patil, 2018). Because test assumptions of normal distribution and equal variance were not met, nonparametric Kruskal-Wallis test, followed by Dunn's test for multiple comparisons were used. The *p* values were Holm-corrected. Linear regression models were run to assess the relationship between contaminant concentrations and various explanatory variables, including lipid content, fish size, $\delta^{15}N$, and $\delta^{13}C$ for each location separately. Differences in accumulation, that is, the relationship between $\delta^{15}N$ and contaminant concentration between habitat types (pelagic, $\delta^{13}C < -15‰$; demersal, $\delta^{13}C > -15‰$) were assessed by adding an interaction term to the model:

$$\text{Log10(POP)} = \delta^{15}\text{N} + \text{Habitat} + \delta^{15}\text{N} * \text{Habitat}$$

For significant interactions between $\delta^{15}N$ and habitat, the linear relationship between the contaminant and $\delta^{15}N$ for the two habitats (demersal and pelagic) is shown by two separate regression lines, when significant.

A crude assessment of adequacy for human consumption was done by comparing contaminant concentrations found in fish from the present study to limit values set by the European Commission. The measured levels were compared to the MRLs, which describe the maximum level of contaminant residue that is tolerated in commercial food items, and to EQS set by the European Union water framework directive to protect human health and the most sensitive species in the ecosystem.

RESULTS AND DISCUSSION

A total of 82 individual samples of fish muscle and pooled samples of herring and prawn were analyzed for 52 organic contaminants. Of these, 14 PCBs, 7 PBDEs, HBCDD, and 14 OCPs including β-HCH, γ-HCH, DDTs, HCB, chlordanes, and Mirex were included in further analyses; PCB-128, PCB-136, α-HCH, heptachlor, PBDE-28, PBT, DPTE, PBEB, HBB, DBDPE, and BTBPE were not detected in any samples. Also, PBDE-183, -196, -206, -207, and -208 were detected in <65% of samples and thus removed from data analyses. The instrumental recoveries of contaminants included in analyses were within acceptable ranges (80–120%), except for *p,p′*- DDT, PBDE-202,

TABLE 2: Contaminant concentrations (mean [median], range [nanograms per gram lipid wt]) in fishes and prawns from Dar es Salaam harbor, Kunduchi, and Zanzibar

Species (common name)	Location	*n*	Lipid% mean (median) range	ΣDDTs[a] mean (median) range	HCB mean (median) range	ΣHCH mean (median) range	ΣPCBs[b] mean (median) range	ΣPBDEs[c] mean (median) range	HBCDD mean (median) range
Silver-stripe herring,	DAR	3[d]	0.97 (0.98)	22.2 (22.5)	1.36 (1.35)	0.50 (0.49)	21.3 (20.8)	2.13 (1.58)	nd
Spratelloides gracilis			0.91–1.01	21.0–23.2	1.29–1.44	0.40–0.61	20.0–23.2	1.47–3.33	
(dagaa lumbunga)	KUN	3[d]	1.31 (1.3)	53.1 (52.3)	1.17 (1.16)	0.28 (0.29)	45.9 (45.8)	10.7 (10.9)	nd
			1.29–1.33	51.3–55.7	1.15–1.19	0.20–0.35	47.1–47.7	10.0–11.3	
	ZNZ[e]	4[d]	1.32 (1.32)	89.0 (88.7)	1.34 (1.21)	nd	19.4 (19.0)	4.09	nd
			1.10–1.54	76.5–102	0.92–2.01		16.8–22.7	3.70–4.50	
Indian mackerel,	DAR	6	1.27 (1.24)	37.2 (37.9)	1.1 (1.1)	0.26 (0.28)	27.0 (26.7)	2.54 (2.27)	nd
Rastrelliger kanagurta			0.74–1.88	32.4–41.0	0.9–1.35	nd–0.41	20.5–33.8	1.84–4.19	
(kibua)	KUN	6	1.30 (1.17)	38.0 (38.0)	1.04 (1.02)	nd	34.0 (34.9)	nd	nd
			0.78–2.24	32.9–42.4	0.98–1.15		28.5–37.0		
	ZNZ[e]	6	1.95 (1.75)	15.7 (13.8)	10.7 (9.18)	0.44 (0.33)	3.31 (3.33)	0.78 (0.83)	1.69 (1.64)
			1.08–3.35	4.13–30.6	5.34–21.5	0.19–1.05	1.99–4.59	(0.33–1.14)	nd–4.72
Pickhandle barracuda,	DAR	–	–	–	–	–	–	–	–
Spyraena jello	KUN	6	0.37 (0.35)	47.1 (37.2)	2.61 (2.57)	1.16 (1.03)	44.1 (37.0)	1.21 (1.14)	nd
(mzia)			0.24–0.59	15.8–104	1.80–3.60	0.75–1.77	(12.1–99.1)	0.78–1.73	
	ZNZ[e]	–	–	–	–	–	–	–	–
Mackerel tuna,	DAR	–	–	–	–	–	–	–	–
Euthynnus affinis	KUN	–	–	–	–	–	–	–	–
(jodari)	ZNZ[e]	6	3.62 (2.81)	27.5 (30.4)	1.68 (1.72)	nd	7.05 (6.35)	3.37 (3.37)	1.88 (1.02)
			0.91–10.3	11.1–42.3	1.13-2.12		3.03–14.1	1.1–5.82	0.34–5.98
Prawn,	DAR	3[d]	0.78 (0.78)	104 (105)	1.61 (0.62)	0.48 (0.51)	127 (126)	9.06 (9.06)	nd
Penaeus sp.			0.75–0.81	99.0–107	1.51–1.69	0.36–0.58	126–129	8.81–9.31	
(kamba)	KUN	3[d]	1.01 (1.01)	47.1 (53.7)	2.0 (1.96)	1.80 (1.08)	51.5 (53.1)	4.61 (5.09)	nd
			0.53–1.49	29.4–58.3	1.33–2.73	0.33–4.0	44.9–56.4	3.27–5.47	
	ZNZ[e]	3[d]	0.55 (0.56)	5.61 (4.13)	nd	nd	nd	4.15 (3.9)	nd
			0.45–0.65	3.92–8.77				1.68–6.85	
Silver biddy,	DAR	5	1.28 (1.27)	417 (501)	0.95 (0.93)	0.81 (0.75)	671 (544)	8.1 (7.42)	9.12 (6.42)
Gerres oyena			0.35–1.85	14.7–615	0.87–1.05	0.57–1.32	5.17–1160	3.95–11.1	nd–19.6
(chaa)	KUN	–	–	–	–	–	–	–	–
	ZNZ[e]	4	0.87 (0.94)	194 (24.6)	0.83 (0.81)	nd	0.79 (0.67)	7.49 (7.47)	nd
			0.48–1.12	3.35–724	0.63–1.09		nd–1.82	3.06–11.9	
Thumbprint emperor,	DAR	6	0.81 (0.60)	73.4 (78.2)	0.80 (0.85)	0.47 (0.38)	50.6 (54.5)	0.77 (0.43)	nd
Lethrinus harak			0.33–1.50	7.87–117	0.62–0.93	nd–1.0	7.7–80.7	0.34–1.33	
(changu)	KUN	6	0.63 (0.48)	19.1 (12.4)	0.54 (0.67)	nd	20.5 (12.6)	nd	nd
			0.28–1.26	nd–63.8	nd–0.73		4.37–65.8		
	ZNZ[e]	6	0.36 (0.34)	8.04 (6.56)	13.6 (14)	nd	5.68 (5.84)	nd	nd
			0.31–0.46	nd–15.7	10.1–16.8		3.97–7.17		
Whitespotted grouper,	DAR	4	0.55 (0.47)	276 (222)	2.07 (1.60)	0.87 (0.72)	274 (275)	13.0 (12.4)	nd
Epinephelus fasciatus			0.27–1.04	168–439	1.08–3.93	nd–1.79	167–367	2–29.5	
	KUN	–	–	–	–	–	–	–	–
	ZNZ[e]	–	–	–	–	–	–	–	–

[a]ΣDDTs: *p,p*′, *o,p*′ (dichlorodiphenyldichloroethylene, dichlorodiphenyldichlorethane, and DDT).
[b]PCBs: CB-28, -52, -74, -99, -101, -105, -118, -138, -153, -156, -170, -180, -183, -187.
[c]PBDEs: BDE-47, -99, -100, -153, -154, -202, -209.
[d]Pooled samples.
[e]Data presented in Haarr et al. (2021).
DDTs = dichlorodiphenyltrichloroethanes; HCB = hexachlorobenzene; HCH = hexachlorocyclohexane; PCB = polychlorinated biphenyl; PBDE = polybrominated diphenyl ether; HBCDD = hexabromocyclododecane; DAR = Dar es Salaam harbor; KUN = Kunduchi; ZNZ = Zanzibar; – = data not analyzed; nd = not determined.

and HBCDD in a few analytical series. These were corrected for recovery percentage. Biometric data, trophic niche, and stable isotope values in all collected samples are shown in Table 1. Combining all species, concentrations of ΣDDTs, HCH, HBCDD, and PCBs were highest in Dar es Salaam, followed by Kunduchi and Zanzibar, while HCB concentrations were the highest on Zanzibar (Table 2).

OCPs

The OCPs were dominated by ΣDDTs (DDE, DDD, and DDT), representing on average 73% of ΣOCPs on Zanzibar and 94 and 90% in Dar es Salaam and Kunduchi, respectively. Median concentrations of ΣDDTs among all species ranged from 12.4 to 53.7 ng/g lipid weight in Kunduchi, 4.13–88.7 ng/g lipid weight on Zanzibar, and 22.7–501 ng/g lipid weight in Dar es Salaam (Table 2). Spatial variation was found for ΣDDTs with lipid-adjusted median concentrations (all species combined, except silver biddy) decreasing in the following order: Dar es Salaam > Kunduchi > Zanzibar (Kruskal-Wallis, $\chi^2 = 20.35$, $p = 0.003$; Figure 2). There was no indication of recent DDT usage because the DDT pattern at all locations was dominated by the main degradation product, *p,p*′-DDE, which accounted for between 68 and 100% of ΣDDTs (Figure 3). No MRL for DDT is set for aquatic animals, but a default value of 100 ng/g wet weight, set to "protect the consumer from the intake of unauthorized or excessive levels of pesticide residues" (European Commission, 2005), can be used as a reference. The maximum ΣDDTs concentration found in the present study was 9.26 ng/g wet weight (in silver biddy from Dar es Salaam), and thus well below this threshold (Supporting Information, Table A2).

The second most detected OCP was HCB, in 96% of all samples, with median concentrations among all species ranging from 0.9 to 1.6 ng/g lipid weight in Dar es Salaam, from 0.7 to 2.6 ng/g lipid weight in Kunduchi, and from <LOD to 14 ng/g lipid weight on Zanzibar (Table 2). No samples exceeded the EQS set for HCB at 10 ng/g wet weight (European Commission, 2013). On Zanzibar, HCB was particularly prominent in the OCP patterns of the mackerel (42% of ΣOCPs) and thumbprint emperor (67% of ΣOCPs), which are mid–trophic level, pelagic and demersal species, respectively. Concentrations of HCB were higher on Zanzibar compared to the mainland (Kruskal-Wallis, $\chi^2 = 11.41$, $p = 0.003$; Figure 2). This could indicate spatial variation in OCP exposure between the mainland and Zanzibar. Although HCB is no longer used as a fungicide on Zanzibar, residues from historical application, leaching from obsolete stockpiles, unregulated use, and waste incineration (Adu-Kumi et al., 2010), could represent sources of HCB to the local environment.

Concentrations of OCPs were in the same range as what has been reported in marine (Mwakalapa et al., 2018) and freshwater (Polder et al., 2014) fish from Tanzania, as well as marine predator species collected off the Seychelles (Munschy et al.,

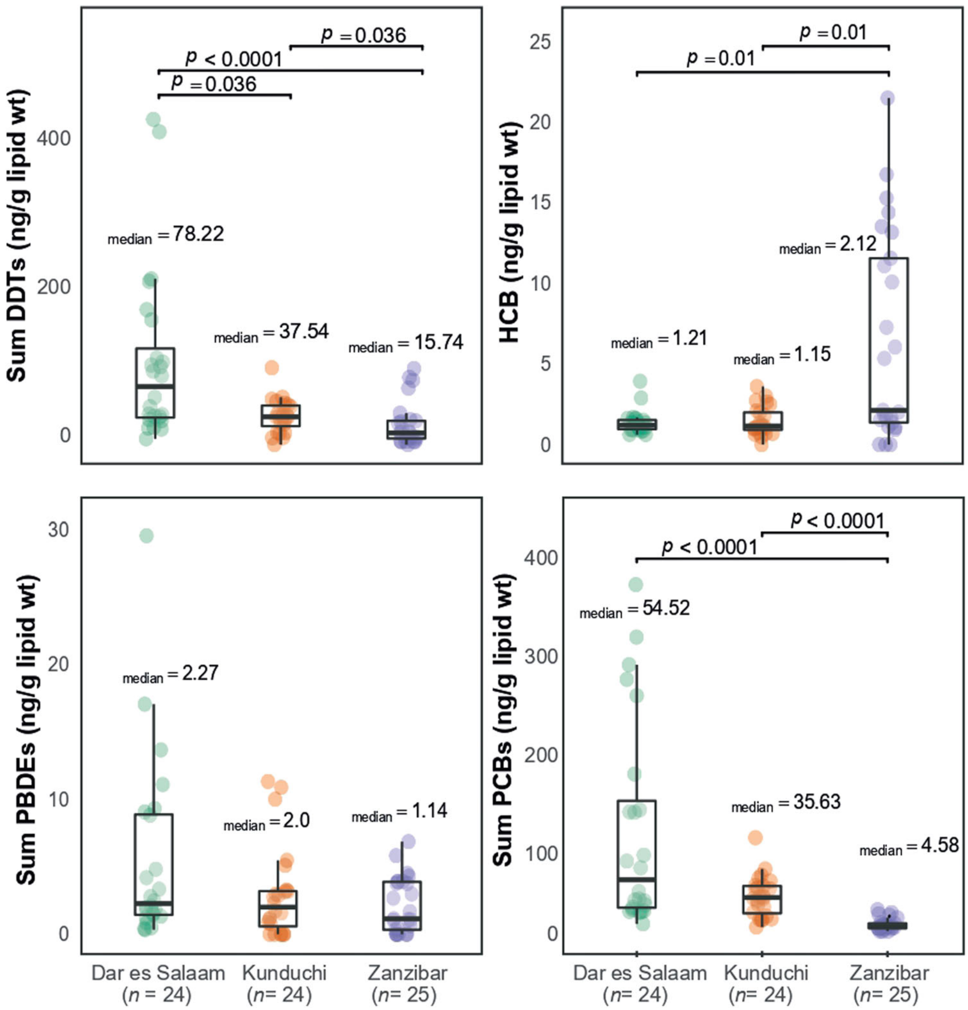

FIGURE 2: Contaminant concentrations in fishes and prawns collected from fish markets at the Dar es Salaam harbor, Kunduchi, and Zanzibar. Significant spatial difference is estimated using the Kruskal-Wallis test, followed by Dunn's pairwise comparisons. *p* values are Holm-corrected and indicate significant difference, where applicable, among the three locations. DDT = dichlorodiphenyltrichloroethane; HCB = hexachlorobenzene; PBDE = polybrominated diphenyl ether; PCB = polychlorinated biphenyl.

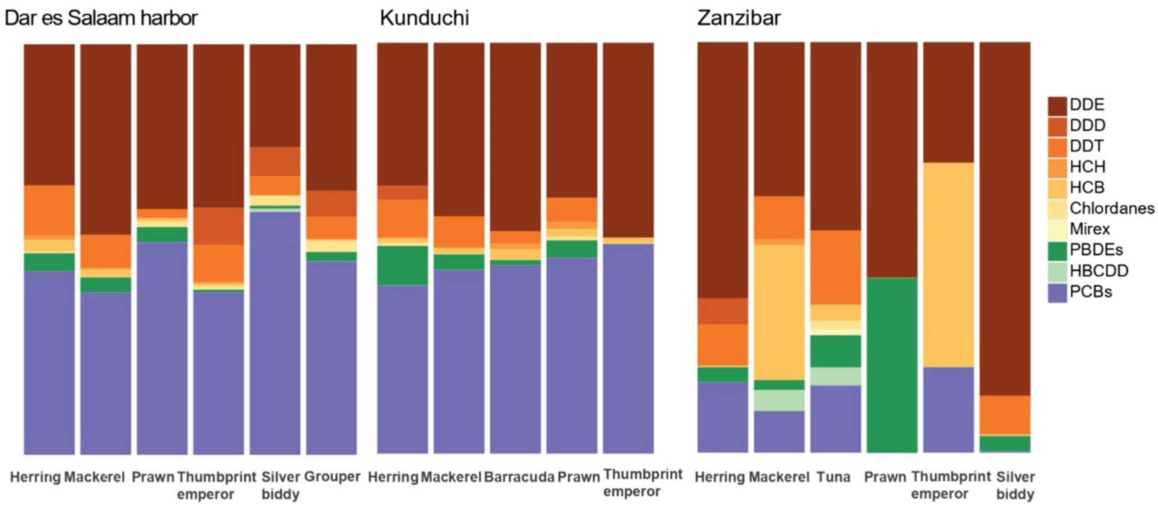

FIGURE 3: Relative contribution of each contaminant group to total contaminant load in marine fishes and prawns collected from fish markets at the Dar es Salaam harbor, Kunduchi, and Zanzibar. DDE = dichlorodiphenyldichloroethylene; DDD = dichlorodiphenyldichlorethane; DDT = dichlorodiphenyltrichloroethane; HCH = hexachlorocyclohexane; HCB = hexachlorobenzene; PBDE = polybrominated diphenyl ether; HBCDD = hexabromocyclododecane; PCB = polychlorinated biphenyl.

2020; Ueno et al., 2003), but lower compared to what has been reported in marine fish collected from the South China Sea (Shi et al., 2013), Italy (Naso et al., 2005), and northern Norway (Bustnes et al., 2012; Table 3). This is attributable to differences in agricultural, domestic, and industrial pollution sources.

Industrial-use contaminants (PCBs and BFRs)

Median concentrations of ΣPCBs among all species ranged from <LOD to 19 ng/g lipid weight on Zanzibar, from 12.6 to 53.1 ng/g lipid weight in Kunduchi, and from 20.8 to 544 ng/g lipid weight in Dar es Salaam (Table 2). The highest ΣPCB concentration was found in one silver biddy from Dar es Salaam (1160 ng/g lipid weight), which exceeded the highest concentration found in the grouper (367 ng/g lipid weight), which is a higher–trophic level species. Congener patterns were similar among all locations, dominated by highly persistent hexa- and hepta-congeners, including PCB-138, -153, and -180. The ΣPCB concentrations differed among locations, with lower levels on Zanzibar compared to the mainland (Kruskal-Wallis, $\chi^2 = 42.9$, $p = 4.9\text{e-}10$; Figure 2). For PCB_6 (six "indicator" PCBs: PCB-28, -52, -101, -138, -153, -180), the MRL for "muscle meat of fish and fishery products" is 75 ng/g wet weight (European Commission, 2011). No samples exceeded this limit as the maximum ΣPCB_6 concentration found in the present study was 9.61 ng/g wet weight (silver biddy from Dar es Salaam).

The ΣPBDEs were similar among locations (Figure 2), median concentrations ranging from <LOD to 7.47 ng/g lipid weight on Zanzibar, from <LOD to 9.06 ng/g lipid weight in Dar es Salaam, and from <LOD to 10.9 ng/g lipid weight in Kunduchi. Congener patterns of PBDEs were dominated by penta-BDE congeners including BDE-47, -99, -100, and -153 in most species. A higher dominance of BDE-209 to ΣPBDEs on Zanzibar relative to the mainland could indicate recent exposure to the commercial deca-BDE mixture. No MRLs exist for PBDEs, but the EQS for Σ_6PBDEs (-28, -47, -99, -100, -153, -154) is set at 0.0085 ng/g wet weight (European Commission, 2013), which is close to the detection limit for several PBDE congeners. Most samples from the present study exceed this limit (Supporting Information, Table A2). Hexabromocyclododecane was only detected in mackerel and tuna from Zanzibar (median concentration 1.64 and 1.02 ng/g lipid wt, respectively) and silver biddy from Dar es Salaam (median concentration 6.42 ng/g lipid wt). The EQS for HBCDD is set at 167 ng/g wet weight (European Commission, 2013), and no samples exceeded this limit. None of the "newer-type" BFRs (PTB, DPTE, PBEB, HBB, DBDPE, and BTBPE) were detected above the LOD in the present study. This could serve as a reference for future studies because these contaminants could be expected to increase in this region because of replacements of PBDEs and increasing urban development, industrialization, and globalization.

In industrialized regions, PCBs are typically the dominant contaminant group compared to PBDEs because of their historical production and use (Breivik et al., 2002). The percent contribution of PCBs and PBDEs relative to the sum of the two (Σ[PCBs + PBDEs]) can be used to identify the importance of the two contaminant groups to the total industrial-use contaminants load and possibly identify a shift between "older" and "newer" types of flame retardants. In silver biddy sampled on Zanzibar, ΣPCBs and ΣPBDEs represented 12 and 87% of Σ(PCBs + PBDEs), respectively. In fishes and prawn from Kunduchi and Dar es Salaam, ΣPCBs represented on average 93% of Σ(PCBs + PBDEs) at both locations, indicating a similarity between the two mainland locations relative to Zanzibar (63%; Figure 3). Less influence from industrialization and urbanization on Zanzibar relative to the mainland could result in less pollution from the older-type industrial-use contaminants, such as PCBs. Similar PBDE concentrations among all three

TABLE 3: Mean contaminant concentrations (nanograms per gram lipid wt) in fishes from other studies

Location	Species	*n*	Year	Tissue	Lipid% (mean)	HCB	HCH	ΣDDTs	ΣPCB	ΣPBDE	Reference
Dar es Salaam, Tanzania	Silver biddy, *Gerres oyena*	5	2019	Muscle	1.28	0.95	0.81	417	671	8.06	Present study[a]
Dar es Salaam, Tanzania	Indian mackerel, *Rastrelliger kanagurta*	6	2019	Muscle	1.27	1.1	0.26	37.2	27.0	2.54	Present study[a]
Zanzibar, Tanzania	Mackerel tuna, *Euthynnus affinis*	6	2018	Muscle	3.62	1.68	nd	27.6	7.71	7.38	Haarr et al. (2021)[a]
Tanzania	Milkfish, *Chanos chanos*	7	2016	Liver	7.81	0.2	0.1	117	0.2	1.3	Mwakalapa et al. (2018)[a]
Tanzania	Mullet, *Mugil cephalus*	8	2016	Liver	4.85	0.2	0.04	73.3	0.6	1.1	Mwakalapa et al. (2018)[a]
Lake Tanganyika	Nile tilapia, *Oreochromis niloticus*	16	2011	Muscle	3.3	1.2	1.1	273	17.2	4.1	Polder et al. (2014)[a]
Lake Malawi	Malawi squeaker, *Synodontis njassae*	5	1996–1997	Muscle	12.8			453.1			Kidd et al. (2001)
Benya lagoon, Ghana	Tilapia, *Sarotherodon melanotheron*	8	2010	Muscle	3.1				150	19	Asante et al. (2013)[b]
Lake Victoria, Uganda	Nile tilapia, *Oreochromis niloticus*	3	2013	Muscle	2.8				22.7		Ssebugere et al. (2014)[c]
Offshore Taiwan	Skipjack tuna, *Katsuwonus pelamis*	3	1999	Muscle	0.9					53	Ueno et al. (2004)[d]
Seychelles	Skipjack tuna, *Katsuwonus pelamis*	5	1999	Liver	3.0	1.7	<0.29	39	14		Ueno et al. (2003)[e]
Seychelles	Swordfish, *Xiphias gladius*	18	2013–2014	Muscle				81.4	9.3		Munschy et al. (2020)[c]
South China Sea	Yellowfin tuna, *Thunnus albacares*	6	2017	Muscle	0.28			169.5			Sun et al. (2020)
Zhoushan, East China Sea	Bullet mackerel, *Auxis rochei*	6	2011	Muscle	16.7				22.9	10.1	Shang et al. (2016)[f]
Pearl River delta, South China Sea	Various fish species	19	2004	Whole body (?)	2.4		8.3	3330		79.2	Guo et al., (2008)[g,h]
South China Sea	Chub mackerel, *Scomber japonicus*	7	2010	Muscle	1.98	3.03	94.4	423.7	2152		Shi et al. (2013)[i]
Gulf of Naples, Italy	Atlantic mackerel, *Scomber scombrus*	10	2003	Muscle	4.47	15.6		180.9	1005		Naso et al. (2005)[j]
Øksfjord, northern Norway	Atlantic cod, *Gadus morhua*	10	2007	Liver	42.8	34.8	2.81	230.3	625.1	20.5	Bustnes et al. (2012)[k]

[a] PCB_{16} (-28, -52, -74, -99, -101, -105, -118, -128, -136, -138, -153, -156, -170, -180, -183, -187); $PBDE_{13}$ (-28, -47, -99, -100, -153, -154, -183, -196, -202, -206, -207, -208, -209).
[b] PCB_{16} (-28, -52, -70, -74, -99, -101, -105, -110, -118, -138, -149, -153, -170, -180, -183, -187); $PBDE_{13}$ (-15, -28, -47, -49, -66, -100, -154, -155, -197 + 204, -206, -207, -208, -209).
[c] PCB_{6i} (PCB-28, -52, -101, -138, -153, -180).
[d] $PBDE_{11}$ (-3, -15, -28, -47, -99, -100, -138, -153, -154, -183, -209).
[e] PCB_{49} (-41 + 64, -42, -44, -47, -49 + 69, -51, -52, -53, -66, -70, -91 + 95, -102, -84 + 92 + 90, -101, -99, -83, -97 + 113, -87 + 117, -85, -82 + 120 + 110, -118, 105, -144 + 149, -134, -133, -132, -128, -153, -141, -137, -138, -159, -156, -176, -178, -187, -285 + 183, -177, -173, -172, -180, -170, -202, -200, -198, -201, -195, -194).
[f] PCB_{6i}; $PBDE_9$ (-28, -47, -66, -85, -99, -100, -153, -154, -183).
[g] Median values.
[h] $PBDE_{15}$ (-3, -15, -28, -47, -60, -85, -99, -100, -138, -153, -154, -183, -197, -207, -209).
[i] PCB_{27} (-1, -8, -18, -28, -29, -44, -50, -52, -66, -77, -87, -101, -104, -105, -118, -126, -128, -138, -153, -154, -170, -180, -187, -188, -195, -200, -206).
[j] PCB_{20} (-28, -52, -66, -74, -99, -101, -105, -118, -128, -138, -146, -153, -170, -177, -180, -183, -187, -194, -196, -201).
[k] PCB_{24} (-28, -52, -47, -74, -66, -101, -99, -110, -149, -118, -153, -105, -138, -187, -183, -128, -156, -157, -180, -170, -196, -189, -194, -206); $PBDE_4$ (-47, -100, -99, -154).
DDTs = dichlorodiphenyltrichloroethanes; HCB = hexachlorobenzene; HCH = hexachlorocyclohexane; PCB = polychlorinated biphenyl; PBDE = polybrominated diphenyl ether; nd = not determined.

locations suggest more diffuse sources and relatively low input from local sources.

Relatively low levels of PCBs in fishes and prawn from Zanzibar are in accordance with findings of organochlorines in blubber of bottlenose (*Tursiops aduncus*) and spinner (*Stenella longirostris*) dolphins from Zanzibar, with PCBs <LOD, while methoxylated PBDEs were quantified at high levels, ranging from 0.6 to 210 mg/g lipid weight (Mwevura et al., 2010). In Ghana, West Africa, mean ΣPCB concentrations in muscle tissue of tilapia fish, which is a relatively low–trophic level species, ranged from 22 to 150 ng/g lipid weight (Asante et al., 2013), which is higher than what was found on Zanzibar and Kunduchi in the present study but comparable to fishes and prawn collected from the Dar es Salaam harbor (Table 3). This might indicate similarities in PCB sources in central Dar es Salaam and Ghana, such as waste disposal practices. Concentrations of PCBs in two mackerel species from areas influenced by industrial and urban pollution (China and Italy) exceeded concentrations in mackerel from Dar es Salaam (present study) by up to 2 orders of magnitude (Table 3). In tilapia fish from Ghana, ΣPBDE concentrations ranged from 0.64 to 52 ng/g lipid weight (Asante et al., 2013), exceeding concentrations in all samples from the present study, which might be a result of a larger e-waste recycling industry in Ghana compared to Tanzania (Asante et al., 2010). It is important to remember, however, that evaluation of spatial variation in contaminant occurrence using different species must be done with care because species-specific variations in contaminant accumulation may vary as a result of, for example, trophic position.

Stable isotopes of carbon and nitrogen: Variations within and among locations

Because of overall low lipid content (<5%; Table 2), low C/N ratio (<3.6; Table 1), and low interspecies variability, the isotope values were not corrected for lipid content (Post et al., 2007). The $\delta^{13}C$ values suggest a separation of pelagic and demersal feeding organisms at −15‰ (Figure 4). There were no clear differences in $\delta^{13}C$ values among locations. The species-specific $\delta^{15}N$ values were higher in fish collected from Dar es Salaam compared to Zanzibar. Median $\delta^{15}N$ values in the thumbprint emperor collected in Dar es Salaam (15.1‰) were higher compared to individuals sampled from Zanzibar (9.6‰; Kruskal-Wallis, $\chi^2 = 6.47$, $p = 0.039$). Median $\delta^{15}N$ values in mackerel collected on Zanzibar (10.7‰) were lower than those in mackerel from the two mainland locations (11.2 and 11.9‰ in Dar es Salaam and Kunduchi, respectively; Kruskal-Wallis, $\chi^2 = 11.56$, $p = 0.003$). Contrary to our expectations, the silver biddy and thumbprint emperor sampled in Dar es Salaam had among the highest $\delta^{15}N$ values, with relatively large variation (Figure 4), suggesting specialized individual feeding behavior. They are both demersal feeders, known to feed on small organisms like crustaceans, polychaetes, and mollusks along shallow, sandy lagoons or coral reefs, and are considered to be low– to mid–trophic level species (Froese & Pauly, 2020). However, their $\delta^{15}N$ values were comparable to the grouper from Dar es Salaam and higher compared to tuna from Zanzibar and barracuda from Kunduchi, which are all considered top predators in the marine system. Spatial variation in $\delta^{15}N$ values could be caused by dietary differences, with individuals from Dar es Salaam feeding on a higher trophic level compared to the same species from other locations; or it can be due to differences in $\delta^{15}N$ at the base of the food web, causing a systematic shift in values through the food web. Baseline $\delta^{15}N$ could vary among locations because of differences in species composition in the lower trophic levels, that is, primary producers/consumers, or differences in nutrient input to the marine system (Guzzo et al., 2011), for example, sewage/industrial wastewater discharge and urban/agricultural runoff in the harbor area of Dar es Salaam.

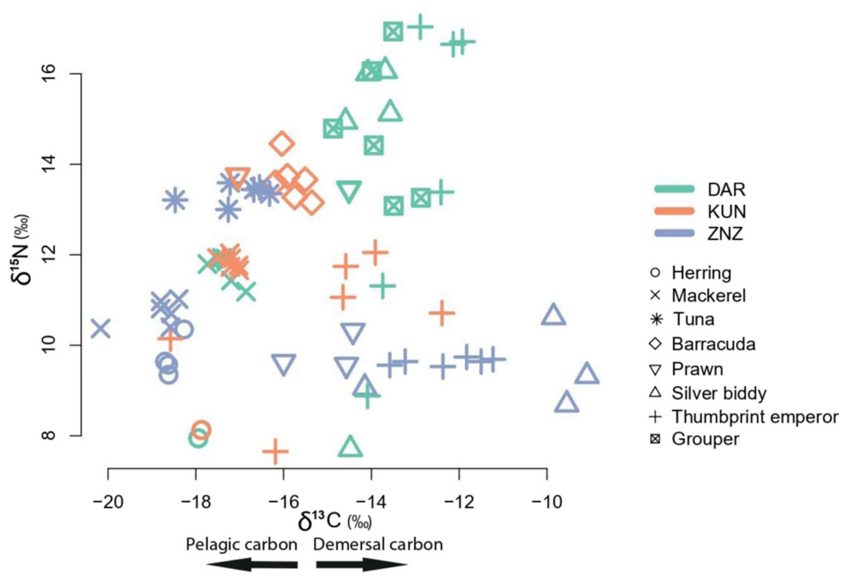

FIGURE 4: Stable isotopes of carbon and nitrogen in various marine species collected from fish markets at the Dar es Salaam harbor, Kunduchi, and Zanzibar. DAR = Dar es Salaam harbor; KUN = Kunduchi; ZNZ = Zanzibar.

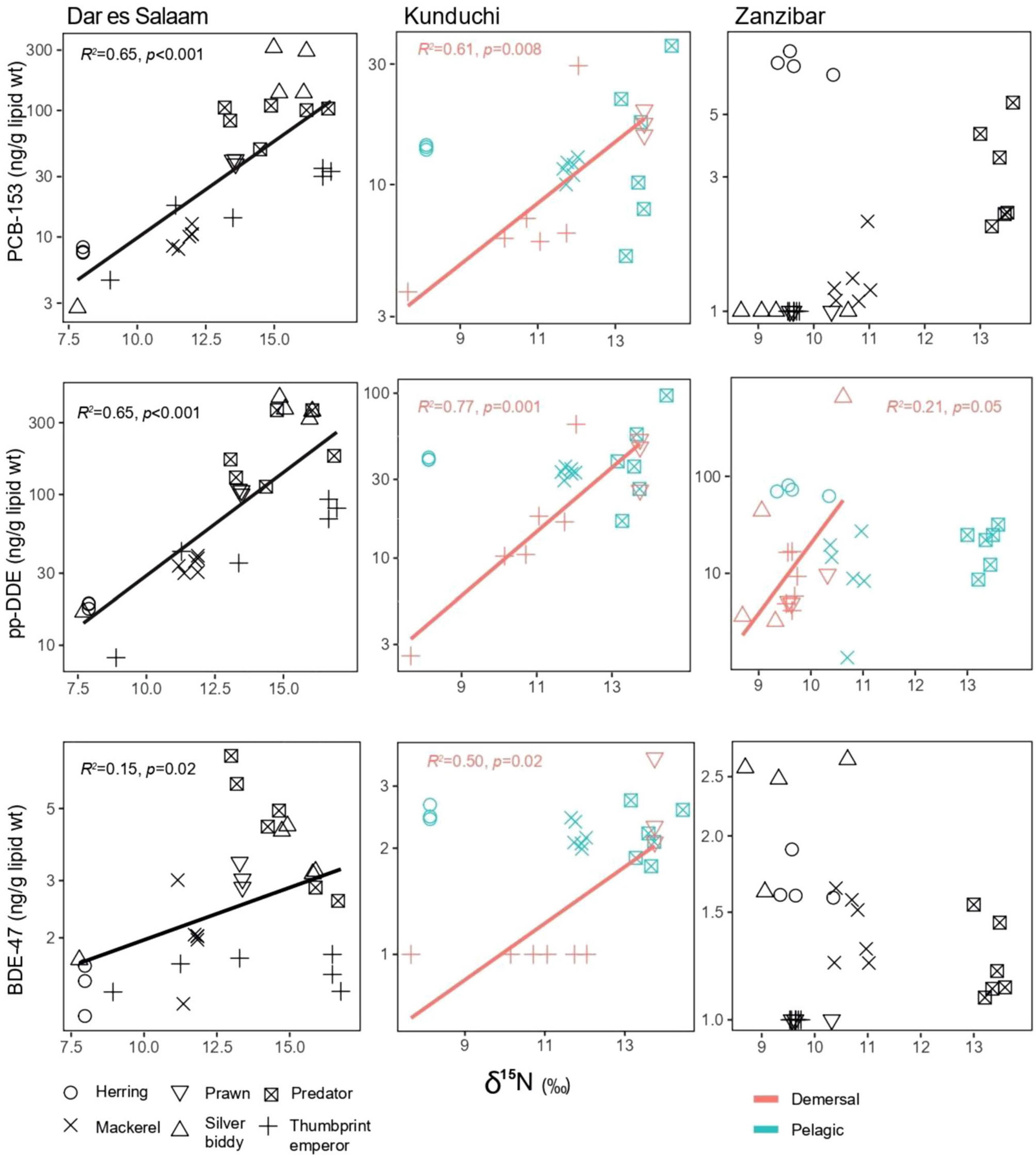

FIGURE 5: Associations between contaminants (PCB-153, pp-DDE, and BDE-47) and $\delta^{15}N$ in marine fishes and prawns collected from fish markets at the Dar es Salaam harbor, Kunduchi, and Zanzibar. Red and blue indicate significant interaction between habitat (pelagic/demersal) and $\delta^{15}N$, while black indicates no significant interaction. Regression line is shown only when the relationship between contaminant and $\delta^{15}N$ was significant. PCB = polychlorinated biphenyl; DDE = dichlorodiphenyldichloroethylene; BDE = brominated diphenyl ether.

Associations between $\delta^{15}N$, habitat, and contaminant concentrations

Linear regression analyses showed that fish size (weight and length) was not a significant explanatory variable for variation in POP concentrations at any location. In Kunduchi and Zanzibar, POP concentrations tended to be higher in organisms with higher lipid content, but this was not the case in fish from the Dar es Salaam harbor area. Contaminant concentrations are therefore presented on a lipid weight basis, while wet weight concentrations are shown in the Supporting Information.

The importance of δ^{15}N in explaining contaminant concentrations varied depending on contaminants and location and between habitats (pelagic/demersal; Figure 5). In Dar es Salaam, concentrations of PCB-153, PBDE-47, and *p, p*′-DDE were positively associated with δ^{15}N; and this association did not differ between habitats. In Kunduchi, a positive association between contaminants and δ^{15}N was found for the demersal system but not the pelagic system. On Zanzibar, a positive association between *p, p*′-DDE and δ^{15}N was found only for the demersal system, and no significant associations were found for the other contaminants. Differences in contaminant accumulation between organisms relying on demersal or pelagic carbon sources have previously been demonstrated in Lake Malawi (Kidd et al., 2001). A more efficient accumulation of ΣDDTs per trophic level was found for the pelagic system compared to the benthic system, possibly due to higher carbon turnover rates at the base of the benthic food web resulting in reduced ΣDDT exposure to consumers (Kidd et al., 2001). Biomagnification was not found in a food web from the subtropical region of the Pearl River delta in China, with no significant relationships between δ^{15}N and contaminants (Guo et al., 2008). Results from the present study could indicate a higher biomagnification potential in the demersal system compared to the pelagic, which could be explained by lipophilic contaminants having a higher affinity to sediments compared to water, and thus a higher exposure potential to organisms feeding in closer association to sediments. However, evaluation of accumulation of POPs in the Tanzanian coastal marine food web must be done with care because the food web was not fully characterized in the present study but biased by the selection of local, common fish species for human consumption. To better understand the trophodynamics of contaminants in this tropical marine system, the trophic niche of a wider range of species including primary consumers should be further investigated.

CONCLUSION

In the present study, occurrence of OCPs and industrial-use contaminants in marine fishes and prawns from coastal Tanzania was investigated among three locations differing in degree of anthropogenic activity. The collected species represented a wide range of different habitats and trophic levels, and contaminant concentrations among and within species varied accordingly. The contaminant concentrations in collected samples from the fish market in central Dar es Salaam were generally higher compared to those in a more rural fish market 25 km north of the city center (Kunduchi) and on Unguja Island of Zanzibar, which is as expected because of the higher anthropogenic activity in this fast-growing urban area. The results indicate spatial variation in concentrations of ΣPCBs, ΣDDT, and HCB with a mainland–island trend. Concentrations of ΣPCBs and ΣDDT were higher and those of HCB lower at the two mainland locations compared to Zanzibar, which may reflect differences in local pollution sources, while ΣPBDE concentrations were similar among locations.

Species-specific spatial variation in δ^{15}N values reflects individual feeding behavior or differences in baseline δ^{15}N values among locations. In general, contaminant concentrations were positively associated with δ^{15}N values, indicating higher contaminant exposure for organisms feeding at higher trophic levels.

Because of the relatively low levels of contaminants found in the present study, consumption of fishes and prawns from coastal Tanzania and Zanzibar does not represent a major risk for humans. However, environmental monitoring of PCBs, PBDEs, and other flame retardants should be conducted because the occurrence of industrial-use contaminants is assumed to increase in this region.

Supporting Information—The Supporting Information is available on the Wiley Online Library at https://doi.org/10.1002/etc.5254.

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Data Availability Statement—Data, associated metadata, and calculation tools are available from the corresponding author (katrine.borga@ibv.uio.no).

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