Occurrence and dynamics of organic contaminants in Tanzanian biota: legacies of the past and emerging concerns

Dissertation for the degree of Philosophiae Doctor

By

Ane Haarr

Department of Biosciences

Faculty of Mathematics and Natural Sciences

University of Oslo

2023

Series of dissertations submitted to the
Faculty of Mathematics and Natural Sciences, University of Oslo
No. 2693

ISSN 1501-7710

Cover: UiO.
Print production: Graphic center, University of Oslo.

Acknowledgements

The work presented in this thesis was carried out under the Anthrotox project, funded through the Life:Science initiative, Department of Biosciences and Department of Social Anthropology, University of Oslo (UiO). Thanks to the project leaders, Paul Wenzel Geissler (Dept. of Social Anthropology, UiO) and Katrine Borgå (Dept. of Biosciences, UiO) and the other affiliates: Knut Breivik (Dept. of Chemistry and NILU – Norsk Institutt for Luftforskning), Susanne Bauer (Center for Technology, Innovation and Culture, UiO), Ruth Prince (Institute of Health and Society, UiO), Rolf David Vogt (Dept. of Chemistry, UiO), and Anders Ruus (NIVA – Norsk Institutt for Vannforskning, Dept. of Biosciences, UiO).

I would like to direct my gratitute to our collaborating partners in Tanzania, who's help, knowledge and expertise have been invaluable: Aviti John Mmochi (IMS - Institute of Marine Science, University of Dar es Salaam, Tanzania), Peter Mangesho (NIMR - National Institute for Medical Research, Tanzania) and Eliezer Brown Mwakalapa (Department of Natural Sciences, Mbeya University of Science and Technology). Without you, Eliezer, much of the fieldwork would not have been possible. I am grateful for the time we spent at busy fish markets, in muddy mangroves, the crazy Dar es Salaam traffic, more or less dramatic boat trips, various waste disposal sites, and the hours we spent dissecting fish.

Thanks to Zhanna Tairova and Martin Mørch Larsen (University of Aarhus, Denmark) I was able to join fieldwork on Zanzibar and collect samples at an early stage of my PhD. Thank you, Zhanna, for your positive attitude, competence and help, and for including me in your work with the students from the State University of Zanzibar. Thank you, Martin, for help and constructive feedback on my first manuscript.

Thanks to co-supervisor Jan Ludvig Lyche and everyone at the "Miljøtoks" lab at the Norwegian University of Life Science. Mahin Karimi, Mariska Hoorweg, and Gabrielle Haddad-Weiser – thank you for welcoming me, training me, and helping me with laboratory work. Anuschka Polder – thank you for your kind consideration, critical comments and for sharing your knowledge and enthusiasm for this field. I know that you are important for a lot of students, and for me as well.

This journey would not have been the same without my fellow PhD students: Maja Nipen (Dept. of Chemistry), Franziska Klaas, Signe Mikkelsen, Samwel Moses Ntapanta, and

Christian Medaas (all from Dept. of Social Anthropology). I am grateful for having shared this experience, with all the excitement, the problems and frustrations, the laughs, and the ~~blood~~, sweat ~~and tears~~, with you. Being part of an interdisciplinary group like this has challenged me and opened my mind in ways of thinking and relating to my own field and the global challenges of environmental pollution.

I have spent many years roaming the halls in the fourth floor of the biology building here at UiO. There are many people I am grateful to have met here, and to still have in my life. Special thanks to my dear friends and colleagues at AKVA: Silje, Julie, Julia, Karoline, Lasse, Jan, Torben, Luka, Clare, Eve, Sabrina, Svenja, Khuong, Sagnik, Danny, Jessica, and Lina, for making these last years especially pleasant and including of things equally important as working on a PhD, i.e. beer, coffee, lunch, dinners, talks, rants, gossip, laughs, and even activities outside the university walls (!) Thanks Silje and Danny for reading and commenting on the final versions of my thesis. And thank you, Anders, for reading, commenting, and being there for me at the very end.

To Katrine: working with you has brought me to so many interesting places, from the high Arctic to the Tropics. Your research group and activities has grown immensely during the years since I started as a master student, and even though you are extremely busy you always seem to find time for your students. Your supervision has given me the right amount of freedom and responsibility, and I am very grateful for this. Thanks for including me and for giving me all these opportunities – It has been a lot of fun, and you are an important reason why I have stayed at UiO for so long.

To my parents – thank you for your unconditional support and trust in my choices.

❤

And to my dearest Jone, Ella and Felix

Table of contents

Summary

Data on the occurrence and dynamics of persistent organic contaminants (POPs) in tropical regions is scarce. While research and monitoring efforts largely have been carried out in the Global North (e.g. the Arctic), the Global South is greatly understudied. In order to meet the demands of a modern world, new contaminants are synthesized and legacy contaminants find new ways of entering the environment. In particular, the active transport of contaminants embedded in modern consumer products (e.g. electronics) from industrialized countries to developing countries, raises concern. A large amount of consumer goods and waste end up in dumspites in developing countries that lack the capacity for safe handling and recycling of waste. Thus, contaminants may leach into the environment, contaminating the air, soil and water bodies, and eventually enter the food chain where they can excert toxic effects in wildlife and humans. Waste from electronic products (e-waste) is particularly problematic due to their increasing demand, high consumer turnover, and contaminants contained within, including polychlorinated biphenyls (PCBs), polybrominated diphenyl ethers (PBDEs), chlorinated paraffins (CPs) and dechloranes.

The aim of this thesis is to understand whether e-waste represents a potential contaminant source to the Tanzanian environment, and increase the understanding of the occurrence and dynamics of both legacy and emerging organic contaminants in tropical biota from various habitats. The sampled material include marine fish from locations differing in degree of human activity, from Dar es Salaam to more rural locations on mainland Tanzania and on Zanzibar. On Zanzibar, marine fish were collected during two seasons in order to assess the effect of terrestrial runoff and/or atmospheric deposition on the dynamics of legacy contaminants such as PCBs, PBDEs and organochlorine pesticides (OCPs). Free-range chicken eggs and soil were collected around dumpsites on Zanzibar and in Dar es Salaam, including one e-waste location. Eggs and soil were analyzed for CPs and dechloranes and sources of these emerging contaminants to the Tanzanian environment were assessed.

As a fast-growing, urban and industrial center of Tanzania, Dar es Salaam is a source of contaminants to the Tanzanian environment. This was reflected in higher contaminant concentrations in marine fish collected around the central harbor fish market compared to a more rural location outside Dar es Salaam and on Zanzibar. Occurrence of BDE-209 and hexachlorobenzene (HCB) in marine fish from Zanzibar could suggest local point sources such as burning of waste. Seasonal variation in contaminant concentrations and patterns was

found in marine fish from Zanzibar. Following a period with heavy rain, concentrations of lower halogenated, more mobile PCB and PBDE congeners increased. Changes in composition patterns of dichlorodiphenyltrichloroethane (DDT) after the rainy season were also found, suggesting remobilization of terrestrial pools of non-degraded DDT and its metabolites into the marine environment. These findings point towards an effect of terrestrial runoff following a period with heavy rain. No clear conclusions could be made on e-waste as a source of emerging contaminants to the Tanzanian environment based on concentrations of CPs and dechloranes in chicken eggs and soil. Rather, these two chemical groups seem to be ubiquitous in the environment, even in a region with no production or extensive use. At one location, concentrations of short chain chlorinated paraffins (SCCPs) in two eggs were close to and exceeding the safety limit set by the EU, suggesting health concern from SCCP intake via egg consumption.

Occurrence of legacy and emerging contaminants in biota from the Tanzanian environment along with recent reports of their increasing trends call for further research and monitoring in this region. The material sampled and analyzed in the present study has enabled a broad assessment of contaminants concentrations and patterns in biota, representing different trophic levels and habitats, as well as their seasonal and spatial variation, for which there was limited knowledge from tropical regions prior to this study.

Sammendrag

Det er lite informasjon om organiske miljøgifter i tropiske regioner, både om forekomst og hvordan disse stoffene oppfører seg i miljøet og i næringskjeden. Mens forsknings- og overvåkningsinnsatsen i stor grad har blitt gjennomført i det globale nord (for eksempel i Arktis), er det globale sør relativt lite studert. For å imøtekomme kravene i en moderne verden, blir nye typer miljøgifter syntetisert og gamle miljøgifter finner nye veier ut i miljøet. Et økende problem er den aktive transporten av miljøgifter tilsatt i moderne forbrukerprodukter (for eksempel elektronikk) fra industrialiserte land til utviklingsland. Store mengder forbruksvarer og avfall ender opp i avfallsdeponier i utviklingsland som mangler kapasitet for sikker håndtering og gjenvinning av avfall. Dermed kan miljøgifter lekke ut i miljøet, forurense luft, jord, vann og forårsake toksiske effekter hos dyr og mennesker. Avfall fra elektroniske produkter er spesielt problematiske på grunn av økende etterspørsel, kort levetid samt tilsatte miljøgifter, som inkluderer polyklorerte bifenyler (PCB), polybrominerte difenyl etere (PBDE), klorparaffiner (CP) og dekloraner.

Målet med denne avhandlingen er å forstå om elektronisk avfall representerer en potensiell kilde til forurensning i det Tanzanianske miljøet, og bidra til forståelsen av forekomst og dynamikk av både eldre og nye typer organiske miljøgifter i tropisk biota fra ulike habitater.

Det innsamlede prøvematerialet inkluderer marin fisk fra steder som varierer i grad av menneskelig aktivitet, fra Dar es Salaam til mer rurale steder på det Tanzanianske fastlandet og på Zanzibar. På Zanzibar ble marin fisk samlet inn i løpet av to sesonger for å vurdere effekten av terrestrisk avrenning og/eller atmosfærisk avsetning på dynamikken til internasjonalt regulerte miljøgifter som PCBer, PBDEer og organiske klorpesticider (OCPer). Egg fra frittgående høns og jord ble samlet inn rundt søppelfyllinger på Zanzibar og i Dar es Salaam, inkludert en lokasjon med elektronisk avfall. Egg og jord ble analysert for klorparaffiner (CPs) og dekloraner, og vi undersøkte potensielle kilder til disse nye og uregulerte miljøgiftene i det Tanzanianske miljøet.

Som et raskt voksende, urbant og industrielt senter i Tanzania, er Dar es Salaam en kilde til forurensning i det Tanzanianske miljøet. Dette ble gjenspeilet i høyere miljøgiftkonsentrasjoner i marin fisk samlet inn rundt det sentrale fiskemarkedet sammenlignet med mer rurale lokasjoner utenfor Dar es Salaam og på Zanzibar. Forekomsten av BDE-209 og HCB i marin fisk fra Zanzibar kan tyde på lokale punktkilder som

forbrenning av avfall. Sesongvariasjon i miljøgiftkonsentrasjon og mønstre ble funnet i marin fisk fra Zanzibar. Etter en periode med kraftig regn økte konsentrasjoner av mindre halogenerte, mer mobile PCB- og PBDE-kongenere. Endringer i dichlorodiphenyltrichloroethan (DDT)- sammensetning etter regntiden ble også funnet, noe som antyder remobilisering av terrestriske kilder og avrenning til det marine miljøet. Ingen klare konklusjoner om elektronisk avfall som en kilde til miljøgifter i det Tanzanianske miljøet kunne trekkes basert på konsentrasjoner av CPer og dekloraner i hønseegg og jord. Disse kjemikaliene ser heller ut til å være til stede overalt i miljøet, selv i en region uten produksjon eller omfattende bruk. På en lokasjon var konsentrasjonene av kortkjedet-klorparaffiner (SCCPer) i to egg nær og under sikkerhetsgrensen satt av EU, noe som antyder risiko knyttet til eksponering av SCCP via inntak av egg.

Forekomsten av eldre og nyere typer miljøgifter i biota fra det Tanzanianske miljøet, sammen med nylige rapporter om økende trender, krever mer forskning og overvåking i denne regionen. Det innsamlede og analyserte materialet presentert i denne avhandlingen har muliggjort en bred vurdering av miljøgiftkonsentrasjoner og mønstre i biota som representerer ulike trofiske nivåer og habitater, samt deres sesongmessige og romlige variasjon, noe det var begrenset kunnskap om fra tropiske regioner.

Abbreviations

aBFRs	Alternative brominated flame retardants
BAF	Bioaccumulation factor
BCF	Bioconcentration factor
BFRs	Brominated flame retardants
CPs	Chlorinated paraffins
DDD	Dichlorodiphenyldichloroethane
DDE	Dichlorodiphenyldichloroethylene
DDT	Dichlorodiphenyltrichloroethane
Dec 602	Dechlorane 602
Dec 603	Dechlorane 603
DP	Dechlorane Plus
E-waste	Electronic waste
EFSA	European Food Safety Authority
EQS	Environmental Quality Standard
FAO	Food and Agriculture Organization of the United Nations
GC/Q-TOF	Gas chromatography quadrupole time-of-flight high-resolution mass spectrometry
GC-HRMS	Gas chromatography high-resolution mass spectrometry
HBCDD	Hexabromocyclododecane
HCB	Hexachlorobenzene
HPLC	High performance liquid chromatography
HCH	Hexachlorocyclohexane
IMS	Institute of Marine Science, Zanzibar
IPEN	International POPs Elimination Network
Kow	Octanol/water equilibrium partitioning coefficient
LC	Liquid chromatography
LCCPs	Long chain chlorinated paraffins
LOD	Limit of detection
LRT	Long-range transport
MCCPs	Medium chain chlorinated paraffins
MRL	Maximum residue limit
MONET	Monitoring Network
NILU	Norsk institutt for luftforskning / Norwegian Institute for Air Research
NMBU	Norges miljø- og biovitenskapelige universitet / Norwegian University of Life Sciences
OCPs	Organochlorine pesticides
PBT	Persistent, Bioaccumulative, Toxic
PBDEs	Polybrominated diphenyl ethers
PCBs	Polychlorinated biphenyls
POPs	Persistent organic pollutants
REACH	Registration, Evaluation, Authorization, and Registration of Chemicals
SCCPs	Short chain chlorinated paraffins
UNEP	United Nations Environment Program
WIO	Western Indian Ocean

List of papers

This thesis is based on the three papers listed below:

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

Haarr A., Mwakalapa E.B., Mmochi A.J., Lyche J.L., Ruus A., Othman H., Larsen M.M., Borgå K.
Science of the Total Environment 774 (2021) 145652

Paper II Spatial variation in contaminant occurrence in marine fishes and prawns from coastal Tanzania

Haarr A., Mwakalapa E.B., Lyche J.L., Mmochi A.J., Polder A., Ruus A., Borgå K.
Environmental Toxicology and Chemistry 41 (2022) 321-333

Paper III Chlorinated paraffins and dechloranes in free-range chicken eggs and soil around waste disposal sites in Tanzania

Haarr A., Nipen M., Mwakalapa E.B., Borgen A.R., Mmochi A.J., Borgå K.
Chemosphere 329 (2023) 138646

1. Background

1.1 The global challenge of environmental pollution

Environmental pollution is a global challenge, as contaminants can disperse over vast distances and reach areas far from their initial production and use. Since the second World War, organic contaminants as industrial and agricultural chemicals have been synthesised and produced in large quanities, and used mostly in industrialized regions of the world (Breivik et al., 2002; Abbasi et al., 2019; Melymuk et al., 2022). The physicochemical properties that make chemicals advantageous for use in industry and agriculture are the same reasons that make them problematic for the environment. These properties include long half-lives and persistence, the ability to bioaccumulate in humans and wildlife, and ability to exert toxic effects, summarized as Persistent, Bioaccumulative and Toxic (PBT) properties. As we have learned about their problematic properties, these chemicals have been characterized as persistent organic pollutants (POPs).

POPs have low water solubility but high solubility in organic phases meaning they readily accumulate in lipid-rich tissues of living organisms, i.e. are lipid soluble. POPs can biomagnify in food webs, reaching high concentrations in top predators, especially in long, marine food webs (AMAP, 1998; Borgå et al., 2004). At the top of the food web, humans are also at risk of accumulating POPs. Intake of animal fats, including dairy products, meat, fish and eggs, are essential sources of nutrients for humans but also an important pathway of contaminant exposure (Fries, 1995; Djien Liem et al., 2000; Bocio et al., 2003). As POPs are semi-volatile and mobile, many can undergo long-range transport (LRT) and be transported to remote regions of the world, such as the Poles, by repeated evaporation and condensation known as the "grasshopper effect". Contaminants can also be transported passively via oceanic and atmospheric currents (Wania & Mackay, 1993), via migrating species, and of increasing societal interest, via trade of products and consumer goods (Wong et al., 2007; Breivik et al., 2015). This active transport of contaminants typically happens along international gradients of economic inequality and regulatory weaknesses, from industrialized countries in the Global North, to developing countries in the Global South (Figure 1). The high costs associated with the environmentally sound recycling practices of modern consumer goods and their increasing demand in the Global South, are also facilitating this form of active transport (Breivik et al., 2011; Breivik et al., 2015).

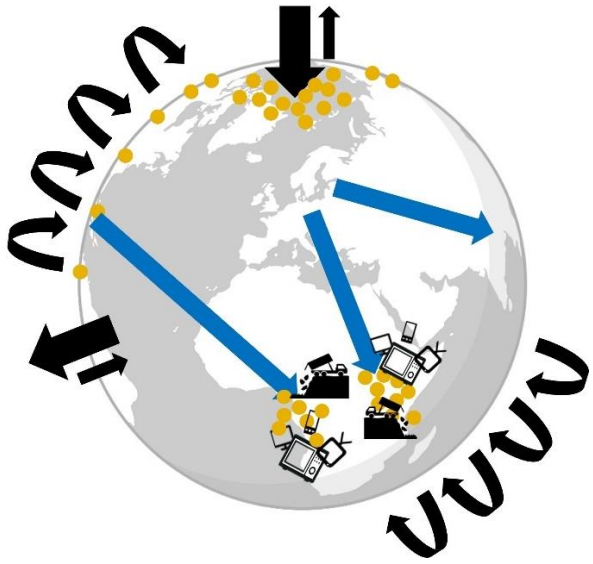


Figure 1. *Illustration of the passive, atmospheric transport (black arrows) of contaminants (yellow dots) and the active transport of contaminants (blue arrows). Passive transport of contaminants typically happens from warmer regions to colder regions due to evaporation and cold condensation, while active transport of contaminants happens via trade of products (e.g. e-waste), in which contaminants are embedded as chemical additives. Illustration inspired and modified from Breivik et al. (2015)*

Waste disposal sites in developing countries represent significant contaminant sources due to lack of sufficient infrastructure and regulations that enable the safe recycling and management practices of various waste types such as waste from electronic products (e-waste), medical- and industrial waste, and other types of hazardous waste. E-waste is considered particularly problematic due to an increasing consumer demand for modern electronic products, trade and use around the globe, as well as their short lifespan (Wong et al., 2007; Perkins et al., 2014; Forti et al., 2020). E-waste may contain a range of contaminants including heavy metals and organic compounds such as polychlorinated biphenyls (PCBs), polybrominated diphenyl ethers (PBDEs), and other halogenated flame retardants, such as chlorinated paraffins (CPs) and dechloranes. The incomplete combustion of e-waste, as part of disposal or crude recycling, can also produce and release toxic by-products such as dioxins and furans (Wong et al., 2007; Yu et al., 2010; Zeng et al., 2018; Ssebugere et al., 2019; UNEP/GMP3, 2021). Contaminants embedded in discarded products can leach into the air, soil, and surrounding water bodies, exposing wildlife, domestic animals and humans, meaning these waste disposal sites are contamination "hot-spots" (Zhao et al., 2009; Arukwe et al., 2012; Petrlik et al., 2022).

Both regional and international governing bodies regulate POPs for the protection of human and environmental health. The Stockholm Convention is administered under the United Nations Environmental Program (UNEP) and was ratified in 2004 by 131 countries, which agreed to work towards the elimination of the world's most problematic chemicals (UNEP, 2001). Regulation through the Stockholm Convention is based on a compound's PBT properties and potential for LRT. Persistence is determined by resistance to degradation, often

defined as a compound's half-life in a given environmental matrix, e.g. water or soil. The identification of bioaccumulative substances is often based on the chemicals' octanol-water partitioning coefficient (K_{ow}), bioconcentration factors (BCF) obtained from laboratory tests, or field-derived bioaccumulation factors (BAF) (Arnot & Gobas, 2006). Detection of a chemical in remote locations, such as the Arctic, is often an indication of its potential for LRT. Within the Stockholm Convention framework, the assessment of toxicity is based on a chemical's potential to exert adverse effects on human health or the environment. Production and import of chemicals (embedded in products or as pure substances) has to be registered within the European Chemical Agency (ECHA) according to the REACH (Registration, Evaluation, Authorisation of Chemicals) regulation. All manufacturers and importers of chemicals in quantities exceeding one tonne per year are thus responsible for assessing and managing the risks associated with the substances in question. The Rotterdam and Basel Conventions are additional international treaties under the UNEP framework working specifically to control international trade of contaminants (UNEP, 2004) and the transboundary movement of hazardous waste (UNEP, 2002), respectively.

A challenge for environmental pollution includes the complex task of managing the global stock of POPs produced over the years. For example, over 80% of PCB-containing materials have still not been eliminated despite regulations being in place since the 1970s and its listing under the Stockholm Convention since 2004. Some countries, such as Canada and the Czech Republic, have succeeded in reducing their PCB stocks by 99% the last 10 years (Melymuk et al., 2022). However, over 10 million tons of PCB-containing materials remain; mainly in countries lacking the ability to manage PCB-containing waste safely, but also in e.g. the USA which has not ratified the Stockholm Convention (Melymuk et al., 2022). PCB concentrations in biota remain high, especially in long-lived marine mammals feeding at high trophic levels. The high PCB body burdens in these mammals are still associated with adverse effects including reproductive impairment, disruption of endocrine and immune functions, cancers, and possibly animal population collapse, as hypothesized for the killer whale (*Orcinus orca*) (Desforges et al., 2018).

Another example is the large stock of organochlorine pesticides (OCPs) in African countries. Large stocks of pesticides, including dichlorodiphenyltrichloroethane (DDT), were recieved as donations from industrialized countries, often after the substances in question had been banned in parts of the global North (Manyilizu, 2019). Stockpiles of these pesticides were inadequately stored and needed safe disposal and destruction. This initiated the "Africa

Stockpiles Programme," aiming to clear all obsolete pesticide stocks from Africa and prevent further build-up and recurrence of OCPs (WorldBank, 2016). Pesticide storage under inadequate conditions represents a significant source of environmental pollution in many African countries, including Tanzania (Elfvendahl et al., 2004; Elibariki & Maguta, 2017).

1.2 Shift in contaminant source regions – from "cradle" to "grave"

The research efforts on environmental contamination have historically been directed to industrialized regions in the Global North, meaning less industrialized and developing regions in the Global South are greatly understudied. Most knowledge on the potential for LRT and the PBT properties of POPs comes from research conducted in the Global North, particularly in the Arctic (de Wit et al., 2010; Letcher et al., 2010; AMAP, 2017; Wong et al., 2021). Decades of monitoring POPs in Arctic air show declining trends of most POPs since their regulation (Wong et al., 2021), but this is typically not the trend in developing regions in the Global South. This has been attributed to increased import of waste and its improper handling in developing regions, e.g. crude recycling of electronic waste and shipwreck dismantling (Gioia et al., 2014; White et al., 2020; de Boer et al., 2023). The reduction in atmospheric levels of POPs in the Northern hemisphere has also been hypothesized to be partly achieved by the transport of waste (e.g. e-waste) from the Global North to the Global South (Breivik et al., 2011; Gioia et al., 2011; Breivik et al., 2014; Breivik et al., 2015). A shift in contaminant inventory and source regions is thus expected, from source regions where contaminants have been produced and used, to source regions in the receiving end of used goods and waste (Breivik et al., 2011; Li et al., 2016; Abbasi et al., 2019; Babayemi et al., 2022).

An increasing proportion of contaminants from consumer products are estimated to end up in dumpsites in developing countries after reductions in production and use in developed countries (Abbasi et al., 2019). For example, estimations show that the global BDE-209 stock will shift from the Global North to the Global South after its listing in the Stockholm Convention (2017), and that 5 kt of BDE-209 will still be in use by 2050, mainly in less industrialized countries (Abbasi et al., 2019). Pharmaceuticals, many of which are considered chemicals of emerging concern, were also found in highest concentrations in rivers in less industrialized regions compared to to industrialized regions. This finding was attributed to poor wastewater treatment and waste handling systems in low- to middle income countries (Wilkinson et al., 2022).

The use of sediment cores can reveal time-trends in contaminant deposition, and investigating contaminant occurrence in dated sediment layers can give insights into the historical pollution in the sampling area. Studies using sediment cores also indicate a shift in contaminant source regions by showing regional differences in peak contaminant concentrations in dated sediment layers. A general trend is that contaminant concentrations in countries that have been industrialized for some time peak around the time when production was highest (typically late 1990s), while in developing and newly industrialized countries contaminant concentrations peak in the top sediment layers representing the most recent years (Iozza et al., 2008; Arp et al., 2011; Nipen et al., 2022b). For example, sediment cores from a Mexican lake show an increase in PCB and PBDE concentrations during the recent years, suggesting that despite their restrictions and bans decades ago, these POPs are still being released to the environment (Ontiveros-Cuadras et al., 2019). These trends are attributed to release of POPs to the environment from products and waste causing delayed emissions in developing and newly industrialized countries compared to industrialized countries.

1.3 Organohalogen contaminants

The contaminants that were studied in this thesis include PCBs, OCPs, CPs, dechloranes, and brominated flame retardants (BFRs), including PBDEs and alternative BFRs (aBFRs). aBFRs is herein used to describe non-PBDE BFRs. A list of all organic contaminants analyzed in the present study along with their start of production, regulation and listing in the Stockholm Convention is given in Table A1. The selected contaminants represent chemicals with different production volumes, usages, sources and regulations. PCBs, BFRs, CPs, and dechloranes are herein referred to as industrial-use contaminants because they are deliberately produced and used for various purposes in industrial and consumer products. OCPs are designed and produced to be toxic to control weeds, insect infestations and diseases in agriculture, but may also be toxic to other organisms, including humans (AMAP, 1998).

PCB production occurred mainly in Europe, USA, Russia and Japan, and peaked in the 1970s before being phased out in the 1990s. PCBs were used in hydraulic fluids, heat transfer fluids, lubricants, and as plasticizers (Kimbrough & Jensen, 1989; Breivik et al., 2002). Production of PBDEs started in the 1970s in China, Japan, Europe, USA, and Israel, and peaked around the year 2000. PBDEs were mainly used as flame retardants in electronics, furniture, textiles, construction materials and vehicles. Penta- and Octa-BDE mixtures were listed under the Stockholm Convention in 2009, while Deca-BDE was the last PBDE mixture to be banned through the Stockholm Convention in 2017 (Abbasi et al., 2019). aBFRs have been

introduced to the market to replace restricted or highly regulated contaminants, and often share structural and functional similarities with the legacy PBDEs (Covaci et al., 2011).

Dechloranes and CPs are high-production chlorinated flame retardants and are currently applied in modern consumer products such as plastics and electronics. Today, China is the biggest manufacturer of CPs and the annual production is estimated to exceed 1 million tons, which is more than the total production volume for PCBs (van Mourik et al., 2016). CPs are mainly used as additives in metal working fluids, in paints, sealing materials, and in the industrial production of rubber, leather and other textiles (Wei et al., 2016). Based on their carbon chain length, CPs can be classified into four groups: very short-chain CPs ($C_{<10}$, VSCCPs), short-chain CPs ($C_{10\text{-}13}$, SCCPs), medium-chain CPs ($C_{14\text{-}17}$, MCCPs), and long-chain CPs ($C_{>17}$, LCCPs). Short-chain CPs have recently been subjected to regulation through the Stockholm Convention (UNEP, 2017), while MCCPs are currently under consideration. Dechlorane Plus (DP) was first manufactured in the USA and later in China (Sverko et al., 2011). DP is mainly used as a flame retardant in electrical wires and cable coatings, computer connectors and plastic materials. Dechlorane 602 and 603 are often detected together with DP in environmental samples, and studies suggest worldwide occurrence of these DP related compounds. However, details on their production and usage are not readily available (Wang et al., 2016).

POPs under international regulation, such as PCBs, PBDEs and several OCPs, including ∑DDT (DDT, DDE and DDD) and hexachlorobenzene (HCB), are often referred to as legacy contaminants, meaning that their presence in the environment is largely a legacy of historical production and use. Contaminants of emerging concern include newer and unregulated chemicals whose production, use, and release in the environment are increasing, such as dechloranes and CPs. However, new releases of legacy POPs to the environment may still occur from stockpiles and waste (De Wit et al., 2019), and from permafrost, sea ice, glaciers and soils, whose capacitiy to store contaminants are affected by changing climatic conditions, e.g. higher temperatures (Ma et al., 2016) and increased precipitation (Ruus et al., 2010). Many legacy POPs are also by-products formed during combustion processes, such as HCB, or are only partly regulated and allowed for use in certain cases, such as indoor spraying of DDT for malaria control (De Wit et al., 2019). Therefore, even though legacy POPs are subjected to regulations, these contaminants also represent an emerging concern, especially in countries where the capacity for safe handling of waste is lacking (Breivik et al., 2011; Breivik et al., 2015; Melymuk et al., 2022; Wilkinson et al., 2022).

1.3.1 Human risk assessment of chlorinated paraffins

Human risk assessment involves hazard identification, determination of the relationship between exposure and an adverse effect (dose-response assessment), assessing the level of exposure (e.g. through intake data) and risk characterization (EFSA, 2023). A toxicity test, performed under controlled laboratory settings, can provide useful data for risk assessment, including a dose-response curve. A dose-response curve provides information on the no-observed adverse effect level (NOAEL) and the lowest-observed acverse effect level (LOAEL). The benchmark dose level (BMDL) describes the dose or concentration for when a specified health risk is induced and can be statistically modelled using toxicity studies, taking into account the uncertainties in the dose-response data. Based on available data from toxicity testing, expert panels, like the European Food Safety Association (EFSA), extrapolate results and determine tolerable levels for contaminants in food. Other region-specific guidelines exist, e.g. American (USEPA) and Chinese guidelines, but there are no regulations specifically for Africa.

Risk assessment of CPs in food was recently conducted by the EFSA CONTAM panel. Based on the available scientific literature, the panel selected BMDLs for SCCP and MCCP exposure. Further, a Margin of Exposure (MOE) approach was used to assess potential health concern from CP intake via food. The MOE is the ratio of the BMDL and estimated exposure dose. For CPs in food, the panel selected a MOE above 1000 to indicate no health concern and MOE below 1000 to indicate health concern (EFSA, 2020).

1.4 Environmental contamination in Tanzania

Most African countries have ratified the Stockholm Convention. However, the information on POPs in Africa is scarce and does not allow proper evaluation of temporal trends and population exposures levels (Bruce-Vanderpuije et al., 2019; UNEP/GMP3, 2021). Tanzania ratified the Stockholm Convention in 2004, but many of the challenges faced here are representative of most of Sub-Saharan Africa: lack of monitoring studies and historical data, lack of capacity to analyse organic contaminants in environmental samples, lack of capacity to safely manage hazardous waste, weak enforcement of legislation, and low public awareness on the issues of pollution (Leslie et al., 2013; White et al., 2020; UNEP/GMP3, 2021).

Sources of POPs to the Tanzanian environment include runoff from agriculture and urban areas, wastewater discharge from sewage and industrial activities, shipwreck dismantling, and improper waste management (Machiwa, 2010; Magashi & Schluep, 2011; Yhdego, 2017; de

Boer et al., 2023). The aquaculture industry in Tanzania is growing (MALF, 2016), and the country is in the initial phases of oil and gas exploration, which may represent additional sources of future contamination. Tanzania is not an officially known recipient of e-waste from industrialized regions, as are other countries in West Africa. However, Tanzania is experiencing rapid economic growth with increased import and domestic use of new and used electrical goods (Ntapanta, 2022).

Elevated concentrations of chlorinated paraffins and dechloranes have been found in the air and soil around urban locations and near waste and e-waste handling facilities around Dar es Salaam in Tanzania (Nipen et al., 2022a). Sediment core sampling has also revealed a recent environmental increase of both legacy (PCBs, PBDEs) and emerging (aBFRs, CPs, Dechloranes) contaminants in this region (Nipen et al., 2022b). Environmental pollution from pesticides have been found in Tanzanian sediments, soil, water, vegetation and biota (Mwevura et al., 2002; Kishimba et al., 2004; Elibariki & Maguta, 2017). Müller et al. (2019) analysed POPs in Tanzanian mothers and infants, and found that BDE-47 concentrations in maternal blood were in the same range as in Californian mothers, a population cohort known to be highly exposed to PBDEs (Sagiv et al., 2015). The intake of non-dioxin-like PCBs (∑6PCBs) from breast milk in Tanzanian infants has also been estimated, and exceeded the provisional tolerable daily intake (PTDI) in more than 50% of the infants (Müller et al., 2017). At present, there are few studies on the occurrence of PCBs, PBDEs and other industrial-use contaminants in Tanzanian biota (Polder et al., 2014; Polder et al., 2016), and even fewer in the Tanzanian marine system (Mwakalapa et al., 2018).

Population growth, industrialization and urbanization in Tanzania may cause an increase in environmental pollution, especially if waste handling practices are not improved. This is particularly important for the major urban areas in coastal Tanzania, such as Dar es Salaam, where the economy is rapidly growing (Kruitwagen et al., 2008; Todd et al., 2019; Nipen et al., 2022a). These results call for further investigations of both legacy and emerging contaminants in Tanzania, their spatial distribution and accumulation in biota.

1.5 Bioaccumulation of contaminants in tropical systems

Our understanding on the factors affecting the environmental fate and dynamics of organic contaminants in food webs is largely based on studies on mid- to high-latitude systems. Limited information exists on the bioaccumulation and dynamics of contaminants in tropical systems (Alava & Gobas, 2012; Walters et al., 2016; Zhang et al., 2017). Arctic and

temperate food webs are adapted to high seasonal variation in irradiance and thus, primary production and food availability. This has an impact on the lipid dynamics for energy storage in organisms, which again influences the accumulation, distribution and metabolism of lipid soluble contaminants (Borgå et al., 2004). Tropical marine systems are rich in biodiversity, have complex food webs, higher microbial activity and metabolic efficiency compared to Arctic and temperate systems (Paine, 1966; Borgå et al., 2012; Brown, 2014; UNEP/GMP3, 2021). Tropical systems are not adapted to large variations in temperature or irradiation, but experience large seasonal changes in precipitation, which can affect environmental (re)distribution and bioavailability of contaminants. Heavy rainfall, which occurs seasonally in tropical regions, is associated with increased atmospheric deposition of contaminants and runoff from land (Fu & Wu, 2006; Shi et al., 2016; Lammel et al., 2018).

In tropical terrestrial environments, low content of organic matter in soil can limit the capacity to bind organic contaminants, and high temperatures facilitate evaporation and lower the environmental half-life of contaminants (Webster et al., 1998). However, recent studies have shown relatively high concentrations of both legacy (e.g. PCBs and PBDEs) and emerging (CPs and dechloranes) contaminants in air and soil measurements from Tanzania, especially around urban areas and waste disposal sites (Nipen et al., 2022a).

2. Aims and objectives

This PhD project is part of the convergence environment "Anthropology of Toxicity" (AnthroTox), which is an interdisciplinary project aiming to study various aspects of environmental pollution in Africa, with Tanzania as the main study site. The AnthroTox project has brought together students and researchers from social anthropology, environmental chemistry and ecotoxicology, from different parts of the world, engaging, communicating and cooperating on challenges regarding environmental pollution. The overarching aim of the AnthroTox project is to "understand how environmental and social processes and their relationships dictate flows and impacts of anthropogenic contaminants within and across societies and ecosystems". Anthropogenic activity, with increasing demand of modern consumer products, results in an increasingly efficient transport of contaminants and strongly influences their global distribution. As contaminants increase in terms of global concern, aspects of social differences and injustice become relevant as societies have different capacities to monitor contaminants and enforce international regulations and safety measurements, including waste handling. The topic of toxic waste is a highly complex issue as it touches upon factors of economic, social, ethical and political concern. Within the AnthroTox project, the main focus has been to study contaminants associated with industrial activities and consumer products, particularly electronic equipment and e-waste such as PCBs, BFRs, chlorinated paraffins and other types of halogenated flame retardants.

The aim of this thesis is to understand whether e-waste represents a potential contaminant source to the Tanzanian environment, and increase understanding of the occurrence and dynamics of both legacy and emerging organic contaminants in various tropical habitats and biota. To pursue this aim, the following objectives were addressed:

- To investigate contaminant concentrations and patterns in coastal marine fishes before and after the rainy season to assess potential effects of terrestrial runoff (**Paper I**)
- To investigate whether the degree of anthropogenic activity affects spatial variation in contaminant occurrence in marine fishes along the Tanzanian coast (**Paper II**)
- To investigate whether waste in general, and e-waste in particular, represent a source of emerging contaminants to soil and chicken eggs near waste disposal sites in Tanzania (**Paper III**)

3. Methods

3.1 Fieldwork

To address the aims and objectives of this study, fieldwork was carried out in Dar es Salaam on the Tanzanian mainland and on Unguja Island of Zanzibar in three rounds: January 2018, August 2018 and January/February 2019. January and February represent the dry and warm season. The main rainy season occurs between March and May, while August is usually associated with lower temperatures. The selection of sampling sites and study species, and the sample collection was conducted together with the AnthroTox-team (students and supervisors) and in close collaboration with local partners and experts. Local anglers, small-scale farmers, residents and landowners were very helpful with advising and assisting in sample collection. Sample size throughout was limited by the high costs associated with analyses of organic contaminants, which is not unusual for ecotoxicological studies of this kind. In general, six replicates per sample type were attempted to collect to make statistical inferences.

The selection of marine organisms for **Paper I** and **II** represent species that are fished for human consumption and thus of commercial interest. Fishes and prawn were bought from local anglers at three fish markets: “Malindi” fish market on Zanzibar, “Harbor” fish market in central Dar es Salaam, and “Kunduchi” fish market, 25 km North of Dar es Salaam city center (Figure 2). Through communication with the anglers, fish from the local, coastal environment was collected as far as possible. In **Paper III,** eggs from free-range chickens (*Gallus domesticus*) and soil samples were collected around three different waste disposal sites: “Maruhubi” on Zanzibar, which is an informal dumpsite (not regulated); “Mtoni” dumpsite in Dar es Salaam, which is an historical dumpsite no longer in use (closed in 2009); and the currently official dumpsite of Dar es Salaam, “Pugu Kinyamwezi.” The official dumpsite is located around 25 km South-East of the city center. Next to the official dumpsite lies a licensed e-waste dealer that buys electronic waste, dismantles, sorts and sends byproducts to other waste dealers. The owner reported that some material was sent to Belgium for recycling of valuable materials. The lack of an incineration oven made it impossible for the owner to properly recycle the material they received. Eggs were also collected at a presumed reference location: a residential area at Bumbwini, Makoba, located around 25km North of Zanzibar city and away from any known waste disposal sites.

Sample processing and subsample collection were conducted at the facilities of the Zanzibar State University (SUZA), the Institute of Marine Science (IMS) on Zanzibar, or outdoors at an adequate place. Established protocols for fish dissection were followed and quality assurance was obtained by use of e.g. clean aluminium foil and nitrile gloves for each fish, cleaning of equipment between samples, and proper storage of samples prior to analyses. Details of sampling and dissection procedures are described in the respective papers.

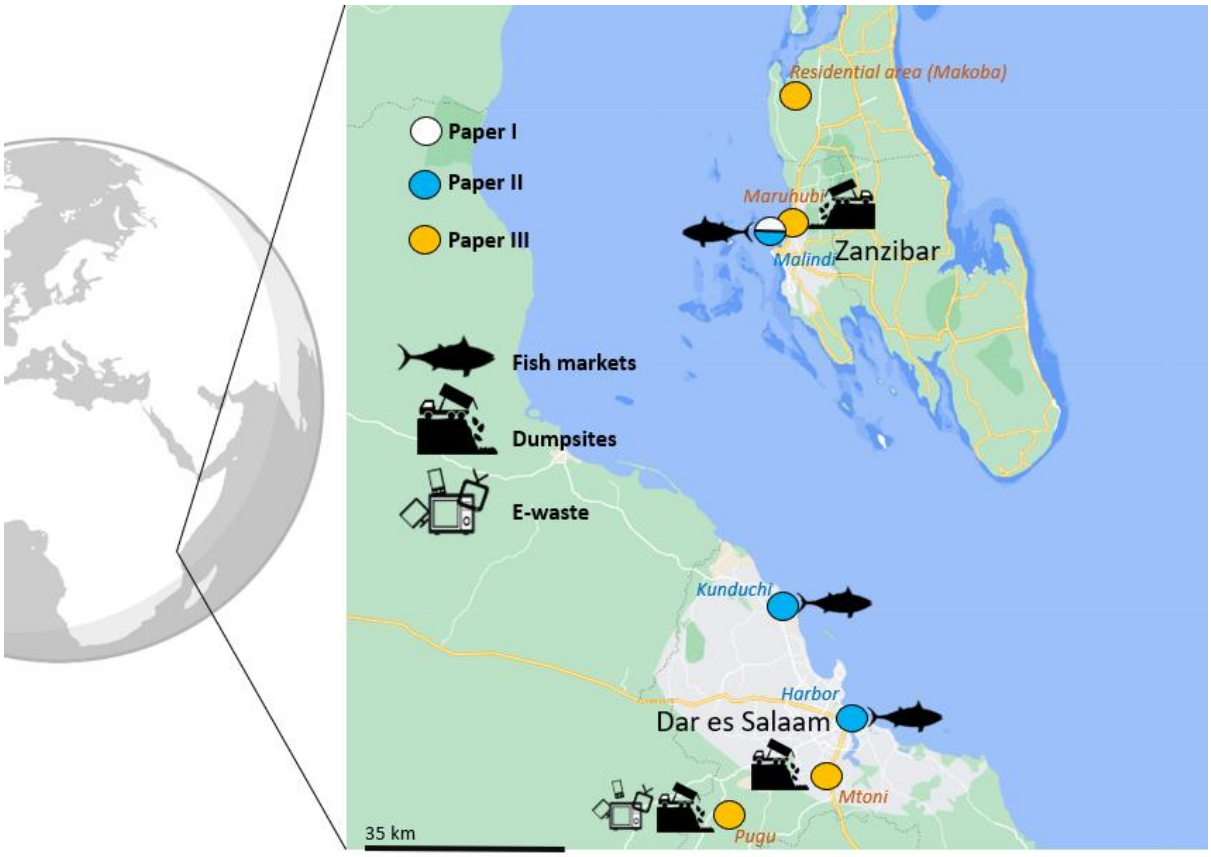


Figure 2. *Figure of sampling sites on the Tanzanian mainland and on Unguja Island, Zanzibar.*

3.2 Permissions and ethical clearance

Ethical clearance for the whole AnthroTox project was granted by the National Institute for Medical Research (NIMR), Tanzania. Permissions to conduct field work was granted by the Tanzanian Commission for Science and Technology (COSTECH), 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.

3.3 Analyses of organic contaminants

Sample preparation, extraction, clean-up, lipid quantification and instrument analyses of PCBs, BFRs and OCPs were conducted at the Norwegian University of Life Science (NMBU), Adamstuen, Norway. Methods are described in detail in Polder et al. (2014) and in **Paper I** and **II**. In short, homogenized samples of fish and shrimp (muscle tissue) was extracted using acetone and cyclohexane (2:3). Extracts were treated with sulfuric acid for lipid removal. Sample extracts were run on a high-resolution gas chromatograph (HRGC) coupled to a mass spectrometer (MS). The laboratory is accredited by "Norwegian Accreditation", which is the national accreditation body of Norway established by the Royal Norwegian Ministry of Trade and Fisheries in accordance with EU Regulation 765. Analyses of POPs in biological materials by the laboratory is conducted according to the requirements of the NS-EN SO/IEC 17025 (test 137). The laboratory follows strict protocols and routines for quality control measures by running parallel samples, procedural blanks, internal laboratory reference material, European certified reference material, and regular participation in international ring tests.

Analyses of the emerging CPs and Dechloranes in egg and soil samples were conducted at the Norwegian Institute for Air Research (NILU), Kjeller, Norway. Methods are described in **Paper III** and references therein. In short, egg and soil samples were extracted using cold column and accelerated solvent extraction, respectively, using acetone and n-hexane (1:1). Sulphuric acid was used for lipid removal. An extra clean-up step was conducted using activated silica and sodium sulphate packed in a column and diluted with diethyl ether in n-hexane before the solvent was changed to isooctane. Concentration of dechloranes and chlorinated paraffins was determined in the final extract using gas chromatography quadrupole time-of-flight high-resolution mass spectrometer (GC-QTOF). A pattern deconvolution method developed by Bogdal et al. (2015) was used for the quantification of CPs. Since CPs are particularly challenging to quantify, the laboratory follows strict quality control measures to ensure method performance, as well as participation in international laboratory certification studies.

3.4 Analysis of stable isotopes

Analyses of stable isotopes were conducted at the Stable Isotope Laboratory of the University of Oslo (UiO:CLIPT). Detailed description of the methods is given in **Paper I**. In short, dried and homogenized tissues were sealed in tin capsules and analysed for carbon ($\delta^{13}C$) and nitrogen ($\delta^{15}N$) isotopes using a Thermo Fisher Scientific EA IsoLink IRMS System with a

Flash Elemental Analyses and DeltaV Isotope Ratio Mass Spectrometer. Calibrated internal laboratory reference material and quality control material (from Fisher Scientific) were run with each sample batch to normalize the data and assess precision and accuracy to the measurements.

Dietary descriptions such as stable isotopes of carbon and nitrogen can be used to characterize an organism's dietary habits of carbon source and relative trophic position, respectively, integrated over time (Hobson & Welch, 1992). Stable isotopes are useful tools providing ecological linkages to contaminant occurrence and distribution in organisms and food webs (Kidd et al., 2001; Borgå et al., 2004). 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 reflects relative trophic status. $\delta^{13}C$ does not change along a trophic level gradient, but changes with different photosynthetic pathways in primary producers (e.g. C3 and C4 plants) (Minagawa & Wada, 1984; Layman et al., 2012). In marine systems, $\delta^{13}C$ can be used to assess gradients of pelagic/demersal and marine/terrestrial carbon sources.

3.5 Data treatment and statistical analyses

All datasets were established in Windows Excel and statistical analyses were conducted using RStudio. In general, assumptions of normal distribution and homogenous variance were not met and so non-parametric testing of group-differences were conducted throughout (or log-transformed for multivariate analyses). Lipid normalized contaminant concentrations were used throughout in order to compare concentrations among species in the present study and with data from the literature.

The use of multivariate statistics is well established in the field of ecotoxicology (Sparks et al., 1999). In ecotoxicology, we often measure multiple parameters in the same samples, and multivariate methods allows for investigations of occurrence and interrelations among responses, and how they can be explained by selected explanatory factors. The *Vegan* package in R was developed for gradient analyses in ecology, but is also useful dealing with other types of datasets that include several response variables (e.g. contaminants concentrations) and few explanatory variables (e.g. location, species). In this study, principal component analyses (PCA) were used to visualize and explore trends and patterns in the dataset (**Paper I and III**), and redundancy analyses (RDA) were conducted for significance testing of relationships between explanatory and response variables **(Paper I**).

4. Results and Discussion

In **Paper I** and **II**, we present and discuss concentrations and patterns of legacy contaminants in marine fishes and prawns. We demonstrate variations in concentrations of PCBs, BFRs and OCPs related to species-specific dietary niches, seasonal dynamics (**Paper I**) and spatial variation (**Paper II**). In **Paper III**, we present and discuss concentrations and composition patterns of two groups of emerging contaminants, CPs and dechloranes, in free-range chicken eggs and soil around Tanzanian dumpsites. **Paper III** is the first peer-reviewed paper to report concentrations of CPs and dechloranes in African biota and to perform risk assessment of human CP intake via egg consumption.

4.1 Legacy contaminants

In general, the levels of legacy, industrial-use contaminants (PCBs and PBDEs) were low in coastal marine fishes and prawns from Dar es Salaam and Zanzibar (**Paper I and II**). This finding is probably representative for the whole Western Indian Ocean (WIO) region. Due to distance from point sources and a history of low degree of industrialization in the region, the WIO is not considered to be very contaminated (Munschy et al., 2020; van der Schyff et al., 2021). The highest contaminant concentrations were found in fish from central Dar es Salaam, which indicates that urban and industrial areas are sources of POPs to the local marine environment (**Paper II**).

A high proportion of pesticide-use contaminants (OCPs) to total POP concentrations was expected due to the importance of agriculture and the historical and present use of pesticides in this region. OCP composition consisted of up to 94% ∑DDTs followed by HCB, HCH, chlordanes and mirex. ∑DDT patterns were largely dominated by *p,p'*-DDE, which is the main DDT metabolite and a highly persistent, bioaccumulative and toxic compound (Mirmigkou & de Boer, 2016). The ratio of the mother compound (DDT) to its metabolites (mainly DDE, but also DDD) is useful when understanding if DDT use has occurred in the past or recently. A high DDE/∑DDT ratio indicates that metabolites dominate the ΣDDT pattern and suggest historic use, while a low ratio means that the ΣDDT pattern is dominated by non-degraded DDT, and indicate recent use. Indications of recent DDT use in Tanzania are somewhat contrasting, as some studies report indications of recent use (Polder et al., 2014; Polder et al., 2016; Mwakalapa et al., 2018), while others do not (Müller et al., 2017; Mwakalapa et al., 2018; Mwevura et al., 2020). Policy for indoor residual spraying for malaria prevention exists in Tanzania and on Zanzibar, but not specifically for DDT use

(WHO, 2021). Results from **Paper I** and **II** indicate historical DDT use, as contaminant patterns were dominated by the main DDT metabolite, DDE.

In **Paper II**, we compile literature data to compare POP concentrations from fish in this study to what has been found in similar fish species from other areas of the world. Figure 3 shows concentrations of legacy POPs (ΣPCBs, ΣPBDEs, ΣDDTs, HCB) in different fish species from Tanzania, various African freshwater lakes, remote locations in the WIO such as the Seychelles, the South China Sea, and Europe. Notably, ΣPCB concentrations in fish from the WIO is low compared to fish sampled from more industrialized regions in China and Europe. The high PCB concentrations in silver biddy presented in **Paper II** are comparable to what is found in Atlantic cod (*Gadus morhua*) from Norway, but the mean concentration is largely affected by very high concentrations in one individual fish, which most likely represents an outlier rather than a general trend. Relatively high DDT levels in fish from Lake Tanganyika and Lake Malawi, which lie on the borders of western and southern inland Tanzania respectively, suggest that fishes from these lakes, receiving runoff from several bordering countries, are more exposed to DDT compared to marine fishes.

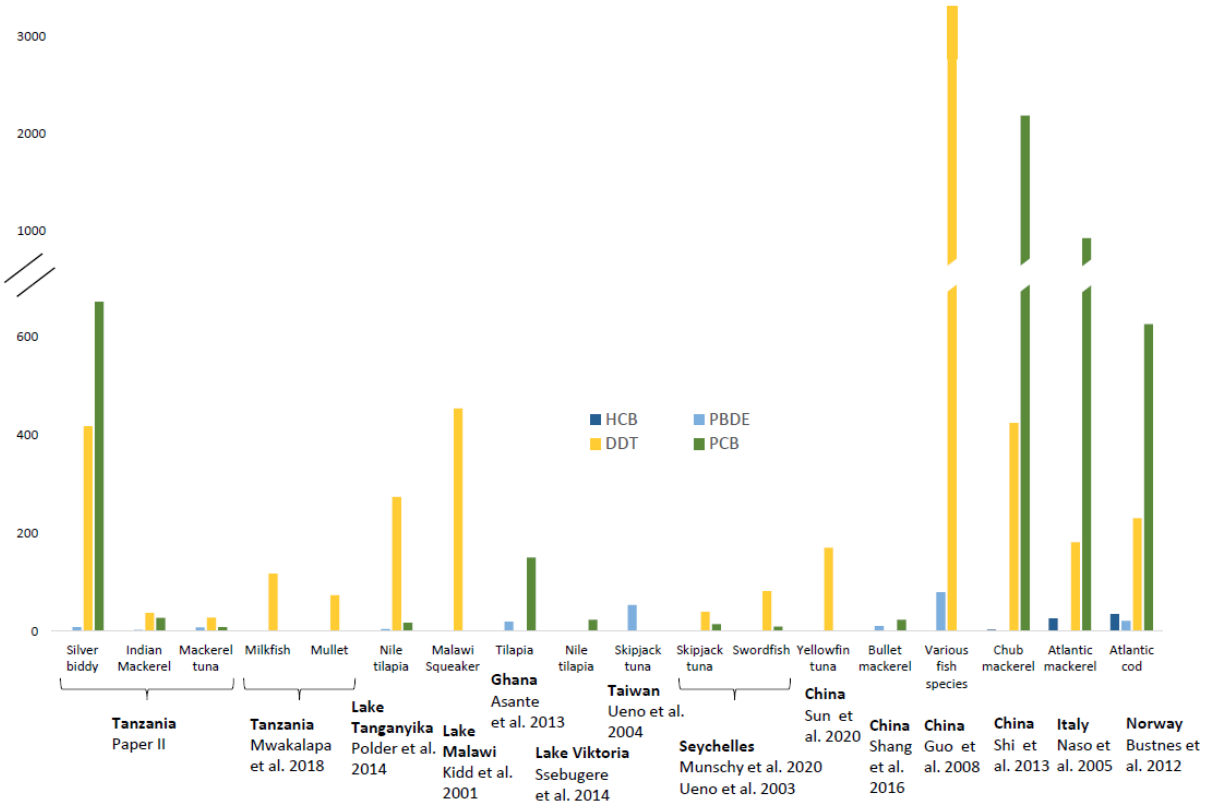


Figure 3. *Concentrations of PCBs, PBDEs, HCB and DDTs (ng/g lipid weight) in marine fishes from the present study and data collected from the literature. Note the axis break at the vertical axis and different scales above and below this break.*

In Paper II, we conduct a crude assessment of adequacy for human consumption by comparing levels of PCBs, PBDEs, HCB, HBCDD and DDT found in marine fish to European thresholds. Maximum Residue Limits (MRLs) describe the maximum allowed concentration of a chemical in food products (E.C. 2011). No samples exceeded the MRLs set for PCBs, HCB, HBCDD or DDTs. Environmental Quality Standards (EQS) are set by the water framework directive to protect human health and top predators from secondary poisoning through biomagnification (E.C. 2013). The EQS_{biota} set for PBDEs is conservative (0.0085 ng/g ww), and close to the detection limit for the analyzed PBDEs in the present study.

Even though PCBs and PBDEs are considered legacy contaminants, studies indicate that there is an environmental increase of both legacy and emerging industrial contaminants in this region (Gioia et al., 2011; Breivik et al., 2015; Nipen et al., 2022b). PBDE levels exceeding levels of PCBs in fish from Zanzibar, as discussed in **Paper I,** was a somewhat unexpected result, as PCBs most often dominate over PBDEs in the contaminant profile in environmental samples due to historical production and use (Breivik et al., 2002). This finding may be related to the difference in use between the two groups of industrial contaminants, where PBDEs are associated with more modern consumer products compared to PCBs, which were used in products produced before 1990s (Nipen et al., 2022b). Tanzania has stopped importing transformers containing PCB oils, but still has a substantial PCB stock associated with this type of electrical equipment (IPEN, 2005). Increasing PBDE concentrations in sediment from Dar es Salaam during recent years have been linked to a range of socioeconomic factors, including population growth, industrialization, and waste generation (Nipen et al., 2022b).

The detection of brominated flame retardants in fishes and prawns could indicate exposure from e-waste. For example, BDE-209 is the main constituent in the commercial deca-BDE mixture that was extensively used as a flame retardant in electronics and other consumer products before it was phased out of production in 2017 (de Wit et al., 2010). Normally, BDE-209 does not readily accumulate in fish because of its high molecular weight, high hydrophobicity and low bioavailability. It is also known to be efficiently metabolized and eliminated and therefore often not detected in fish (Viganò et al., 2011). Therefore, the detection of BDE-209 in fish could indicate recent exposure to the commercial deca-BDE mixture. Higher concentration and detection frequency of HCB and BDE-209 in fish from Zanzibar compared to mainland Tanzania (**Paper I** and **II**) could point towards a point source

on Unguja Island, such as the use of fire foam on the airfield or open burning of waste (Mwakalapa et al., 2018; Zhang et al., 2018). The aBFRs were only detected in very few marine species (**Paper I and II**). Due to low instrumental sensitivity and detection frequency, most aBFRs were omitted from data analyses. It could be hypothesized that the relatively recent phase-out of deca-BDE usage and the substitution by aBFRs has yet to reflect current contaminant composition in marine organisms in Tanzania. However, this should be further monitored as a shift from the legacy BDEs to emerging BFRs is expected also in the future (Covaci et al., 2011).

4.2 Seasonal dynamics and bioaccumulation of contaminants

In **Paper I**, we document a general increase in POP concentrations in fishes and prawns after the rainy season, as well as a shift in contaminant pattern to a higher relative occurrence of lighter, more mobile PCB and PBDE congeners. These findings suggest that there may be atmospheric deposition and/or terrestrial runoff of these congeners into the marine environment. Differences in the composition patterns of ∑DDT between January and August also indicates a signal of terrestrial runoff. We found a higher contribution of DDT and DDD to total ΣDDTs in samples from August compared to January, suggesting that terrestrial pools of DDT and DDD are remobilized and entering the marine environment following a period with heavy rain. The two months with most precipitation on Unguja Island in 2018 was April and May. Although the following months had relatively little precipitation, there was likely still a signal from increased rainfall during sampling in August. More data over time is needed in order to better understand differences in contaminant concentrations and patterns driven by the seasonal rain.

Results from **Paper I** and **II** indicate that trophic level ($\delta^{15}N$) is the most important explanatory factor for contamiant concentrations in marine fishes and prawns from Tanzania. The biomagnification potential, i.e. the increase in contaminant concentrations with increasing trophic level, varied among locations and between habitats, but appeared to be the greatest in Dar es Salaam and in the demersal system at Kunduchi. Higher biomagnification in the demersal system may be due to a higher affinity of organic contaminants to sediment and that organisms that feed closer to sediment are more easily exposed. Spatial variation in $\delta^{15}N$ was observed, as fish from Dar es Salaam generally contained a higher $\delta^{15}N$ signal compared to the same species from the other locations. For example, two demersal fish species (thumbprint emperor and silver biddy) sampled at the central fish market in Dar es Salaam had higher $\delta^{15}N$ values compared to the same species from the other locations. The

two species are known to be demersal mid-trophic level feeders, but had $\delta^{15}N$ values comparable to predator fish species from the present study. They also contained high contaminant concentrations indicating that they were feeding at higher trophic levels (**Paper II**). Variations in individual feeding behaviour and differences in baseline $\delta^{15}N$ among locations could explain the observed patterns, but is difficult to assess with this limited sample size. The sampling of marine fishes and prawns was based on what is sold on the fish markets, which results in a human consumption-biased selection of organisms. Full characterization of the local marine food web, including primary producers, was beyond the scope of the present study. Rather, a broad selection of different species representing low to high trophic level feeders and both the demersal and pelagic systems was sampled.

4.3 Contaminants of emerging concern

Chlorinated paraffins and dechloranes are considered contaminants of emerging concern even though they are subjected to international regulation. The regulation of SCCPs is associated with several exceptions for production and usage for certain purposes, as well as the unintentional presence of SCCPs in other CP mixtures or as degradation products (Guida et al., 2020). Very little data exist on the occurrence of emerging contaminants in the African environment, especially in biota. Eggs from free-range household chickens (*Gallus domesticus*) are ideal indicators for local contamination, and represent a commonly used matrix in global monitoring programs. The chicken egg is lipid-rich, important for human consumption, and can be used to make inferences about bioaccumulation (soil to hen) and maternal transfer (hen to egg) of lipophilic contaminants (Polder et al., 2016; Petrlik et al., 2022). The soil-chicken-egg pathway is typically studied when estimating human risk and intake of dioxins and dioxin-like PCBs (Polder et al., 2016), but is also relevant for other semi-volatile contaminants such as CPs. Relatively high levels of CPs were found in house dust in South Africa (Brits et al., 2020) and in breastmilk from African mothers (Krätschmer et al., 2021), demonstrating their ubiquitous presence.

In **Paper III,** we report high detection frequency of CPs and Dechlorane Plus in free-range chicken eggs and soil. Elevated concentrations of CPs and DP in soil were found at the two active dumpsites: the official dumpsite of Dar es Salaam located next to an e-waste handler and the informal dumpsite on Zanzibar (Figure 4). This finding is in accordance with air and soil concentrations from the same region (Nipen et al., 2022a). Elevated concentrations of SCCPs in eggs at the e-waste location was also found, but this difference was not statistically significant. Due to a low sample size, low spatial variation across sites, high variation in CP

concentrations in eggs within sites, and the relatively high concentrations in eggs from the reference location, no clear conclusions regarding an e-waste source could be made. Even though comparison between different laboratories and quantification methods is challenging, CP concentrations in soil and eggs from all locations in the present study was higher compared to eggs from reference locations (no e-waste) in Europe, Indonesia and Ghana, but lower compared to e-waste locations in China (Figure 4).

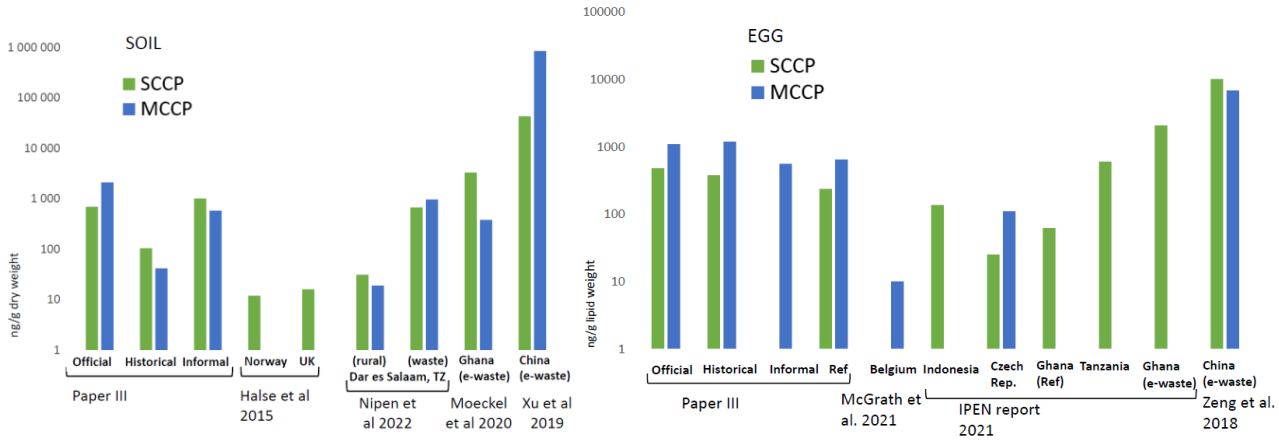


Figure 4. *Mean concentrations of SCCPs and MCCPs in soil and egg from the present study and data collected from the literature. Data is presented on a logarithmic scale.*

The waste disposal sites described in **Paper III** were all characterized as dumpsites because there are no particular safety measures in place for leachate management, and no regulations on types of waste permitted for disposal. E-waste and other valuable material are not separated from general waste before ending up at the dumpsites. However, valuable materials are removed from the dumpsites by local waste scavengers for recycling, refurbishing purposes or are sold to other traders (Ntapanta, 2022). Only the sampling site by the e-waste handler and the official dumpsite in Dar es Salaam were considered a "true" e-waste location. At the other dumpsites, we could not determine how long potential hazardous waste had been stored. The assessment of potential sources of emerging contaminants to local soil and chickens would have been improved by collecting any industrial feed that the chickens received, and by including a representative reference location. Unknown contaminant sources at the presumed reference location, e.g. open burning of waste, cannot be ruled out.

We found no apparent association in contaminant concentrations or composition pattern between soil and egg (**Paper III**). In general, MCCPs dominated in the eggs, and SCCPs dominated in the soil, except from at the official dumpsite, where MCCP concentrations were

higher than SCCPs (Figure 4). Dechlorane Plus dominated the dechlorane pattern in eggs and soil, except from soil at the historical dumpsite in Dar es Salaam, where concentrations of Dechlorane 603 was very high. This might be linked to a waste signal, but Dechlorane 603 also appears as an impurity in pesticides, and its occurrence here may instead be a signal of pesticide use (Nipen et al 2022b). A dominance of MCCPs over SCCPs in eggs is probably because MCCPs have higher K_{ow} and affinity to lipid-rich tissues. CPs with 5-8 Chlorine atoms were dominating the CP composition patterns in eggs, suggesting that these congeners are more readily accumulated in the hen and transferred to the egg during egg formation. The sampling of other tissues from the hen would allow a better assessment on the accumulation of contaminants from soil to chicken, tissue distribution, and the maternal transfer of contaminants from the hen to the egg during egg formation.

4.3.1 Human risk assessment of chlorinated paraffins

A complete assessment of the human health risks associated with the contaminants addressed herein is beyond the scope of this study. In **Paper III**, we assess the potential health concern from CP exposure via egg consumption based on the newly identified BMDLs and a MOE approach.

Based on the BMDLs and estimated daily intake of eggs in the Tanzanian population, SCCP concentrations in one egg represented potential human health concern (MOE<1000, **Paper III**). Even though only one out of 24 sampled eggs had MOE<1000, this finding indicates that concentration ranges in the upper percentiles could represent a health concern. Uncertainties and limitations following these assessments must be taken into account as the sample size is low and the assessment only describe a limited exposure scenario. Importantly, exposure from other sources (air, dust, and other types of food) or contaminant mixtures are not accounted for. Exposure risk to infants, toddlers and other sensitive population member are also not accounted for. Thus, the current risk assessment might be an underestimation. Further research need to be conducted on the sources of contaminants to the Tanzanian population and the associated health risks.

4.4 Shift in contaminant source regions: need for effective regulation of contaminants in products and waste

Monitoring of several legacy POPs, including PCBs, PBDEs and OCPs, in African air using passive samplers has been conducted through the MONitoring NETwork (MONET). These results have been implemented in the global monitoring plan (GMP) to assess the

effectiveness of the Stockholm Convention regulatory framework (White et al., 2020; UNEP/GMP3, 2021).

Ten years of monitoring POPs in African air conclude that ∑DDTs still dominate the contaminant profile, but are decreasing over time (White et al., 2020). Trends of PCB and PBDE concentrations in air vary among locations, but evidence point towards continuous input of these contaminants to the African environment (White et al., 2020; de Boer et al., 2023), including Tanzania (Nipen et al., 2022b). Important sources of environmental pollution from both legacy and emerging contaminants in Tanzania include urban areas, i.e. Dar es Salaam, and dumpsites (**Paper II and Paper III**, Nipen et al. 2022a).

Although the transboundary movement of hazardous waste is restricted by regulations, the e-waste trade still occurs and typically happens covertly, often under the disguise of being intended for reuse or recycling, and is difficult to measure or control. Regulatory bodies such as the Basel and Rotterdam Conventions lack the ability to efficiently regulate the movement of contaminants embedded in products. The Prior Informed Consent (PIC) regulation aiming to share information on how to safely store, transport, use, and dispose of chemicals is deficient, and there are currently no requirements for labelling of chemical additives in products (Babayemi et al., 2022; Guida et al., 2022). Because of this, large volumes of contaminants, such as CPs, can be transported to developing countries embedded in products such as plastics and electronics (Babayemi et al., 2015; Babayemi et al., 2019; Nevondo & Okonkwo, 2021). The recently established International Panel on Chemical Pollution (IPCP) work beyond geographical regions and legislations such as the Stockholm, Rotterdam and Basel Conventions to tackle the issues of chemical pollution and waste. Importantly, IPCP recognizes the significant role of less industrialized countries in the Global South regarding the burden of chemical pollution and consequences of waste mismanagement (Ågerstrand et al., 2023).

5. Conclusion

This thesis has presented and discussed the occurrence of legacy and emerging contaminants in the Tanzanian environment. Terrestrial and marine habitats, and environmental matrices ranging from soil to chicken eggs and marine biota have been sampled. This information has enabled a broad assessment of contaminant concentrations, patterns, dynamics and potential sources in a region where contaminant data has been very limited. The main conclusions drawn from the present work were as follows:

Spatial variation in concentrations of OCPs, PCBs and BFRs in the Tanzanian coastal marine system (**Paper I and II**):

- Concentrations of legacy contaminants were relatively low and intake of marine fishes and prawns from mainland Tanzania and Zanzibar do not represent high contaminant exposure
- Highest contaminant concentrations in marine fishes and prawns from central Dar es Salaam confirm that major urban and industrialized areas are important contaminant sources
- Higher concentration and detection frequency of HCB and BDE-209 on Zanzibar could indicate local point sources such as open burning of waste
- In general, contaminant concentrations increased with $\delta^{15}N$ indicating food web biomagnification
- Spatial variation in $\delta^{15}N$ across species reflects individual feeding behaviour or differences in baseline $\delta^{15}N$ among locations

Seasonal variation in the occurrence of OCPs, PCBs and BFRs in marine fishes and prawns from Zanzibar (**Paper I**):

- Increasing concentrations and changes in contaminant composition patterns following a period with heavy rain indicate an effect of atmospheric deposition and/or terrestrial runoff

Occurrence of CPs and dechloranes in chicken eggs and soil from various waste disposal sites in Tanzania (**Paper III**):

- High detection frequency of CPs and Dechlorane Plus in chicken eggs and soil highlights their ubiquitous presence even in regions with no production or intensive use
- No direct relationship in contaminant concentrations or patterns between soil and egg suggest specific mechanisms of accumulation and/or maternal transfer
- Risk assessment of CPs gave margin of exposures (MOE) close to or below the safety limit in two eggs from one location, suggesting human health concern for SCCP exposure via egg consumption

References

Abbasi, G., Li, L., & Breivik, K. (2019). Global historical stocks and emissions of PBDEs. *Environmental Science & Technology, 53*, 6330-6340. doi:https://doi.org/10.1021/acs.est.8b07032

Alava, J. J., & Gobas, F. (2012). Assessing biomagnification and trophic transport of persistent organic pollutants in the food chain of the Galapagos sea lion (Zalophus wollebaeki): conservation and management implications. *New Approaches to the Study of Marine Mammals*, 77-108. doi:https://doi.org/10.5772/51725

AMAP. (1998). *AMAP Assessment Report: Arctic Pollution Issues. Arctic Monitoring and Assessment Programme (AMAP), Oslo Norway.* Retrieved from https://www.amap.no/documents/doc/amap-assessment-report-arctic-pollution-issues/68

AMAP. (2017). AMAP Assessment 2016: Chemical of Emerging Arctic Concern. Arctic Monitoring and Assessment Programme (AMAP), Oslo, Norway. Retrieved from: https://www.amap.no/documents/doc/amap-assessment-2016-chemicals-of-emerging-arctic-concern/1624

Arnot, J. A., & Gobas, F. A. (2006). A review of bioconcentration factor (BCF) and bioaccumulation factor (BAF) assessments for organic chemicals in aquatic organisms. *Environmental Reviews, 14*(4), 257-297. doi:https://doi.org/10.1139/a06-005

Arp, H. P. H., Villers, F., Lepland, A., Kalaitzidis, S., Christanis, K., Oen, A. M., Breedveld, G. D., & Cornelissen, G. (2011). Influence of historical industrial epochs on pore water and partitioning profiles of polycyclic aromatic hydrocarbons and polychlorinated biphenyls in Oslo Harbor, Norway, sediment cores. *Environmental Toxicology and Chemistry, 30*(4), 843-851. doi: https://doi.org/10.1002/etc.466

Arukwe, A., Eggen, T., & Möder, M. (2012). Solid waste deposits as a significant source of contaminants of emerging concern to the aquatic and terrestrial environments—A developing country case study from Owerri, Nigeria. *Science of the Total Environment, 438*, 94-102. doi:https://doi.org/10.1016/j.scitotenv.2012.08.039

Babayemi, J., Sindiku, O., Osibanjo, O., & Weber, R. (2015). Substance flow analysis of polybrominated diphenyl ethers in plastic from EEE/WEEE in Nigeria in the frame of Stockholm Convention as a basis for policy advice. *Environmental Science and Pollution Research, 22*(19), 14502-14514. doi:https://doi.org/10.1007/s11356-014-3228-6

Babayemi, J. O., Nnorom, I. C., Osibanjo, O., & Weber, R. (2019). Ensuring sustainability in plastics use in Africa: consumption, waste generation, and projections. *Environmental Sciences Europe, 31*(1), 1-20. doi:https://doi.org/10.1186/s12302-019-0254-5

Babayemi, J. O., Nnorom, I. C., & Weber, R. (2022). Initial assessment of imports of chlorinated paraffins into Nigeria and the need of improvement of the Stockholm and Rotterdam Conventions. *Emerging Contaminants, 8*, 360-370. doi:https://doi.org/10.1016/j.emcon.2022.07.004

Barrie, L. A., Gregor, D., Hargrave, B., Lake, R., Muir, D., Shearer, R., Tracey, B., & Bidleman, T. (1992). Arctic contaminants: sources, occurrence and pathways. *Science of the Total Environment, 122*(1-2), 1-74.

Bocio, A., Llobet, J., Domingo, J., Corbella, J., Teixido, A., & Casas, C. (2003). Polybrominated diphenyl ethers (PBDEs) in foodstuffs: human exposure through the diet. *Journal of Agricultural and Food Chemistry, 51*(10), 3191-3195. doi:https://doi.org/10.1021/jf0340916

Bogdal, C., Alsberg, T., Diefenbacher, P. S., MacLeod, M., & Berger, U. (2015). Fast quantification of chlorinated paraffins in environmental samples by direct injection high-resolution mass spectrometry with pattern deconvolution. *Analytical Chemistry, 87*(5), 2852-2860. doi:https://doi.org/10.1021/ac504444d

Borgå, K., Fisk, A. T., Hoekstra, P. F., & Muir, D. C. (2004). Biological and chemical factors of importance in the bioaccumulation and trophic transfer of persistent organochlorine contaminants in arctic marine food webs. *Environmental Toxicology and Chemistry, 23*(10), 2367-2385. doi:https://doi.org/10.1897/03-518

Borgå, K., Kidd, K. A., Muir, D. C., Berglund, O., Conder, J. M., Gobas, F. A., Kucklick, J., Malm, O., & Powell, D. E. (2012). Trophic magnification factors: considerations of ecology, ecosystems, and study design. *Integrated Environmental Assessment and Management, 8*(1), 64-84. doi:https://doi.org/10.1002/ieam.244

Breivik, K., Armitage, J. M., Wania, F., & Jones, K. C. (2014). Tracking the global generation and exports of e-waste. Do existing estimates add up? *Environmental Science & Technology, 48*(15), 8735-8743. doi:https://doi.org/10.1021/es5021313

Breivik, K., Armitage, J. M., Wania, F., Sweetman, A. J., & Jones, K. C. (2015). Tracking the global distribution of persistent organic pollutants accounting for e-waste exports to developing regions. *Environmental Science & Technology, 50*(2), 798-805. doi:https://doi.org/10.1021/acs.est.5b04226

Breivik, K., Gioia, R., Chakraborty, P., Zhang, G., & Jones, K. C. (2011). Are reductions in industrial organic contaminants emissions in rich countries achieved partly by export of toxic wastes? *Environmental Science & Technology, 45*(21), 9154-9160. doi:https://doi.org/10.1021/es202320c

Breivik, K., Sweetman, A., Pacyna, J. M., & Jones, K. C. (2002). Towards a global historical emission inventory for selected PCB congeners - a mass balance approach: 1. Global production and consumption. *Science of the Total Environment, 290*(1-3), 181-198. doi:https://doi.org/10.1016/S0048-9697(01)01075-0

Brits, M., De Boer, J., Rohwer, E. R., De Vos, J., Weiss, J. M., & Brandsma, S. H. (2020). Short-, medium-, and long-chain chlorinated paraffins in South African indoor dust and cat hair. *Chemosphere, 238*, 124643. doi:https://doi.org/10.1016/j.chemosphere.2019.124643

Brown, J. H. (2014). Why are there so many species in the tropics? *Journal of biogeography, 41*(1), 8-22. doi: https://doi.org/10.1111/jbi.12228

Bruce-Vanderpuije, P., Megson, D., Reiner, E. J., Bradley, L., Adu-Kumi, S., & Gardella, J. A. (2019). The state of POPs in Ghana- A review on persistent organic pollutants: Environmental and human exposure. *Environmental Pollution, 245*, 331-342. doi:https://doi.org/10.1016/j.envpol.2018.10.107

Covaci, A., Harrad, S., Abdallah, M. A.-E., Ali, N., Law, R. J., Herzke, D., & de Wit, C. A. (2011). Novel brominated flame retardants: a review of their analysis, environmental fate and behaviour. *Environment International, 37*(2), 532-556. doi:https://doi.org/10.1016/j.envint.2010.11.007

de Boer, J., van Dijk, R., Abalos, M., & Abad, E. (2023). Persistent organic pollutants in air from Asia, Africa, Latin America, and the Pacific. *Chemosphere, 324*, 138271. doi:https://doi.org/10.1016/j.chemosphere.2023.138271

De Wit, C. A., Balmer, J., Muir, D. C., Vorkamp, K., & Wilson, S. (2019). Chemicals of emerging Arctic concern: preface. *Emerging Contaminants, 5*, 1-3. doi:https://doi.org/10.1016/j.emcon.2018.12.001

de Wit, C. A., Herzke, D., & Vorkamp, K. (2010). Brominated flame retardants in the Arctic environment—trends and new candidates. *Science of the Total Environment, 408*(15), 2885-2918. doi:https://doi.org/10.1016/j.scitotenv.2009.08.037

Desforges, J.-P., Hall, A., McConnell, B., Rosing-Asvid, A., Barber, J. L., Brownlow, A., De Guise, S., Eulaers, I., Jepson, P. D., & Letcher, R. J. (2018). Predicting global killer whale population collapse from PCB pollution. S*cience, 361*(6409), 1373-1376. doi:10.1126/science.aat1953

Djien Liem, A., Furst, P., & Rappe, C. (2000). Exposure of populations to dioxins and related compounds. *Food Additives & Contaminants, 17*(4), 241-259. doi:https://doi.org/10.1080/026520300283324

EFSA (2020). Risk assessment of chlorinated paraffins in feed and food. EFSA Journal, 18(3), e05991. doi:https://doi.org/10.2903/j.efsa.2020.5991

EFSA (2023). Glossary. Retrieved from: https:// www.efsa.europa.eu/en/glossary/risk-assessment

E.C. (2011). Commission Regulation (EC) No 1259/2011of 2 December 2011 amending Regulation (EC) No 1881/2006 as regards maximum levels for dioxins, dioxin-like PCBs and non dioxin-like PCBs in foodstuffs. Retrieved from: https://eur-lex.europa.eu/LexUriServ/LexUriServ.do?uri=OJ:L:2011:320:0018:0023:EN:PDF

E.C. (2013) Directive 2013/39/EC of the European Parliament and of the Council of 12 August 2013

amending Directives 2000/60/EC and 2008/105/EC as regards priority substances in the field of water policy. Retrieved from: https://eur-lex.europa.eu/LexUriServ/LexUriServ.do?uri=OJ:L:2013:226:0001:0017:en:PDF

Elfvendahl, S., Mihale, M., Kishimba, M. A., & Kylin, H. (2004). Pesticide pollution remains severe after cleanup of a stockpile of obsolete pesticides at Vikuge, Tanzania. *AMBIO: A Journal of the Human Environment, 33*(8), 503-508. doi:https://doi.org/10.1579/0044-7447-33.8.503

Elibariki, R., & Maguta, M. M. (2017). Status of pesticides pollution in Tanzania–A review. *Chemosphere, 178*, 154-164. doi:https://doi.org/10.1016/j.chemosphere.2017.03.036

Forti, V., Baldé, C. P., Kuehr, R., & Bel, G. (2020). The Global E-waste Monitor 2020: Quantities, flows and the circular economy potential. United Nations University (UNU)/United Nations Institute for Training and Research (UNITAR) – co-hosted SCYCLE Programme, International Telecommunication Union (ITU) & International Solid Waste Association (ISWA), Bonn/Geneva/Rotterdam., 2020

Fries, G. F. (1995). A review of the significance of animal food products as potential pathways of human exposures to dioxins. *Journal of Animal Science, 73*(6), 1639-1650. doi:https://doi.org/10.2527/1995.7361639x

Fu, C.-T., & Wu, S.-C. (2006). Seasonal variation of the distribution of PCBs in sediments and biota in a PCB-contaminated estuary. *Chemosphere, 62*(11), 1786-1794. doi:https://doi.org/10.1016/j.chemosphere.2005.07.034

Gioia, R., Akindele, A. J., Adebusoye, S. A., Asante, K. A., Tanabe, S., Buekens, A., & Sasco, A. J. (2014). Polychlorinated biphenyls (PCBs) in Africa: a review of environmental levels. *Environmental Science and Pollution Research, 21*(10), 6278-6289. doi:https://doi.org/10.1007/s11356-013-1739-1

Gioia, R., Eckhardt, S., Breivik, K., Jaward, F. M., Prieto, A., Nizzetto, L., & Jones, K. C. (2011). Evidence for major emissions of PCBs in the West African region. *Environmental Science & Technology, 45*(4), 1349-1355. doi:https://doi.org/10.1021/es1025239

Guida, Y., Capella, R., Kajiwara, N., Babayemi, J. O., Torres, J. P. M., & Weber, R. (2022). Inventory approach for short-chain chlorinated paraffins for the Stockholm Convention implementation in Brazil. *Chemosphere, 287*, 132344. doi:https://doi.org/10.1016/j.chemosphere.2021.132344

Guida, Y., Capella, R., & Weber, R. (2020). Chlorinated paraffins in the technosphere: A review of available information and data gaps demonstrating the need to support the Stockholm Convention implementation. *Emerging Contaminants, 6*, 143-154. doi:https://doi.org/10.1016/j.emcon.2020.03.003

Hobson, K. A., & Welch, H. E. (1992). Determination of trophic relationships within a high Arctic marine food web using δ 13 C and δ 15 N analysis. *Marine Ecology Progress Series*, 9-18. doi:https://www.jstor.org/stable/24829721

Iozza, S., Müller, C. E., Schmid, P., Bogdal, C., & Oehme, M. (2008). Historical profiles of chlorinated paraffins and polychlorinated biphenyls in a dated sediment core from Lake Thun (Switzerland). *Environmental Science & Technology, 42*(4), 1045-1050. doi:https://doi.org/10.1021/es702383t

IPEN (2005). Hotspot Report for Pattern of Practices: PCBs Sources and Releases in Tanzania. The International POPs Elimination Project. Retrieved from: https://ipen.org/sites/default/files/documents/3urt_pcb_sources_and_releases-en.pdf

Kidd, K. A., Bootsma, H. A., Hesslein, R. H., Muir, D. C., & Hecky, R. E. (2001). Biomagnification of DDT through the benthic and pelagic food webs of Lake Malawi, East Africa: importance of trophic level and carbon source. *Environmental Science & Technology, 35*(1), 14-20. doi:https://doi.org/10.1021/es001119a

Kimbrough, R. D., & Jensen, A. A. (1989). Chemistry, usage and environmental contamination. In Halogenated biphenyls, terphenyls, naphthalenes, dibenzodioxins and related products

Kishimba, M., Henry, L., Mwevura, H., Mmochi, A., Mihale, M., & Hellar, H. (2004). The status of pesticide pollution in Tanzania. *Talanta, 64*(1), 48-53. doi:https://doi.org/10.1016/j.talanta.2003.11.047

Kruitwagen, G., Pratap, H., Covaci, A., & Bonga, S. W. (2008). Status of pollution in mangrove ecosystems along the coast of Tanzania. Ecology and toxicology of mangrove fauna in Tanzania, 51. doi:https://doi.org/10.1016/j.marpolbul.2008.02.018

Krätschmer, K., Malisch, R., & Vetter, W. (2021). Chlorinated paraffin levels in relation to other persistent organic pollutants found in pooled human milk samples from primiparous mothers in 53 countries. *Environmental Health Perspectives, 129*(8), 087004. doi:https://doi.org/10.1289/EHP7696

Lammel, G., Degrendele, C., Gunthe, S. S., Mu, Q., Muthalagu, A., Audy, O., Biju, C. V., Kukučka, P., Mulder, M. D., & Octaviani, M. (2018). Revolatilisation of soil-accumulated pollutants triggered by the summer monsoon in India. *Atmospheric Chemistry and Physics, 18*(15), 11031-11040. doi:https://doi.org/10.5194/acp-18-11031-2018

Layman, C. A., Araujo, M. S., Boucek, R., Hammerschlag-Peyer, C. M., Harrison, E., Jud, Z. R., Matich, P., Rosenblatt, A. E., Vaudo, J. J., & Yeager, L. A. (2012). Applying stable isotopes to examine food-web structure: an overview of analytical tools. *Biological Reviews, 87*(3), 545-562. doi:https://doi.org/10.1111/j.1469-185X.2011.00208.x

Leslie, H., van Bavel, B., Abad, E., & De Boer, J. (2013). Towards comparable POPs data worldwide with global monitoring data and analytical capacity building in Africa, Central and Latin America, and the South Pacific. *TrAC: Trends in Analytical Chemistry, 46*, 85-97. doi:https://doi.org/10.1016/j.trac.2013.01.009

Letcher, R. J., Bustnes, J. O., Dietz, R., Jenssen, B. M., Jørgensen, E. H., Sonne, C., Verreault, J., Vijayan, M. M., & Gabrielsen, G. W. (2010). Exposure and effects assessment of persistent organohalogen contaminants in arctic wildlife and fish. *Science of the Total Environment, 408*(15), 2995-3043. doi:https://doi.org/10.1016/j.scitotenv.2009.10.038

Li, Y., Lin, T., Hu, L., Feng, J., & Guo, Z. (2016). Time trends of polybrominated diphenyl ethers in East China Seas: response to the booming of PBDE pollution industry in China. *Environment International, 92*, 507-514. doi:https://doi.org/10.1016/j.envint.2016.04.033

Ma, J., Hung, H., & Macdonald, R. W. (2016). The influence of global climate change on the environmental fate of persistent organic pollutants: A review with emphasis on the Northern Hemisphere and the Arctic as a receptor. *Global and Planetary Change, 146*, 89-108. doi:https://doi.org/10.1016/j.gloplacha.2016.09.011

Machiwa, J. F. (2010). Coastal Marine Pollution in Dar es Salaam (Tanzania) relative to Recommended Environmental Quality Targets for the Western Indian Ocean. *Western Indian Ocean Journal of Marine Science, 9*(1), 17-30.

Magashi, A., & Schluep, M. (2011). E-Waste Assessment Tanzania. Retrieved from: https://www.unido.org/sites/default/files/2011-03/110120_Assessment-Tanzania_FINAL_01_0.PDF

MALF. (2016). The Tanzanian fisheries sector: Challenges and opportunities. Retrieved from: https://www.wiomsa.org/download/the-tanzanian-fisheries-sectors-challenges-and-opportunities/

Manyilizu, W. B. (2019). Pesticides, anthropogenic activities, history and the health of our environment: lessons from Africa. doi:https://doi.org/10.5772/intechopen.82600

McGrath, T. J., Limonier, F., Poma, G., Bombeke, J., Winand, R., Vanneste, K., Andjelkovic, M., Van Hoeck, E., Joly, L., & Covaci, A. (2021). Concentrations and distribution of chlorinated paraffins in Belgian foods. *Environmental Pollution, 291*, 118236. doi:https://doi.org/10.1016/j.envpol.2021.118236

Melymuk, L., Blumenthal, J., Sáňka, O. e., Shu-Yin, A., Singla, V., Šebková, K. i., Pullen Fedinick, K., & Diamond, M. L. (2022). Persistent Problem: Global Challenges to Managing PCBs. *Environmental Science & Technology*. doi:https://doi.org/10.1021/acs.est.2c01204

Minagawa, M., & Wada, E. (1984). Stepwise enrichment of 15N along food chains: further evidence and the relation between δ15N and animal age. *Geochimica et cosmochimica acta, 48*(5), 1135-1140.

Mirmigkou, S., & de Boer, J. (2016). DDT and metabolites. *Dioxin and Related Compounds: Special Volume in Honor of Otto Hutzinger*, 355-378.

Munschy, C., Vigneau, E., Bely, N., Héas-Moisan, K., Olivier, N., Pollono, C., Hollanda, S., & Bodin, N. (2020). Legacy and emerging organic contaminants: Levels and profiles in top predator fish from the western Indian Ocean in relation to their trophic ecology. *Environmental research, 188*, 109761. doi:https://doi.org/10.1016/j.envres.2020.109761

Mwakalapa, E. B., Mmochi, A. J., Müller, M. H. B., Mdegela, R. H., Lyche, J. L., & Polder, A. (2018). Occurrence and levels of persistent organic pollutants (POPs) in farmed and wild marine fish from Tanzania. A pilot study. *Chemosphere, 191*, 438-449. doi:https://doi.org/10.1016/j.chemosphere.2017.09.121

Mwevura, H., Bouwman, H., Kylin, H., Vogt, T., & Issa, M. A. (2020). Organochlorine pesticides and polycyclic aromatic hydrocarbons in marine sediments and polychaete worms from the west coast of Unguja island, Tanzania. *Regional Studies in Marine Science, 36*, 101287. doi:https://doi.org/10.1016/j.rsma.2020.101287

Mwevura, H., Othman, O. C., & Mhehe, G. L. (2002). Organochlorine pesticide residues in sediments and biota from the coastal area of Dar es Salaam city, Tanzania. *Marine Pollution Bulletin, 45*(1-12), 262-267. doi:https://doi.org/10.1016/S0025-326X(01)00331-9

Müller, M., Polder, A., Brynildsrud, O., Karimi, M., Lie, E., Manyilizu, W., Mdegela, R., Mokiti, F., Murtadha, M., & Nonga, H. (2017). Organochlorine pesticides (OCPs) and polychlorinated biphenyls (PCBs) in human breast milk and associated health risks to nursing infants in Northern Tanzania. *Environmental Research, 154*, 425-434. doi:https://doi.org/10.1016/j.envres.2017.01.031

Müller, M. H. B., Polder, A., Brynildsrud, O., Grønnestad, R., Karimi, M., Lie, E., Manyilizu, W. B., Mdegela, R., Mokiti, F., & Murtadha, M. (2019). Prenatal exposure to persistent organic pollutants in Northern Tanzania and their distribution between breast milk, maternal blood, placenta and cord blood. *Environmental Research, 170*, 433-442. doi:https://doi.org/10.1016/j.envres.2018.12.026

Nevondo, V., & Okonkwo, O. J. (2021). Status of short-chain chlorinated paraffins in matrices and research gap priorities in Africa: a review. *Environmental Science and Pollution Research, 28*(38), 52844-52861. doi:https://doi.org/10.1007/s11356-021-15924-w

Nipen, M., Vogt, R. D., Bohlin-Nizzetto, P., Borgå, K., Mwakalapa, E. B., Borgen, A. R., Jørgensen, S. J., Ntapanta, S. M., Mmochi, A. J., & Schlabach, M. (2022a). Spatial trends of chlorinated paraffins and dechloranes in air and soil in a tropical urban, suburban, and rural environment. *Environmental Pollution, 292*, 118298. doi:https://doi.org/10.1016/j.envpol.2021.118298

Nipen, M., Vogt, R. D., Bohlin-Nizzetto, P., Borgå, K., Mwakalapa, E. B., Borgen, A. R., Schlabach, M., Christensen, G., Mmochi, A. J., & Breivik, K. (2022b). Increasing Trends of Legacy and Emerging Organic Contaminants in a Dated Sediment Core From East-Africa. *Frontiers in Environmental Science*, 711. doi:https://doi.org/10.3389/fenvs.2021.805544

Ntapanta, S. M. (2022). *Gathering the African Technosphere: An Ethnography of Informal Electronic Waste Recycling in Tanzania*: Series of dissertations submitted to the Faculty of Social Sciences, University of Oslo No. 927.

Ontiveros-Cuadras, J. F., Ruiz-Fernández, A. C., Sanchez-Cabeza, J.-A., Sericano, J., Pérez-Bernal, L. H., Páez-Osuna, F., Dunbar, R. B., & Mucciarone, D. A. (2019). Recent history of persistent organic pollutants (PAHs, PCBs, PBDEs) in sediments from a large tropical lake. *Journal of Hazardous Materials, 368*, 264-273. doi:https://doi.org/10.1016/j.jhazmat.2018.11.010

Paine, R. T. (1966). Food web complexity and species diversity. *The American Naturalist, 100*(910), 65-75. Retrieved from: http://links.jstor.org/sici?sici=0003-0147%28196601%2F02%29100%3A910%3C65%3AFWCASD%3E2.0.CO%3B2-D

Perkins, D. N., Drisse, M.-N. B., Nxele, T., & Sly, P. D. (2014). E-waste: a global hazard. *Annals of Global Health, 80*(4), 286-295. doi:https://doi.org/10.1016/j.aogh.2014.10.001

Petrlik, J., Bell, L., DiGangi, J., Allo'o, S. M. A. o., Kuepouo, G., Ochola, G. O., Grechko, V., Jelinek, N., Strakova, J., & Skalsky, M. (2022). Monitoring dioxins and PCBs in eggs as sensitive indicators for environmental pollution and global contaminated sites and recommendations for reducing and controlling releases and exposure. *Emerging Contaminants, 8*, 254-279. doi:https://doi.org/10.1016/j.emcon.2022.05.001

Polder, A., Müller, M., Brynildsrud, O., De Boer, J., Hamers, T., Kamstra, J., Lie, E., Mdegela, R., Moberg, H., & Nonga, H. (2016). Dioxins, PCBs, chlorinated pesticides and brominated flame retardants in free-range chicken eggs from peri-urban areas in Arusha, Tanzania: levels and implications for human health. *Science of the Total Environment, 551*, 656-667. doi:https://doi.org/10.1016/j.scitotenv.2016.02.021

Polder, A., Müller, M., Lyche, J., Mdegela, R., Nonga, H., Mabiki, F., Mbise, T., Skaare, J., Sandvik, M., & Skjerve, E. (2014). Levels and patterns of persistent organic pollutants (POPs) in tilapia (*Oreochromis* sp.) from four different lakes in Tanzania: geographical differences and implications for human health. *Science of the Total Environment, 488*, 252-260. doi:https://doi.org/10.1016/j.scitotenv.2014.04.085

Ruus, A., Green, N. W., Maage, A., Amundsen, C. E., Schøyen, M., & Skei, J. (2010). Post World War II orcharding creates present day DDT-problems in The Sørfjord (Western Norway)–A case study. *Marine Pollution Bulletin, 60*(10), 1856-1861. doi:https://doi.org/10.1016/j.marpolbul.2010.06.048

Sagiv, S. K., Kogut, K., Gaspar, F. W., Gunier, R. B., Harley, K. G., Parra, K., Villasenor, D., Bradman, A., Holland, N., & Eskenazi, B. (2015). Prenatal and childhood polybrominated diphenyl ether (PBDE) exposure and attention and executive function at 9–12 years of age. *Neurotoxicology and Teratology, 52*, 151-161. doi:https://doi.org/10.1016/j.ntt.2015.08.001

Shi, J., Li, P., Li, Y., Liu, W., Zheng, G. J.-S., Xiang, L., & Huang, Z. (2016). Polychlorinated biphenyls and organochlorine pesticides in surface sediments from Shantou Bay, China: sources, seasonal variations and inventories. *Marine Pollution Bulletin, 113*(1-2), 585-591. doi:https://doi.org/10.1016/j.marpolbul.2016.09.006

Sparks, T. H., Scott, W. A., & Clarke, R. T. (1999). Traditional multivariate techniques: potential for use in ecotoxicology. *Environmental Toxicology and Chemistry: An International Journal, 18*(2), 128-137. doi:https://doi.org/10.1002/etc.5620180206

Ssebugere, P., Sillanpää, M., Matovu, H., & Mubiru, E. (2019). Human and environmental exposure to PCDD/Fs and dioxin-like PCBs in Africa: A review. *Chemosphere, 223*, 483-493. doi:https://doi.org/10.1016/j.chemosphere.2019.02.065

Sverko, E., Tomy, G. T., Reiner, E. J., Li, Y.-F., McCarry, B. E., Arnot, J. A., Law, R. J., & Hites, R. A. (2011). Dechlorane plus and related compounds in the environment: a review. *Environmental Science & Technology, 45*(12), 5088-5098. doi:https://doi.org/10.1021/es2003028

Todd, G., Msuya, I., Levira, F., & Moshi, I. (2019). City Profile: Dar es Salaam, Tanzania. *Environment and Urbanization ASIA, 10*(2), 193-215. doi:https://doi.org/10.1177/0975425319859175

UNEP (2001). Final Act of the Conference of Plenipotentiaries on the Stockholm Convention on Persistent Organic Pollutants. United Nations Environmental Programme, Stockholm, Sweden.

UNEP (2002). Basel Convention on the control of transboundary movemets of hazardous wastes and their disposal. Retrieved from: http://www.basel.int/Portals/4/Basel%20Convention/docs/text/BaselConventionText-e.pdf

UNEP (2004). Rotterdam Convention in the prior informed consent procedure for certain hazardous chemicals and pesticides in international trade. Retrieved from: http://www.pic.int/TheConvention/Overview/TextoftheConvention/tabid/1048/language/en-US/Default.aspx.

Stockholm Convention on Persistent Organic Pollutants. Annex A, decision SC-8/11 (2017). Retrieved from: http://chm.pops.int/Implementation/Alternatives/AlternativestoPOPs/ChemicalslistedinAnnexA/Shortchainchlorinatedparaffins(SCCPs)/tabid/5986/Default.aspx.

UNEP/GMP3 (2021). Global monitoring plan for persistent organic pollutants. Under the Stockholm Convention article 16 on effectiveness evaluation. Third regional monitoring report. Africa region. Retrieved from: http://chm.pops.int/implementation/globalmonitoringplan/monitoringreports/tabid/525/default.aspx

van der Schyff, V., Yive, N. S. C. K., Polder, A., Cole, N. C., Tatayah, V., Kylin, H., & Bouwman, H. (2021). Persistent organic pollutants in sea bird eggs from the Indian Ocean's Mascarene Basin. *Science of the Total Environment, 771*, 145348. doi:https://doi.org/10.1016/j.scitotenv.2021.145348

van Mourik, L. M., Gaus, C., Leonards, P. E., & de Boer, J. (2016). Chlorinated paraffins in the environment: A review on their production, fate, levels and trends between 2010 and 2015. *Chemosphere, 155*, 415-428. doi:https://doi.org/10.1016/j.chemosphere.2016.04.037

Viganò, L., Roscioli, C., & Guzzella, L. (2011). Decabromodiphenyl ether (BDE-209) enters the food web of the River Po and is metabolically debrominated in resident cyprinid fishes. *Science of the Total Environment, 409*(23), 4966-4972. doi:https://doi.org/10.1016/j.scitotenv.2011.07.062

Walters, D., Jardine, T., Cade, B. S., Kidd, K., Muir, D., & Leipzig-Scott, P. (2016). Trophic magnification of organic chemicals: A global synthesis. *Environmental Science & Technology, 50*(9), 4650-4658. doi:https://doi.org/10.1021/acs.est.6b00201

Wang, P., Zhang, Q., Zhang, H., Wang, T., Sun, H., Zheng, S., Li, Y., Liang, Y., & Jiang, G. (2016). Sources and environmental behaviors of Dechlorane Plus and related compounds—A review. *Environment International, 88*, 206-220. doi:https://doi.org/10.1016/j.envint.2015.12.026

Wania, F., & Mackay, D. (1993). Global fractionation and cold condensation of low volatility organochlorine compounds in polar regions. *Ambio*, 10-18.

Webster, E., Mackay, D., & Wania, F. (1998). Evaluating environmental persistence. *Environmental Toxicology and Chemistry: An International Journal, 17*(11), 2148-2158.

Wei, G.-L., Liang, X.-L., Li, D.-Q., Zhuo, M.-N., Zhang, S.-Y., Huang, Q.-X., Liao, Y.-S., Xie, Z.-Y., Guo, T.-L., & Yuan, Z.-J. (2016). Occurrence, fate and ecological risk of chlorinated paraffins in Asia: a review. *Environment International, 92*, 373-387. doi:https://doi.org/10.1016/j.envint.2016.04.002

White, K. B., Kalina, J. í., Scheringer, M., Přibylová, P., Kukučka, P., Kohoutek, J. í., Prokeš, R., & Klánová, J. (2020). Temporal trends of persistent organic pollutants across Africa after a decade of MONET passive air sampling. *Environmental Science & Technology, 55*(14), 9413-9424. doi:https://dx.doi.org/10.1021/acs.est.0c03575

WHO (2021). *World Malaria Report* Retrieved from: https://www.who.int/teams/global-malaria-programme/reports/world-malaria-report-2021

Wilkinson, J. L., Boxall, A. B., Kolpin, D. W., Leung, K. M., Lai, R. W., Galbán-Malagón, C., Adell, A. D., Mondon, J., Metian, M., & Marchant, R. A. (2022). Pharmaceutical pollution of the world's rivers. *Proceedings of the National Academy of Sciences, 119*(8), e2113947119. doi:https://doi.org/10.1073/pnas.2113947119

Wong, F., Hung, H., Dryfhout-Clark, H., Aas, W., Bohlin-Nizzetto, P., Breivik, K., Mastromonaco, M. N., Lundén, E. B., Ólafsdóttir, K., & Sigurðsson, Á. (2021). Time trends of persistent organic pollutants (POPs) and Chemicals of Emerging Arctic Concern (CEAC) in Arctic air from 25 years of monitoring. *Science of the Total Environment, 775*, 145109. doi:https://doi.org/10.1016/j.scitotenv.2021.145109

Wong, M., Wu, S., Deng, W. J., Yu, X., Luo, Q., Leung, A., Wong, C., Luksemburg, W., & Wong, A. (2007). Export of toxic chemicals–a review of the case of uncontrolled electronic-waste recycling. *Environmental Pollution, 149*(2), 131-140. doi:https://doi.org/10.1016/j.envpol.2007.01.044

WorldBank. (2016). Africa Stockpiles Program, Report No. 108524. Retrieved from: https://documents1.worldbank.org/curated/en/168841477341223021/pdf/108524-PPAR-PUBLIC.pdf

Yhdego, M. (2017). From a sanitary landfill to a dump site: Pugu Kinyamwezi community curse in Dar es Salaam, Tanzania. *ERC—Environmental Resources Consultancy*. doi:https://doi.org/10.13140/RG.2.2.27828.04483

Yu, Z., Lu, S., Gao, S., Wang, J., Li, H., Zeng, X., Sheng, G., & Fu, J. (2010). Levels and isomer profiles of Dechlorane Plus in the surface soils from e-waste recycling areas and industrial areas in South China. *Environmental Pollution, 158*(9), 2920-2925. doi:https://doi.org/10.1016/j.envpol.2010.06.003

Zeng, Y. H., Huang, C. C., Luo, X. J., Liu, Y. N., Ren, Z. H., & Mai, B. X. (2018). Polychlorinated biphenyls and chlorinated paraffins in home-produced eggs from an e-waste polluted area in South China: Occurrence and human dietary exposure. *Environment International, 116*, 52-59. doi:https://doi.org/10.1016/j.envint.2018.04.006

Zhang, H., Jiang, L., Zhou, X., Zeng, T., He, Z., Huang, X., Chen, J., & Song, S. (2018). Determination of hexachlorobutadiene, pentachlorobenzene, and hexachlorobenzene in waste incineration fly ash using ultrasonic extraction followed by column cleanup and GC-MS analysis. *Analytical and Bioanalytical Chemistry, 410*(7), 1893-1902. doi:https://doi.org/10.1007/s00216-018-0849-5

Zhang, H., Teng, Y., Doan, T. T. T., Yat, Y. W., Chan, S. H., & Kelly, B. C. (2017). Stable nitrogen and carbon isotopes in sediments and biota from three tropical marine food webs: Application to chemical bioaccumulation assessment. *Environmental Toxicology and Chemistry, 36*(9), 2521-2532. doi: https://doi.org/10.1002/etc.3796

Zhao, G., Zhou, H., Wang, D., Zha, J., Xu, Y., Rao, K., Ma, M., Huang, S., & Wang, Z. (2009). PBBs, PBDEs, and PCBs in foods collected from e-waste disassembly sites and daily intake by local residents. *Science of the Total Environment, 407*(8), 2565-2575. doi:https://doi.org/10.1016/j.scitotenv.2008.11.062

Ågerstrand, M., Arinaitwe, K., Backhaus, T., Barra, R. O., Diamond, M. L., Grimalt, J. O., Groh, K., Kandie, F., Kurt-Karakus, P. B., & Letcher, R. J. (2023). Key Principles for the Intergovernmental Science–Policy Panel on Chemicals and Waste. *Environmental Science & Technology, 57*(6), 2205-2208. doi:https://doi.org/10.1021/acs.est.2c08283

Appendix

Table A1. *Contaminants (groups and individual congeners) analyzed in this study. Information on their production start, start of regulation and listing in the Stockholm Convention is added where applicable.*

Compound group / congener	**Name**	**Start of production**	**Start of regulation**	**Listed under the Stockholm Connvention**
PCBs [1), 2)]		1930s	1970s	2004
PCB-28	2,4,4'-Trichlorobiphenyl			
PCB-52	2,2',5,5'-Tetrachlorobiphenyl			
PCB-74	2,4,4',5-Tetrachlorobiphenyl			
PCB-99	2,2',4,4',5-Pentachlorobiphenyl			
PCB-101	2,2'4,5,5'-Pentachlorobiphenyl			
PCB-105	2,3,3',4,4'-Pentachlorobiphenyl			
PCB-118	2,3',4,4',5-Pentachlorobiphenyl			
PCB-153	2,2',4,4'5,5'-Hexachlorobiphenyl			
PCB-128	2,2',3,3'4,4'-Hexachlorobiphenyl			
PCB-136	2,2',3,3',6,6'-Hexachlorobiphenyl			
PCB-138	2,2',3,4,4',5-Hexachlorobiphenyl			
PCB-156	2,3,3'4,4',5-Hexachlorobiphenyl			
PCB-170	2,2',3,3'4,4',5-Heptachlorobiphenyl			
PCB-180	2,2',3,4,4'5,5'-Heptachlorobiphenyl			
PCB-183	2,2',3,4,4',5',6-Heptachlorobiphenyl			
PCB-187	2,2'3,4'5,5',6-Heptachlorobiphenyl			
OCPs [1)]				
HCB	Hexachlorobenzene			2004
α-HCH	Hexachlorohexane			2010
β-HCH	Hexachlorohexane			2011
γ-HCH	Hexachlorohexane / lindane			2015
Heptachlor				2004
oxychlordane				2004
trans-chlordane				2004
cis-chlordane				2004
trans-Nonachlor				2004
cis-Nonachlor				2004
DDTs [1), 3)]		1940s	1970s	2004

p,p'-DDE	Dichlorodiphenyldichloroethylene			
o,p'-DDD	Dichlorodiphenyldichloroethane			
p,p'-DDD				
o,p'-DDT	Dichlorodiphenyltrichloroethane			
p,p'-DDT				
Mirex		1950s	1970s	2004
PBDEs [1), 4)]		1970s	1990s	
Penta-BDE mix			2004	2009
BDE-28	2,4-Dibromo-1-(4-bromophenoxy)benzene			
BDE-47	2,2'4,4'-Tetrabromodiphenyl ether			
BDE-99	2,2',4,4',5-Pentabromodiphenyl ether			
BDE-100	1,3,5-Tribromo-2-2(2,4-dibromophenoxy)benzene			
BDE-153	2,2',4,4'5,5'-Hexabromodiphenyl ether			
Octa-BDE mix			2004	2009
BDE-154	2,2',4,4'5,6'-Hexabromodiphenyl ether			
BDE-183	2,2',3,4,4'5',6'-Heptabromodiphenyl ether			
BDE-196	2,2',3,3'4,4',5,6'-Octabromodiphenyl ether			
BDE-202	2,2',3,3',5,5',6,6'-Octabromodiphenyl ether			
BDE-206	2,2',3,3',4,4',5,5',6-Nonabromodiphenyl ether			
BDE-207	2,2',3,3',4,4',5,6,6'-Nonabromodiphenyl ether			
BDE-208	2,2',3,3',4,5,5',6,6'-Nonabromodiphenyl ether			
Deca-BDE mix			2008	2017
BDE-209	Decabromodiphenyl oxide			
Non-PBDE BFRs				
HBCDD	Hexabromocyclododecane			2013
DPTE	1,3,5-tribromo-2-benzene			
HBB	1H-Benzimidiazol-2-yl(phenyl)methanol			
PBT	pbt-1033 hydrochloride			
PBEB	Pentabromoethylbenzene			
BTBPE	1,2-Bis(2,4,6-tribromophenoxy)ethane			
DBDPE	Decabromodiphenyl ethane			
Chlorinated paraffins [1)]				

SCCPs	Short-chain chlorinated paraffins (C10-C13)			2017
MCCPs	Medium-chain chlorinated paraffins (C14-C17)			Under consideration
Dechloranes [1)]				
DP *anti*	Dechlorane Plus *anti* isomer			2023
DP *syn*	Dechlorane Plus *syn* isomer			2023
Dec 601	Dechlorane 601			
Dec 602	Dechlorane 602			
Dec 603	Dechlorane 603			
Dec 604	Dechlorane 604			

[1)] UNEP (2001)
[2)] Breivik et al. (2002)
[3)] Barrie et al. (1992)
[4)] PolyBDEs EQS dossier (2011)

Paper I



I

Science of the Total Environment 774 (2021) 145652

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

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

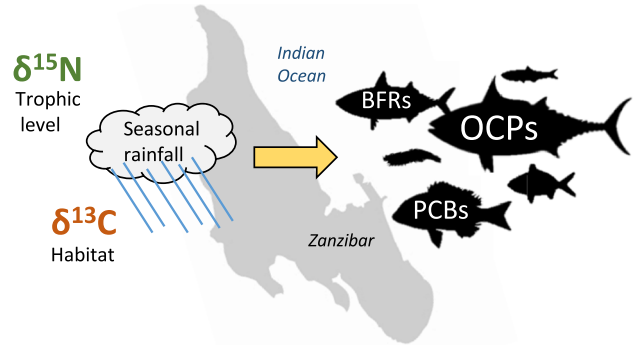
Ane Haarr [a,*], Eliezer B. Mwakalapa [b], Aviti J. Mmochi [c], Jan L. Lyche [d], Anders Ruus [e,a], Halima Othman [f], Martin M. Larsen [g], Katrine Borgå [a,h]

[a] Department of Biosciences, University of Oslo, P.O.Box 1066, 0316 Oslo, Norway
[b] Department of Natural Sciences, Mbeya University of Science and Technology, P.O. Box 131, Mbeya, Tanzania
[c] Institute of Marine Science, University of Dar es Salaam, P.O. Box 668, Zanzibar, Tanzania
[d] Norwegian University of Life Sciences, Ullevålsveien 72, 0474 Oslo, Norway
[e] Norwegian Institute for Water Research, Gaustadalleen 21, 0349 Oslo, Norway
[f] State University of Zanzibar, P.O.BOX 146, Tunguu, Zanzibar, Tanzania
[g] University of Aarhus, Institute of Bioscience, Frederiksborgvej 399, 4000 Roskilde, Denmark
[h] Center for Biogeochemistry in the Anthropocene, University of Oslo, PB 1066, 0316 Oslo, Norway

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



ARTICLE INFO

Article history:
Received 14 November 2020
Received in revised form 5 January 2021
Accepted 1 February 2021
Available online 9 February 2021

Editor: Adrian Covaci

Keywords:
Seasonal variation
Tropical ecotoxicology
Coastal ecosystems
Brominated flame retardants
Organochlorines
PCB

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-

* Corresponding author at: Department of Biosciences, University of Oslo, P.O.Box 1066, 0316 Oslo, Norway.
E-mail addresses: ane.haarr@ibv.uio.no (A. Haarr), mwakalapaeb@gmail.com (E.B. Mwakalapa), avitimmochi@gmail.com (A.J. Mmochi), jan.l.lyche@nmbu.no (J.L. Lyche), anders.ruus@niva.no (A. Ruus), halimaothman2007@gmail.com (H. Othman), mml@bios.au.dk (M.M. Larsen), katrine.borga@ibv.uio.no (K. Borgå).

https://doi.org/10.1016/j.scitotenv.2021.145652

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,

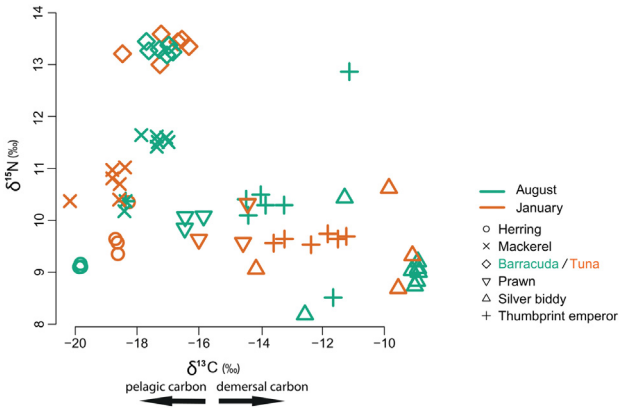


Fig. 1. Stable isotopes of carbon and nitrogen in homogenized muscle tissue, or whole body (herring), of marine fish and prawn sampled on Zanzibar in January and August 2018. The tuna and the barracuda are both representative of a pelagic predator, and different colors indicate that they are sampled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

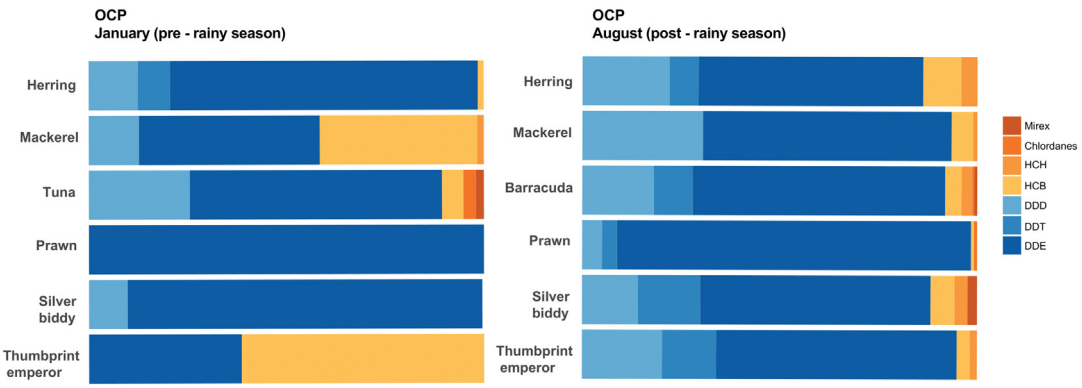


Fig. 2. Relative contribution of each compound to total organochlorine pesticide (OCP) concentration in fish and prawn from Zanzibar, Tanzania in January (pre- rainy season) and August (post- rainy season) 2018.

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

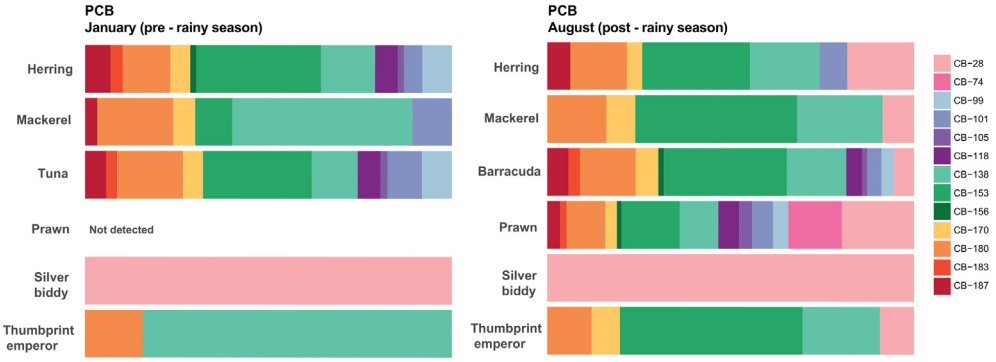


Fig. 3. Relative contribution of each polychlorinated biphenyl (PCB) congener to total PCB concentration in fish and prawn from Zanzibar, Tanzania in January (pre-rainy season) and August (post-rainy season) 2018.

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

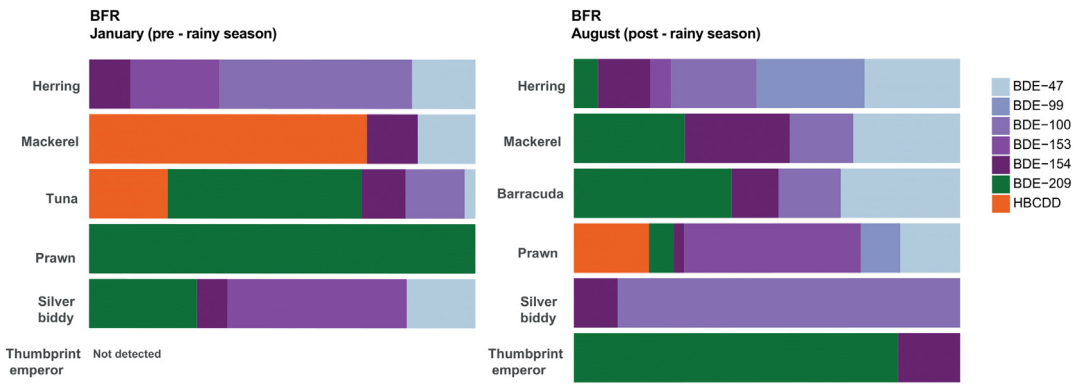


Fig. 4. Relative contribution of each compound to total brominated flame retardant (BFR) concentration in fish and prawn from Zanzibar, Tanzania in January (pre- rainy season) and August (post- rainy season) 2018.

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.

Acknowledgements

The present study was conducted as part of the University of Oslo: Life Science Convergence project ANTHROTOX, with the overall aim to combine social and natural sciences to understand and manage global toxicants. The project is funded through the University of Oslo:Life Science and the Norwegian Ministry of Education and Research. Acknowledgements are directed to local authorities for support and local fishermen for all help with the collection of samples, and everyone involved in fish dissection and field work. Authors thank Zhanna Tairova, Aarhus University Institute of Bioscience, State university of Zanzibar and the "Building Stronger Universities programme Phase III project", Aarhus University's Work Package 2 "Marine and Coastal Ecosystem Health and Services" (MaCES), for facilitating the field campaign in January. Authors thank employees and colleagues at the Institute of Marine Science on Zanzibar for the hospitality and assistance during field work in August. Thank you to Mahin Karimi, Mariska Hoorweg and Gabrielle Haddad-Weiser at the Miljøtoks Lab at NMBU for assistance with the analyses of organic contaminants.

Appendix A. Supplementary data

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

References

Abbasi, G., Li, L., Breivik, K., 2019. Global historical stocks and emissions of PBDEs. Environmental Science & Technology. https://doi.org/10.1021/acs.est.8b07032.

Asante, K.A., Adu-Kumi, S., Nakahiro, K., Takahashi, S., Isobe, T., Sudaryanto, A., Devanathan, G., Clarke, E., Ansa-Asare, O.D., Dapaah-Siakwan, S., 2011. Human exposure to PCBs, PBDEs and HBCDs in Ghana: temporal variation, sources of exposure

and estimation of daily intakes by infants. Environ. Int. 37 (5), 921–928. https://doi.org/10.1016/j.envint.2011.03.011.

Asante, K.A., Takahashi, S., Itai, T., Isobe, T., Devanathan, G., Muto, M., Agyakwah, S.K., Adu-Kumi, S., Subramanian, A., Tanabe, S., 2013. Occurrence of halogenated contaminants in inland and coastal fish from Ghana: levels, dietary exposure assessment and human health implications. Ecotoxicol. Environ. Saf. 94, 123–130. https://doi.org/10.1016/j.ecoenv.2013.05.008.

Borgå, K., Fisk, A.T., Hoekstra, P.F., Muir, D.C., 2004. Biological and chemical factors of importance in the bioaccumulation and trophic transfer of persistent organochlorine contaminants in arctic marine food webs. Environ. Toxicol. Chem. 23 (10), 2367–2385. https://doi.org/10.1897/03-518.

Breivik, K., Sweetman, A., Pacyna, J.M., Jones, K.C., 2002. Towards a global historical emission inventory for selected PCB congeners - a mass balance approach: 1. Global production and consumption. Sci. Total Environ. 290 (1–3), 181–198. https://doi.org/10.1016/S0048-9697(01)01075-0.

Breivik, K., Armitage, J.M., Wania, F., Sweetman, A.J., Jones, K.C., 2015. Tracking the global distribution of persistent organic pollutants accounting for e-waste exports to developing regions. Environmental Science & Technology 50 (2), 798–805. https://doi.org/10.1021/acs.est.5b04226.

Castro, T.F., Yoshida, T., 1971. Degradation of organochlorine insecticides in flooded soils in the Philippines. J. Agric. Food Chem. 19 (6), 1168–1170. https://doi.org/10.1021/jf60178a041.

Cheung, K., Zheng, J., Leung, H., Wong, M.H., 2008. Exposure to polybrominated diphenyl ethers associated with consumption of marine and freshwater fish in Hong Kong. Chemosphere 70 (9), 1707–1720. https://doi.org/10.1016/j.chemosphere.2007.07.043.

Connor, M.S., Davis, J.A., Leatherbarrow, J., Greenfield, B.K., Gunther, A., Hardin, D., Mumley, T., Oram, J.J., Werme, C., 2007. The slow recovery of San Francisco Bay from the legacy of organochlorine pesticides. Environ. Res. 105 (1), 87–100. https://doi.org/10.1016/j.envres.2006.07.001.

Darnerud, P., Atuma, S., Aune, M., Bjerselius, R., Glynn, A., Grawé, K.P., Becker, W., 2006. Dietary intake estimations of organohalogen contaminants (dioxins, PCB, PBDE and chlorinated pesticides, eg DDT) based on Swedish market basket data. Food Chem. Toxicol. 44 (9), 1597–1606. https://doi.org/10.1016/j.fct.2006.03.011.

De Wit, C.A., 2002. An overview of brominated flame retardants in the environment. Chemosphere 46 (5), 583–624. https://doi.org/10.1016/S0045-6535(01)00225-9.

Djien Liem, A., Furst, P., Rappe, C., 2000. Exposure of populations to dioxins and related compounds. Food Additives & Contaminants 17 (4), 241–259. https://doi.org/10.1080/026520300283324.

Elibariki, R., Maguta, M.M., 2017. Status of pesticides pollution in Tanzania–a review. Chemosphere 178, 154–164. https://doi.org/10.1016/j.chemosphere.2017.03.036.

Feidi, I., 2005. The Fisheries of Zanzibar: Potential for New Investments.

Fu, C.-T., Wu, S.-C., 2006. Seasonal variation of the distribution of PCBs in sediments and biota in a PCB-contaminated estuary. Chemosphere 62 (11), 1786–1794. https://doi.org/10.1016/j.chemosphere.2005.07.034.

Gerber, R., Wepener, V., Smit, N., 2015. Application of multivariate statistics and toxicity indices to evaluate the water quality suitability for fish of three rivers in the Kruger National Park, South Africa. Afr. J. Aquat. Sci. 40 (3), 247–259. https://doi.org/10.2989/16085914.2015.1073139.

Gioia, R., Eckhardt, S., Breivik, K., Jaward, F.M., Prieto, A., Nizzetto, L., Jones, K.C., 2011. Evidence for major emissions of PCBs in the West African region. Environmental Science & Technology 45 (4), 1349–1355 doi:0.1021/es1025239.

Gioia, R., Akindele, A.J., Adebusoye, S.A., Asante, K.A., Tanabe, S., Buekens, A., Sasco, A.J., 2014. Polychlorinated biphenyls (PCBs) in Africa: a review of environmental levels. Environ. Sci. Pollut. Res. 21 (10), 6278–6289. https://doi.org/10.1007/s11356-013-1739-1.

Guo, J., Wu, F., Shen, R., Zeng, E.Y., 2010. Dietary intake and potential health risk of DDTs and PBDEs via seafood consumption in South China. Ecotoxicol. Environ. Saf. 73 (7), 1812–1819. https://doi.org/10.1016/j.ecoenv.2010.08.009.

Henry, L., Kishimba, M., 2006. Pesticide residues in Nile tilapia (Oreochromis niloticus) and Nile perch (Lates niloticus) from Southern Lake Victoria, Tanzania. Environ. Pollut. 140 (2), 348–354. https://doi.org/10.1016/j.envpol.2005.06.029.

Huang, H.-J., Liu, S.-M., Kuo, C.-E., 2001. Anaerobic biodegradation of DDT residues (DDT, DDD, and DDE) in estuarine sediment. J. Environ. Sci. Health B 36 (3), 273–288. https://doi.org/10.1081/PFC-100103569.

Jari Oksanen, F. G. B., Michael Friendly, Roeland Kindt, Pierre Legendre, Dan McGlinn, Peter R. Minchin, R. B. O'Hara, Gavin L. Simpson, Peter Solymos, M. Henry H. Stevens, Eduard Szoecs, Helene Wagner (2019). Community Ecology Package (Version 2.5-6).

Johnstone, R.W., Muhando, C.A., Francis, J., 1998. The status of the coral reefs of Zanzibar: one example of a regional predicament. Ambio, 700–707. https://www.jstor.org/stable/4314818.

Kidd, K.A., Bootsma, H.A., Hesslein, R.H., Muir, D.C., Hecky, R.E., 2001. Biomagnification of DDT through the benthic and pelagic food webs of Lake Malawi, East Africa: importance of trophic level and carbon source. Environmental Science & Technology 35 (1), 14–20. https://doi.org/10.1021/es001119a.

Kishimba, M., Henry, L., Mwevura, H., Mmochi, A.J., Mihale, M., Hellar, H., 2004. The status of pesticide pollution in Tanzania. Talanta 64 (1), 48–53. https://doi.org/10.1016/j.talanta.2003.11.047.

Kruitwagen, G., Pratap, H., Covaci, A., Bonga, S.W., 2008. Status of pollution in mangrove ecosystems along the coast of Tanzania. Ecology and toxicology of mangrove fauna in Tanzania 51. https://doi.org/10.1016/j.marpolbul.2008.02.018.

Layman, C.A., Araujo, M.S., Boucek, R., Hammerschlag-Peyer, C.M., Harrison, E., Jud, Z.R., Matich, P., Rosenblatt, A.E., Vaudo, J.J., Yeager, L.A., 2012. Applying stable isotopes to examine food-web structure: an overview of analytical tools. Biol. Rev. 87 (3), 545–562. https://doi.org/10.1111/j.1469-185X.2011.00208.x.

Lema, E., Machunda, R., Njau, K.N., 2014. Agrochemicals use in horticulture industry in Tanzania and their potential impact to water resources. International Journal of Biological and Chemical Sciences 8 (2), 831–842. https://doi.org/10.4314/ijbcs.v8i2.38.

Machiwa, J.F., 2010. Coastal marine pollution in Dar es Salaam (Tanzania) relative to recommended environmental quality targets for the Western Indian Ocean. Western Indian Ocean Journal of Marine Science 9 (1), 17–30. https://www.ajol.info/index.php/wiojms/article/view/73958.

Mackay, D., Fraser, A., 2000. Bioaccumulation of persistent organic chemicals: mechanisms and models. Environ. Pollut. 110 (3), 375–391. https://doi.org/10.1016/S0269-7491(00)00162-7.

Mahugija, J.A., Nambela, L., Mmochi, A.J., 2017. Levels and distribution of pesticide residues in soil and sediments in Eastern Lake Tanganyika environs. International Journal of Biological and Chemical Sciences 11 (5), 2537–2547. https://doi.org/10.4314/ijbcs.v11i5.46.

Mahugija, J.A., Nambela, L., Mmochi, A.J., 2018. Determination of Dichlorodiphenyltrichloroethane (DDT) and metabolites residues in fish species from eastern Lake Tanganyika. South African Journal of Chemistry 71 (1), 86–93. https://doi.org/10.17159/0379-4350/2018/v71a11.

Mdegela, R., Braathen, M., Pereka, A., Mosha, R.D., Sandvik, M., Skaare, J., 2009. Heavy metals and organochlorine residues in water, sediments, and fish in aquatic ecosystems in urban and peri-urban areas in Tanzania. Water Air Soil Pollut. 203 (1–4), 369–379. https://doi.org/10.1007/s11270-009-0019-7.

Mmochi, A. J., & Francis, J. (2003). Land based activities and sources of pollution to the marine, coastal and associated fresh water ecosystems in the Western Indian Ocean Region. http://hdl.handle.net/1834/209

Mmochi, A.J., Mberek, R.S., 1998. Trends in the types, amounts, and toxicity of pesticides used in Tanzania: efforts to control pesticide pollution in Zanzibar, Tanzania. Ambio 27 (8), 669–676. https://doi.org/10.1016/j.talanta.2003.11.047.

Müller, M., Polder, A., Brynildsrud, O., Karimi, M., Lie, E., Manyilizu, W., Mdegela, R., Mokiti, F., Murtadha, M., Nonga, H., 2017. Organochlorine pesticides (OCPs) and polychlorinated biphenyls (PCBs) in human breast milk and associated health risks to nursing infants in Northern Tanzania. Environ. Res. 154, 425–434. https://doi.org/10.1016/j.envres.2017.01.031.

Mwakalapa, E.B., Mmochi, A.J., Müller, M.H.B., Mdegela, R.H., Lyche, J.L., Polder, A., 2018. Occurrence and levels of persistent organic pollutants (POPs) in farmed and wild marine fish from Tanzania. A pilot study. Chemosphere 191, 438–449. https://doi.org/10.1016/j.chemosphere.2017.09.121.

Mwevura, H., Othman, O.C., Mhehe, G.L., 2002. Organochlorine pesticide residues in sediments and biota from the coastal area of Dar es Salaam city, Tanzania. Mar. Pollut. Bull. 45 (1−12), 262–267. https://doi.org/10.1016/S0025-326X(01)00331-9.

Mwevura, H., Bouwman, H., Kylin, H., Vogt, T., Issa, M.A., 2020. Organochlorine pesticides and polycyclic aromatic hydrocarbons in marine sediments and polychaete worms from the west coast of Unguja island, Tanzania. Regional studies in marine science 101287.

Nonga, H., Mdegela, R., Lie, E., Sandvik, M., Skaare, J., 2011. Assessment of farming practices and uses of agrochemicals in Lake Manyara basin, Tanzania. Afr. J. Agric. Res. 6 (10), 2216–2230. http://www.suaire.suanet.ac.tz:8080/xmlui/handle/123456789/1381.

OCGS. (2018). Zanzibar Statistical Abstract 2018. Retrieved from Office of the Chief Government Statistican (OCGS): http://www.ocgs.go.tz/

Oczkowski, A., Markham, E., Hanson, A., Wigand, C., 2014. Carbon stable isotopes as indicators of coastal eutrophication. Ecol. Appl. 24 (3), 457–466. https://doi.org/10.1890/13-0365.1.

Polder, A., Müller, M., Lyche, J., Mdegela, R., Nonga, H., Mabiki, F., Mbise, T., Skaare, J., Sandvik, M., Skjerve, E., 2014. Levels and patterns of persistent organic pollutants (POPs) in tilapia (*Oreochromis* sp.) from four different lakes in Tanzania: geographical differences and implications for human health. Sci. Total Environ. 488, 252–260. https://doi.org/10.1016/j.scitotenv.2014.04.085.

Polder, A., Müller, M., Brynildsrud, O., De Boer, J., Hamers, T., Kamstra, J., Lie, E., Mdegela, R., Moberg, H., Nonga, H., 2016. Dioxins, PCBs, chlorinated pesticides and brominated flame retardants in free-range chicken eggs from peri-urban areas in Arusha, Tanzania: levels and implications for human health. Sci. Total Environ. 551, 656–667. https://doi.org/10.1016/j.scitotenv.2016.02.021.

Post, D.M., Layman, C.A., Arrington, D.A., Takimoto, G., Quattrochi, J., Montana, C.G., 2007. Getting to the fat of the matter: models, methods and assumptions for dealing with lipids in stable isotope analyses. Oecologia 152 (1), 179–189. https://doi.org/10.1007/s00442-006-0630-x.

Richmond, M.D., 2011. In: Richmond, M.D. (Ed.), A Field Guide to the Seashores of Eastern Africa and the Western Indian Ocean Islands, Third ed. Sida/WIOMSA.

Rigét, F., Bignert, A., Braune, B., Dam, M., Dietz, R., Evans, M., Green, N., Gunnlaugsdóttir, H., Hoydal, K.S., Kucklick, J., 2019. Temporal trends of persistent organic pollutants in Arctic marine and freshwater biota. Sci. Total Environ. 649, 99–110. https://doi.org/10.1016/j.scitotenv.2018.08.268.

Roberts, S.C., Noyes, P.D., Gallagher, E.P., Stapleton, H.M., 2011. Species-specific differences and structure— activity relationships in the debromination of PBDE congeners in three fish species. Environmental Science & Technology 45 (5), 1999–2005. https://doi.org/10.1016/j.scitotenv.2018.08.268.

Ruus, A., Green, N.W., Maage, A., Amundsen, C.E., Schøyen, M., Skei, J., 2010. Post World War II orcharding creates present day DDT-problems in the Sørfjord (Western Norway)–a case study. Mar. Pollut. Bull. 60 (10), 1856–1861. https://doi.org/10.1016/j.marpolbul.2010.06.048.

Ruus, A., Daae, I.A., Hylland, K., 2012. Accumulation of polychlorinated biphenyls from contaminated sediment by Atlantic cod (*Gadus morhua*): direct accumulation from resuspended sediment and dietary accumulation via the polychaete Nereis virens. Environ. Toxicol. Chem. 31 (11), 2472–2481. https://doi.org/10.1002/etc.1973.

Shilla, D.A., 2016. Distribution of Pb, Cr, Cu and Zn in the marine-coastal region of Zanzibar (Tanzanian archipelago, East Africa). Chem. Ecol. 32 (8), 774–785. https://doi.org/10.1080/02757540.2016.1178727.

Sorensen, J., Lapworth, D., Nkhuwa, D., Stuart, M., Gooddy, D., Bell, R., Chirwa, M., Kabika, J., Liemisa, M., Chibesa, M., 2015. Emerging contaminants in urban groundwater sources in Africa. Water Res. 72, 51–63. https://doi.org/10.1016/j.watres.2014.08.002.

Sparks, T.H., Scott, W.A., Clarke, R.T., 1999. Traditional multivariate techniques: potential for use in ecotoxicology. Environmental Toxicology and Chemistry: An International Journal 18 (2), 128–137. https://doi.org/10.1002/etc.5620180206.

Ssebugere, P., Kiremire, B.T., Kishimba, M., Wandiga, S.O., Nyanzi, S.A., Wasswa, J., 2009. DDT and metabolites in fish from Lake Edward, Uganda. Chemosphere 76 (2), 212–215. https://doi.org/10.1016/j.chemosphere.2009.03.049.

Stapleton, H.M., Alaee, M., Letcher, R.J., Baker, J.E., 2004. Debromination of the flame retardant decabromodiphenyl ether by juvenile carp (Cyprinus carpio) following dietary exposure. Environmental Science & Technology 38, 112–119. https://doi.org/10.1021/es034746j.

Stewart, A.R., Stern, G.A., Lockhart, W.L., Kidd, K.A., Salki, A.G., Stainton, M.P., Koczanski, K., Rosenberg, G.B., Savoie, D.A., Billeck, B.N., 2003. Assessing trends in organochlorine concentrations in Lake Winnipeg fish following the 1997 Red River flood. J. Great Lakes Res. 29 (2), 332–354. https://doi.org/10.1016/S0380-1330(03)70438-9.

UNEP, I. o. M. S., University of Dar es Salaam/FAO/SIDA. (1998). Overview of Land-based Sources and Activities Affecting the Marine, Coastal and Associated Freshwater Environment in the Easter African Region. Retrieved from https://www.unenvironment.org/resources/report/overview-land-based-sources-and-activities-affecting-marine-coastal-and-associated

UNEP/Nairobi Convention Secretariate, C. a. W., 2009. Regional Synthesis Report on the Status of Pollution in the Western Indian Ocean Region. Retrieved from UNEP, Nairobi, Kenya http://hdl.handle.net/20.500.11822/8749.

Verhaert, V., Covaci, A., Bouillon, S., Abrantes, K., Musibono, D., Bervoets, L., Verheyen, E., Blust, R., 2013. Baseline levels and trophic transfer of persistent organic pollutants in sediments and biota from the Congo River Basin (DR Congo). Environ. Int. 59, 290–302. https://doi.org/10.1016/j.envint.2013.05.015.

Verhaert, V., Newmark, N., D'Hollander, W., Covaci, A., Vlok, W., Wepener, V., Addo-Bediako, A., Jooste, A., Teuchies, J., Blust, R., 2017. Persistent organic pollutants in the Olifants River Basin, South Africa: bioaccumulation and trophic transfer through a subtropical aquatic food web. Sci. Total Environ. 586, 792–806. https://doi.org/10.1016/j.scitotenv.2017.02.057.

Wania, F., Mackay, D., 1993. Global fractionation and cold condensation of low volatility organochlorine compounds in polar regions. Ambio, 10–18. http://www.jstor.com/stable/4314030.

Wetengere, K., Moehl, J., Malya, R., Halwart, A., 2008. Strategic review of aquaculture extension in Tanzania. FAO Aquaculture Newsletter FAN 36–38.

Zhang, K., Wei, Y.-L., Zeng, E.Y., 2013. A review of environmental and human exposure to persistent organic pollutants in the Pearl River Delta, South China. Sci. Total Environ. 463, 1093–1110. https://doi.org/10.1016/j.scitotenv.2012.10.104.

Zhou, S., Tang, Q., Jin, M., Liu, W., Niu, L., Ye, H., 2014. Residues and chiral signatures of organochlorine pesticides in mollusks from the coastal regions of the Yangtze River Delta: source and health risk implication. Chemosphere 114, 40–50. https://doi.org/10.1016/j.chemosphere.2014.03.108.

Supplementary Information

For:

Seasonal rainfall affect occurrence of organohalogen contaminants in marine fishes and prawn from Zanzibar, Tanzania

Contains:

Table A1. Mean monthly precipitation (mm) on Unguja Island, Zanzibar. Based on "Zanzibar Statistical Abstract, 2018" (OCGS, 2018, Zanzibar Statistical Abstract 2018. Office of the Chief Government Statistician (OCGS), http://www.ocgs.go.tz/)

Year	Jan	Feb	Mar	Apr	May	June	July	Aug	Sept	Oct	Nov	Dec
2016	120.2	0.5	218.1	630.9	27.6	7.8	0.9	34.8	58.9	20	89.6	181.6
2017	0	12.5	69.9	516.5	670.6	38.5	2.5	30.1	28.8	285.7	200.2	143
2018	60.1	39.6	119.5	610.8	528.2	25.3	122.9	23.3	24.1	97.7	53.9	171

Table A2. Detection frequency (% detection) of all analytes in all samples

Analyte	Detection frequency (%)
b-HCH	13.8
g-HCH	53.8
ΣHCHs	53.8
p,p'-DDE	98.5
o,p'-DDE	0
p,p'-DDD	32.3
o,p'-DDD	6.2
p,p'-DDT	84.6
o,p'-DDT	38.5
ΣDDTs	
Oxychlordane	4.6
trans-chlordane	0
cis-chlordane	12.3
trans-nonachlor	21.5
cis-nonachlor	12.3
ΣChlordanes	21.5
Mirex	23.1
HCB	95.4
CB-28	56.9
CB-52	0
CB-74	4.6
CB-99	24.6
CB-101	41.5
CB-105	26.2
CB-118	29.2
CB-138	72.3
CB-153	64.6
CB-156	20
CB-170	61.5
CB-180	73.8
CB-183	29.2
CB-187	40
ΣPCBs	95.4
BDE-47	61.5
BDE-99	9.2
BDE-100	46.2
BDE-153	20
BDE-154	75.4
BDE-183	0
BDE-196	0
BDE-202	0
BDE-206	0
BDE-207	0
BDE-208	0
BDE-209	47.7
ΣPBDEs	90.8
HBCDD	20

Table A3. Contaminant concentrations (mean, range) in fishes and prawn from Zanzibar in January and August

Species	Season	Lipid%	ΣDDT[1)]	ΣOCP[2)]	ΣPCB[3)]	ΣPBDE[4)]	HBCDD
Herring	January	1.32	1.16	1.18	0.25	0.05	nd
(*S.gracilis*)		1.10-1.54	1.13-1.21	1.15-1.23	0.24-0.26	0.05-0.06	
	August	1.97	0.20	0.24	0.05	0.06	nd
		1.91-2.02	0.20-0.21	0.24-0.24	0.05-0.05	0.05-0.08	
Mackerel	January	1.95	0.33	0.51	0.06	0.01	0.04
(*R.kanagurta*)		1.08-3.35	0.06-0.80	0.22-0.96	0.04-0.12	0.01-0.03	0.01-0.07
	August	1.24	0.37	0.40	1.55	0.07	nd
		0.25-3.89	0.03-1.05	0.04-1.11	0.02-6.34	0.01-0.15	
Barracuda	August	1.50	0.48	0.51	0.24	0.05	nd
(*S. jello*)		0.91-2.93	0.28-0.93	0.31-0.99	0.12-0.35	0.03-0.07	
Tuna	January	3.62	0.79	0.90	0.24	0.18	0.03
(*E.affinis*)		0.91-10.3	0.30-1.62	0.32-1.78	0.05-0.60	0.04-0.58	0.02-0.05
Prawn	January	0.55	0.03	0.03	nd	0.02	nd
(*Penaeus sp.*)		0.45-0.65	0.02-0.04	0.02-0.04		0.01-0.03	
	August	0.59	1.63	1.66	0.59	0.11	0.02
		0.37-0.79	1.38-1.84	1.40-1.87	0.40-0.79	0.10-0.14	0.01-0.05
Silver biddy	January	0.89	1.01	1.02	0.01	0.06	nd
(*G.oyena*),		0.48-1.12	0.03-3.47	0.04-3.48	0.002-0.02	0.03-0.12	
	August	0.41	0.10	0.11	0.01	0.07	nd
		0.31-0.59	0.06-0.18	0.07-0.19	0.01-0.03	0.01-0.13	
Thumbprint	January	0.36	0.03	0.08	0.02	nd	nd
emperor		0.31-0.46	0.01-0.07	0.06-0.12	0.02-0.02		
(*L.harak*)	August	0.25	0.22	0.23	0.10	0.02	nd
		0.18-0.33	0.06-1.0	0.07-1.0	0.01-0.50	0.01-0.05	

2018. Concentrations are reported in ng/g wet weight (ww).

1) ΣDDTs: *p,p′, o,p′*(DDE, DDD, DDT)
2) ΣHCH, Σchlordanes, HCB
3) PCBs: CB-28, -74, -99, -101, -105, -118, -138, -153, -156, -170, -180, -183, -187
4) PBDEs: BDE-28, -47, -99, -100, -153, -154, -209

Table A4. Contaminant concentrations (mean, range (min-max) (ng/g lipid weight) in all species, January and August.

	January						August					
	S.gracilis	***R.kanagurta***	***E.affinis***	***Penaeus sp.***	***G.oyena***	***L.harak***	***S. gracilis***	***R.kanagurta***	***S.jello***	***Penaeus sp.***	***G.oyena***	***L.harak***
Lipid%	1.32 (1.10-1.54)	1.95 (1.08-3.35)	3.62 (0.91-10.3)	0.55 (0.45-0.65)	0.89 (0.48-1.12)	0.36 (0.31-0.46)	1.97 (1.91-2.02)	1.24 (0.25-3.89)	1.50 (0.91-2.93)	0.59 (0.37-0.79)	0.41 (0.31-0.59)	0.25 (0.18-0.33)
ΣHCH	nd	0.44 (0.19-1.05)	nd	nd	nd	nd	0.49 .(0.30-0.63)	0.80 (0.08-1.60)	0.92 (0.31-1.33)	nd	0.90 (0.30-0.63)	1.59 (0.80-2.81)
HCB	1.34 (0.92-2.01)	10.7 (5.34-21.5)	1.68 (1.13-2.12)	nd	0.83 (0.63-1.09)	13.6 (10.1-16.7)	1.16 (1.11-1.21)	4.01 (0.60-14.2)	1.44 (1.11-1.72)	2.51 (1.67-3.53)	1.66 (1.16-2.26)	3.00 (1.82-4.25)
ΣChlordanes	nd	nd	0.98 (0.37-1.64)	nd	nd	nd	nd	nd	0.20 (0.18-0.41)	2.70 (2.16-3.53)	nd	nd
Mirex	nd	nd	0.62 (0.18-1.15)	nd	nd	nd	nd	nd	0.25 (0.18-0.41)	nd	0.66 (0.25-1.18)	nd
ΣDDE	70.4 (61.5-79.7)	12.3 (0.36-26.2)	19.7 (7.62-30.8)	5.60 (3.92-8.76)	175 (2.21-652)	8.56 (3.15-15.7)	6.82 (6.42-7.15)	46.1 (0.61-179)	22.1 (19.4-25.2)	301 (210-414)	15.9 (9.01-25.5)	54.6 (12.1-221)
ΣDDD	7.35 (6.11-8.91)	nd	nd	nd	nd	nd	0.88 (0.73-1.11)	nd	3.41 (2.72-5.27)	12.8 (9.36-17.3)	4.31 (1.46-8.40)	12.3 (2.75-27.6)
ΣDDT	11.3 (8.82-13.8)	3.42 (2.01-4.43)	7.90 (3.50-11.5)	nd	19.1 (1.07-71.4)	nd	2.65 (2.46-2.86)	22.4 (1.38-82.3)	6.27 (3.69-9.23)	16.9 (11.4-26.2)	3.86 (1.77-7.96)	18.2 (5.43-83.5)
ΣOCPs	90.4 (77.5-104)	26.9 (14.9-36.9)	30.9 (13.4-46.5)	5.60 (3.92-8.76)	195 (4.07-725)	22.2 (17.4-32.2)	12.0 (11.4-12.3)	73.3 (3.24-277)	34.8 (31.3-41.0)	336 (237-464)	27.3 (18.1-44.6)	89.6 (26.0-337)
DDE/ ΣDDTs	0.79	0.68	0.71	1	0.80	1	0.66	0.60	0.69	0.91	0.64	0.60
CB-28	nd	nd	nd	nd	0.88 (0.36-1.82)	nd	0.43 (0.39-0.45)	11.00 (0.12-69.4)	0.98 (0.61-1.31)	22.7 (17.7-31.6)	2.80 (1.43-6.36)	3.59 (2.12-5.86)
CB-101	0.97 (0.65-1.35)	0.39 (0.19-0.60)	0.73 (0.26-1.20)	nd	nd	nd	0.18 (0.15-0.24)	nd	0.69 (0.51-0.91)	6.50 (4.68-8.38)	nd	nd
CB-118	1.21 (1.08-1.41)	nd	0.48 (0.22-0.96)	nd	nd	nd	nd	nd	0.78 (0.61-1.19)	6.61 (3.69-9.55)	nd	nd
CB-138	2.84 (2.51-3.30)	1.63 (0.95-2.11)	0.96 (0.15-2.36)	nd	nd	4.78 (3.55-5.66)	0.44 (0.44-0.45)	29.9 (0.08-148)	2.89 (1.92-4.78)	12.1 (10.6-15.1)	nd	8.12 (0.91-35.2)
CB-153	6.58 (5.91-7.38)	0.33 (0.09-1.09)	2.29 (1.0-4.51)	nd	nd	nd	0.69 (0.64-0.72)	56.8 (0.23-265)	5.98 (3.74-12.0)	18.4 (15.8-23.3)	nd	19.2 (0.8396.6)
CB-180	2.51 (2.29-2.82)	0.69 (0.36-0.86)	1.38 (0.55-2.67)	nd	nd	0.90 (0.43-1.52)	0.36 (0.39-0.40)	20.8 (0.08-100)	2.70 (1.60-4.59)	12.2 (9.88-15.1)	nd	4.68 (0.30-17.9)
Σ_{13} PCBs	19.3 (16.8-22.7)	3.34 (1.93-4.43)	7.71 (3.28-15.6)	nd	0.88 (0.36-1.82)	5.68 (3.97-7.18)	2.33 (2.29-2.43)	129 (0.56-562)	17.8 (12.1-31.6)	116 (87.4-160)	2.80 (1.43-6.36)	38.6 (5.76-167)
BDE-47	0.67 (0.58-0.90)	0.42 (0.24-0.64)	0.26 (0.09-0.54)	nd	1.34 (0.62-1.67)	nd	0.81 (0.72-0.95)	1.86 (0.55-5.14)	1.27 (0.85-1.74)	4.24 (3.37-5.09)	nd	nd
BDE-99	nd	nd	nd	nd	nd	nd	0.92 (0.73-1.21)	nd	nd	2.81 (2.70-2.98)	nd	nd
BDE-100	2.04 (1.75-2.33)	nd	1.42 (0.37-2.17)	nd	nd	nd	0.72 (0.44-1.16)	1.11 (0.43-2.40)	0.66 (0.20-1.10)	nd	16.6 (0.93-27.4)	nd
BDE-153	0.94 (0.82-1.08)	nd	nd	nd	3.50 (0.80-9.31)	nd	0.18 (0.15-0.25)	nd	nd	12.6 (7.98-16.5)	nd	nd
BDE-154	0.44 (0.39-0.45)	0.37 (0.09-0.77)	1.06 (0.49-1.68)	nd	0.60 (0.22-1.04)	nd	0.44 (0.39-0.8)	1.83 (0.42-4.0)	0.50 (0.31-0.61)	0.71 (0.64-0.81)	2.13 (0.25-7.65)	1.25 (0.61-1.67)
BDE-209	nd	nd	4.64 (0.17-21.6)	4.15 (1.68-6.85)	2.10 (1.04-4.04)	nd	0.21 (0.10-0.42)	1.93 (0.27-3.94)	1.68 (0.71-3.37)	1.78 (1.09-2.73)	nd	6.55 (2.03-14.6)
Σ_8PBDEs	4.09 (3.70-4.50)	0.79 (0.33-1.14)	7.38 (1.11-23.8)	4.15 (1.68-6.85)	7.54 (3.06-12.2)	nd	3.28 (2.65-4.43)	6.73 (2.61-9.94)	4.10 (2.32-6.31)	22.1 (17.2-27.8)	18.8 (1.38-34.4)	7.80 (3.63-16.0)
HBCDD	nd	2.05 (1.08-4.72)	1.88 (0.34-5.98)	nd	nd	nd	nd	nd	nd	5.33 (1.53-10.2)	nd	nd
Σ(PCB/ PBDE+PCB)	0.83	0.82	0.58	0	0.13	1	0.42	0.75	0.81	0.84	0.24	0.65

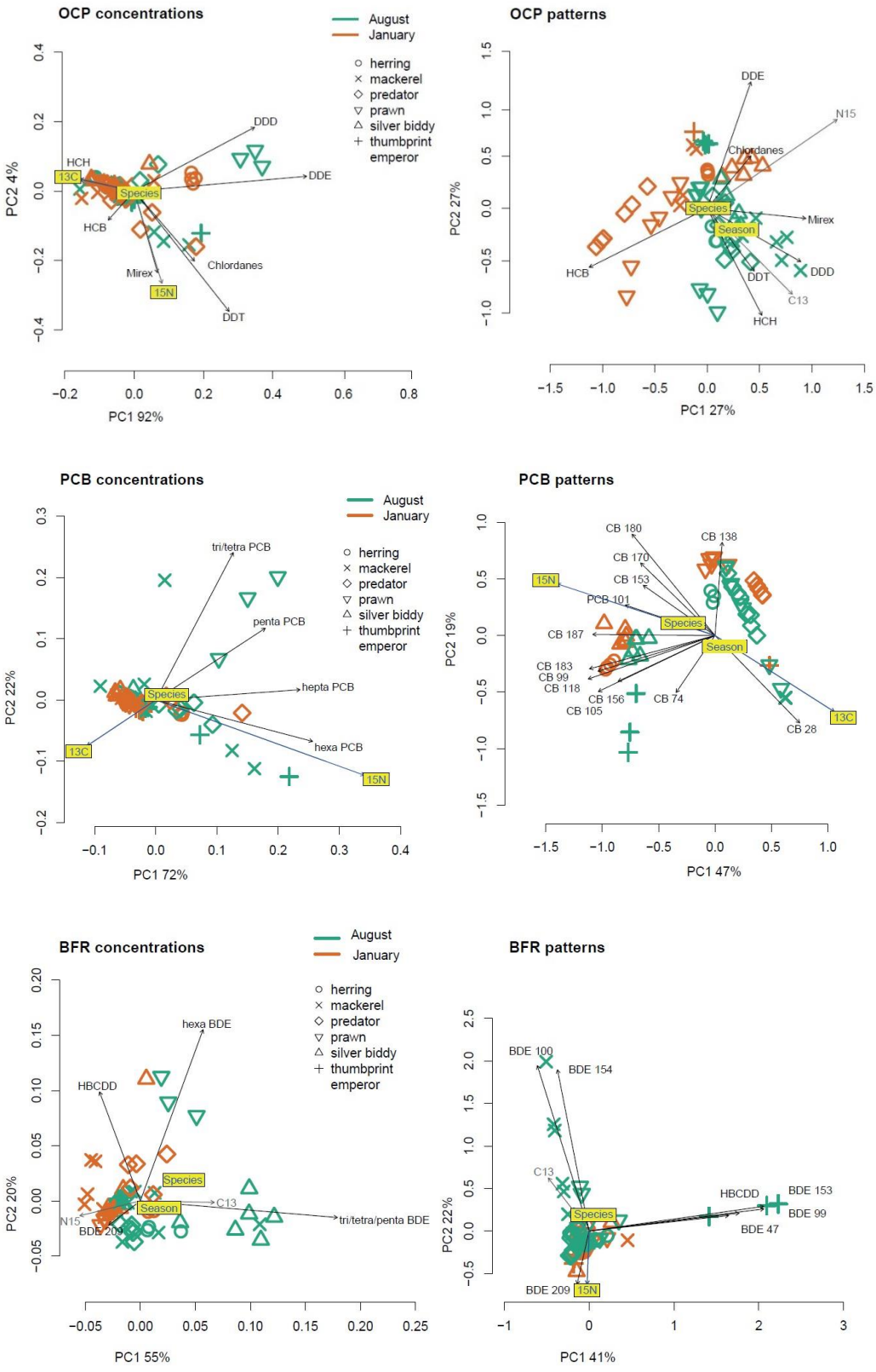


Figure A1. PCA triplots of contaminant concentrations and patterns. Lipid is included as covariate for concentrations. Patterns are relative contribution of each contaminant to its respective contaminant group. Explanatory variables are passively displayed, and significant explanatory variables are indicated with yellow boxes. Outliers are removed due to high loadings in the PCA: One outlier removed for OCP concentrations, two outliers removed for PCB conc, and one outlier removed for BFR concentrations.

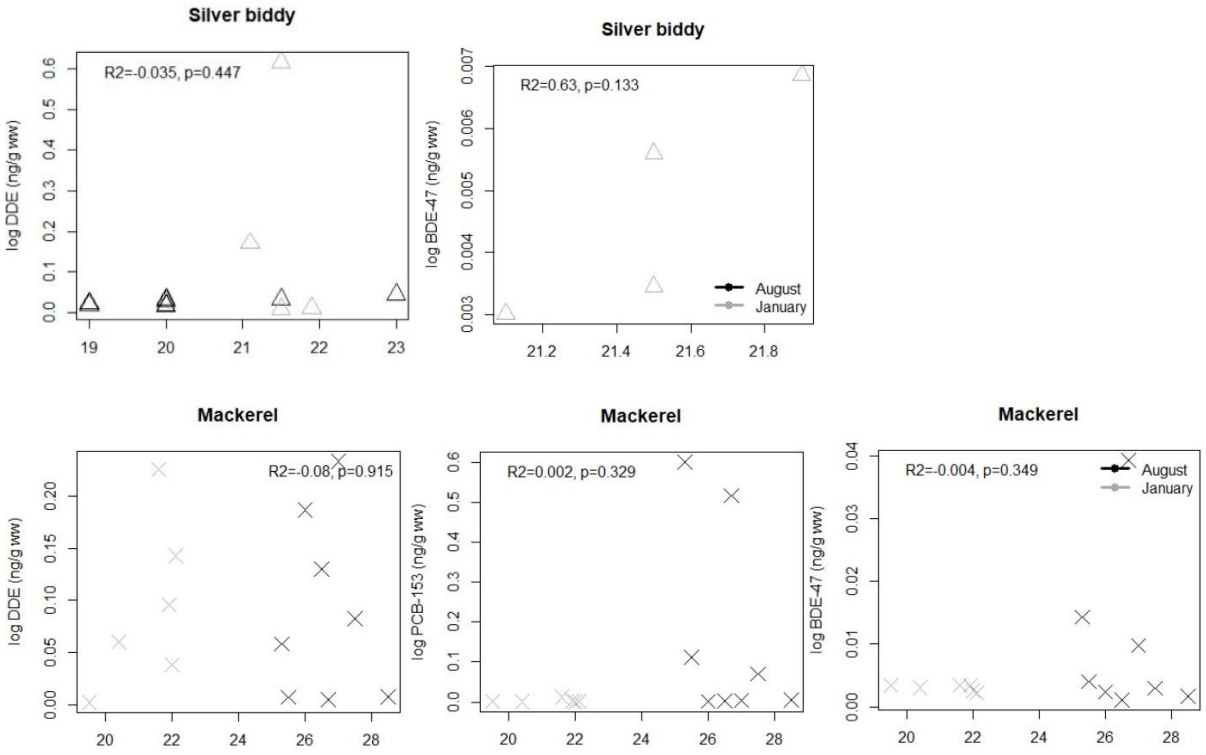


Figure A2. Linear regression plots showing the relationship between fish size (shown here as total weight) and selected POPs (log10 transformed, ng/g wet weight) for the two species where fish size was different between seasons (mackerel and silver biddy). Black symbols indicate fish collected in August, and grey symbols indicate fish collected in January. No significant interaction term for weight and season was found. PCB-153 was not detected in silver biddy in January or August, and BDE-47 was not detected in silver biddy in August.

Paper II



II

Environmental Toxicology and Chemistry—Volume 41, Number 2—pp. 321–333, 2022
Received: 31 May 2021 | Revised: 9 November 2021 | Accepted: 10 November 2021

Environmental Toxicology

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

Ane Haarr,[a] Eliezer B. Mwakalapa,[b] Jan L. Lyche,[c] Aviti J. Mmochi,[d] Anuschka Polder,[c] Anders Ruus,[a,e] and Katrine Borgå[a,f,*]

[a]Department of Biosciences, University of Oslo, Oslo, Norway
[b]Department of Natural Sciences, Mbeya University of Science and Technology, Mbeya, Tanzania
[c]Department of Paraclinical Sciences, Norwegian University of Life Sciences, Ås, Norway
[d]Institute of Marine Sciences, University of Dar es Salaam, Zanzibar, Tanzania
[e]Norwegian Institute for Water Research, Oslo, Norway
[f]Center for Biogeochemistry in the Anthropocene, University of Oslo, Oslo, Norway

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

This article includes online-only Supporting information.

* Address correspondence to katrine.borga@ibv.uio.no
Published online xxxx in Wiley Online Library (wileyonlinelibrary.com).
DOI: 10.1002/etc.5254

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

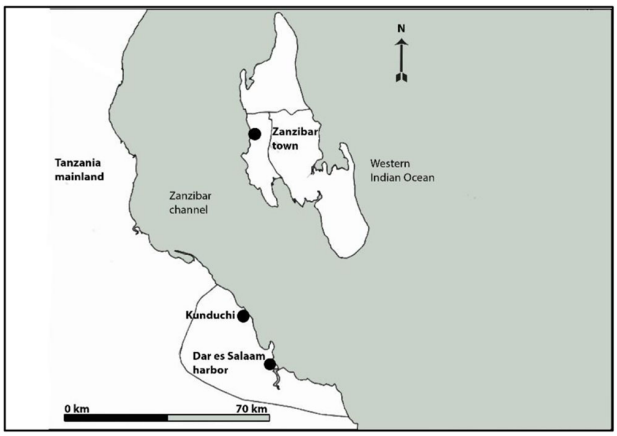


FIGURE 1: Study area and fish marked locations at mainland Tanzania (Dar es Salaam harbor and Kunduchi) and on Unguja Island, Zanzibar.

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, *Spratelloides gracilis* (dagaa lumbunga)	Low, pelagic Fish	DAR	1[a]	–	100[a]	−17.9	7.94	0.29
		KUN	1[a]	–	100[a]	−17.9	8.13	3.3
		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, *Rastrelliger kanagurta* (kibua)	Medium, pelagic Fish	DAR	6	29.5	267	−17.3	11.7	3.47
				27.0–31.0	197–297	−(17.7–16.9)	11.2–11.9	3.3–3.7
		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, *Spyraena jello* (mzia)	High, pelagic Fish	DAR	–	–	–	–	–	–
		KUN	6	70.8	1470	−15.8	13.6	3.1
				59.0–88.0	835–2300	−(16.2–15.4)	13.2–14.5	3.1–3.2
		ZNZ[b]	–	–	–	–	–	–
Mackerel tuna, *Euthynnus affinis* (jodari)	High, pelagic Fish	DAR	–	–	–	–	–	–
		KUN	–	–	–	–	–	–
		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, *Penaeus* sp. (kamba)	Low, demersal (scavenging)	DAR	1[a]	–	100[a]	−14.5	13.5	3.2
		KUN	1[a]	–	100[a]	−17.0	13.7	3.3
		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, *Gerres oyena* (chaa)	Low/medium, demersal Fish	DAR	5	19.4	105	−14.1	14.0	2.9
				17.0–21.0	69.1–130	−(14.6–13.6)	7.71–16.1	0.29–4.1
		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, *Lethrinus harak* (changu)	Low/medium, demersal Fish	DAR	6	23.8	224	−12.9	14.0	3.2
				22.0–25.5	172–273	−(14.1–11.9)	8.88–17.0	3.1–3.4
		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, *Epinephelus fasciatus*	High, demersal Fish	DAR	4	24.8	243	−13.8	14.8	3.2
				17.0–33.0	73.7–499	−(14.9 to 12.9)	13.1–16.9	3.2–3.3
		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, *Spratelloides gracilis* (dagaa lumbunga)	DAR	3[d]	0.97 (0.98) 0.91–1.01	22.2 (22.5) 21.0–23.2	1.36 (1.35) 1.29–1.44	0.50 (0.49) 0.40–0.61	21.3 (20.8) 20.0–23.2	2.13 (1.58) 1.47–3.33	nd
	KUN	3[d]	1.31 (1.3) 1.29–1.33	53.1 (52.3) 51.3–55.7	1.17 (1.16) 1.15–1.19	0.28 (0.29) 0.20–0.35	45.9 (45.8) 47.1–47.7	10.7 (10.9) 10.0–11.3	nd
	ZNZ[e]	4[d]	1.32 (1.32) 1.10–1.54	89.0 (88.7) 76.5–102	1.34 (1.21) 0.92–2.01	nd	19.4 (19.0) 16.8–22.7	4.09 3.70–4.50	nd
Indian mackerel, *Rastrelliger kanagurta* (kibua)	DAR	6	1.27 (1.24) 0.74–1.88	37.2 (37.9) 32.4–41.0	1.1 (1.1) 0.9–1.35	0.26 (0.28) nd–0.41	27.0 (26.7) 20.5–33.8	2.54 (2.27) 1.84–4.19	nd
	KUN	6	1.30 (1.17) 0.78–2.24	38.0 (38.0) 32.9–42.4	1.04 (1.02) 0.98–1.15	nd	34.0 (34.9) 28.5–37.0	nd	nd
	ZNZ[e]	6	1.95 (1.75) 1.08–3.35	15.7 (13.8) 4.13–30.6	10.7 (9.18) 5.34–21.5	0.44 (0.33) 0.19–1.05	3.31 (3.33) 1.99–4.59	0.78 (0.83) (0.33–1.14)	1.69 (1.64) nd–4.72
Pickhandle barracuda, *Spyraena jello* (mzia)	DAR	–	–	–	–	–	–	–	–
	KUN	6	0.37 (0.35) 0.24–0.59	47.1 (37.2) 15.8–104	2.61 (2.57) 1.80–3.60	1.16 (1.03) 0.75–1.77	44.1 (37.0) (12.1–99.1)	1.21 (1.14) 0.78–1.73	nd
	ZNZ[e]	–	–	–	–	–	–	–	–
Mackerel tuna, *Euthynnus affinis* (jodari)	DAR	–	–	–	–	–	–	–	–
	KUN	–	–	–	–	–	–	–	–
	ZNZ[e]	6	3.62 (2.81) 0.91–10.3	27.5 (30.4) 11.1–42.3	1.68 (1.72) 1.13-2.12	nd	7.05 (6.35) 3.03–14.1	3.37 (3.37) 1.1–5.82	1.88 (1.02) 0.34–5.98
Prawn, *Penaeus* sp. (kamba)	DAR	3[d]	0.78 (0.78) 0.75–0.81	104 (105) 99.0–107	1.61 (0.62) 1.51–1.69	0.48 (0.51) 0.36–0.58	127 (126) 126–129	9.06 (9.06) 8.81–9.31	nd
	KUN	3[d]	1.01 (1.01) 0.53–1.49	47.1 (53.7) 29.4–58.3	2.0 (1.96) 1.33–2.73	1.80 (1.08) 0.33–4.0	51.5 (53.1) 44.9–56.4	4.61 (5.09) 3.27–5.47	nd
	ZNZ[e]	3[d]	0.55 (0.56) 0.45–0.65	5.61 (4.13) 3.92–8.77	nd	nd	nd	4.15 (3.9) 1.68–6.85	nd
Silver biddy, *Gerres oyena* (chaa)	DAR	5	1.28 (1.27) 0.35–1.85	417 (501) 14.7–615	0.95 (0.93) 0.87–1.05	0.81 (0.75) 0.57–1.32	671 (544) 5.17–1160	8.1 (7.42) 3.95–11.1	9.12 (6.42) nd–19.6
	KUN	–	–	–	–	–	–	–	–
	ZNZ[e]	4	0.87 (0.94) 0.48–1.12	194 (24.6) 3.35–724	0.83 (0.81) 0.63–1.09	nd	0.79 (0.67) nd–1.82	7.49 (7.47) 3.06–11.9	nd
Thumbprint emperor, *Lethrinus harak* (changu)	DAR	6	0.81 (0.60) 0.33–1.50	73.4 (78.2) 7.87–117	0.80 (0.85) 0.62–0.93	0.47 (0.38) nd–1.0	50.6 (54.5) 7.7–80.7	0.77 (0.43) 0.34–1.33	nd
	KUN	6	0.63 (0.48) 0.28–1.26	19.1 (12.4) nd–63.8	0.54 (0.67) nd–0.73	nd	20.5 (12.6) 4.37–65.8	nd	nd
	ZNZ[e]	6	0.36 (0.34) 0.31–0.46	8.04 (6.56) nd–15.7	13.6 (14) 10.1–16.8	nd	5.68 (5.84) 3.97–7.17	nd	nd
Whitespotted grouper, *Epinephelus fasciatus*	DAR	4	0.55 (0.47) 0.27–1.04	276 (222) 168–439	2.07 (1.60) 1.08–3.93	0.87 (0.72) nd–1.79	274 (275) 167–367	13.0 (12.4) 2–29.5	nd
	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.,

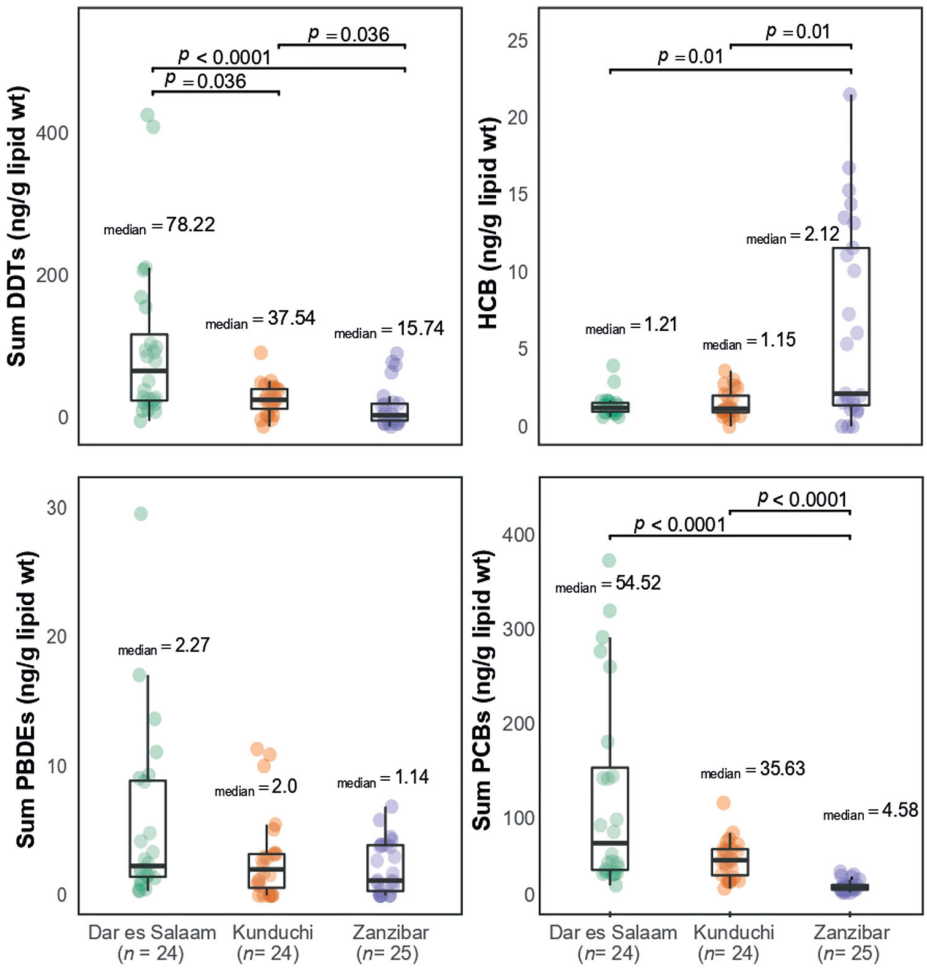


FIGURE 2: Contaminant concentrations in fishes and prawns collected from fish markets at the Dar es Salaam harbor, Kunduchi, and Zanzibar. Significant spatial difference is estimated using the Kruskal-Wallis test, followed by Dunn's pairwise comparisons. *p* values are Holm-corrected and indicate significant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

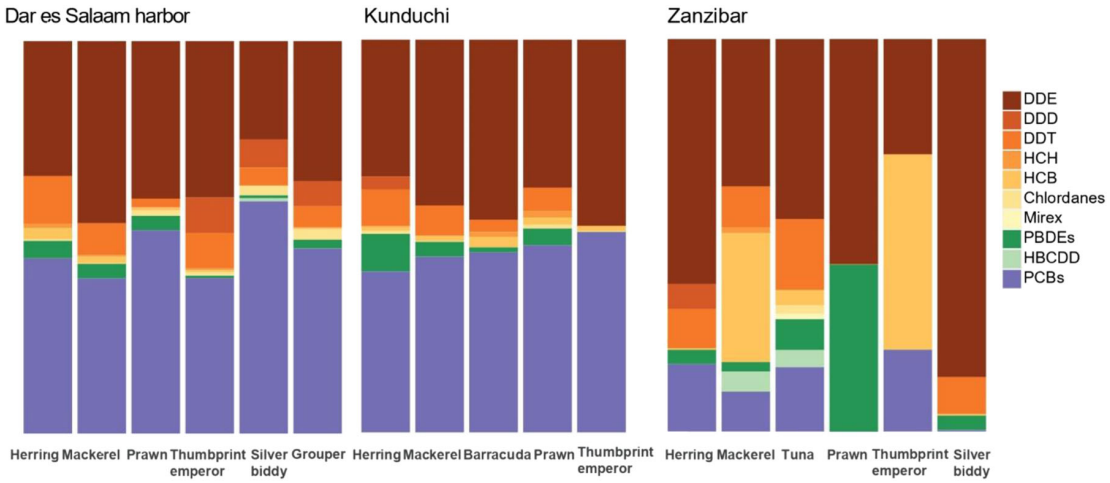


FIGURE 3: Relative contribution of each contaminant group to total contaminant load in marine fishes and prawns collected from fish markets at the Dar es Salaam harbor, Kunduchi, and Zanzibar. DDE = dichlorodiphenyldichloroethylene; DDD = dichlorodiphenyldichlorethane; DDT = dichlorodiphenyltrichloroethane; 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, $\chi^2 = 42.9$, $p = 4.9e\text{-}10$; 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.

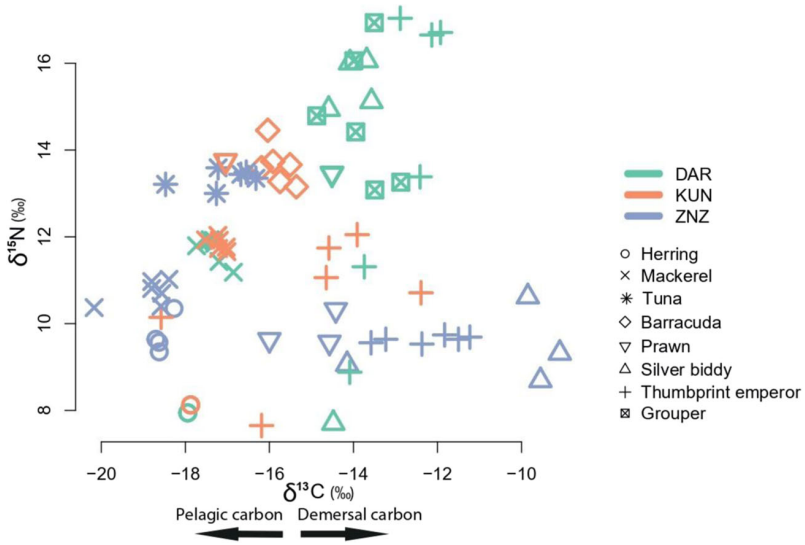


FIGURE 4: Stable isotopes of carbon and nitrogen in various marine species collected from fish markets at the Dar es Salaam harbor, Kunduchi, and Zanzibar. DAR = Dar es Salaam harbor; KUN = Kunduchi; ZNZ = Zanzibar.

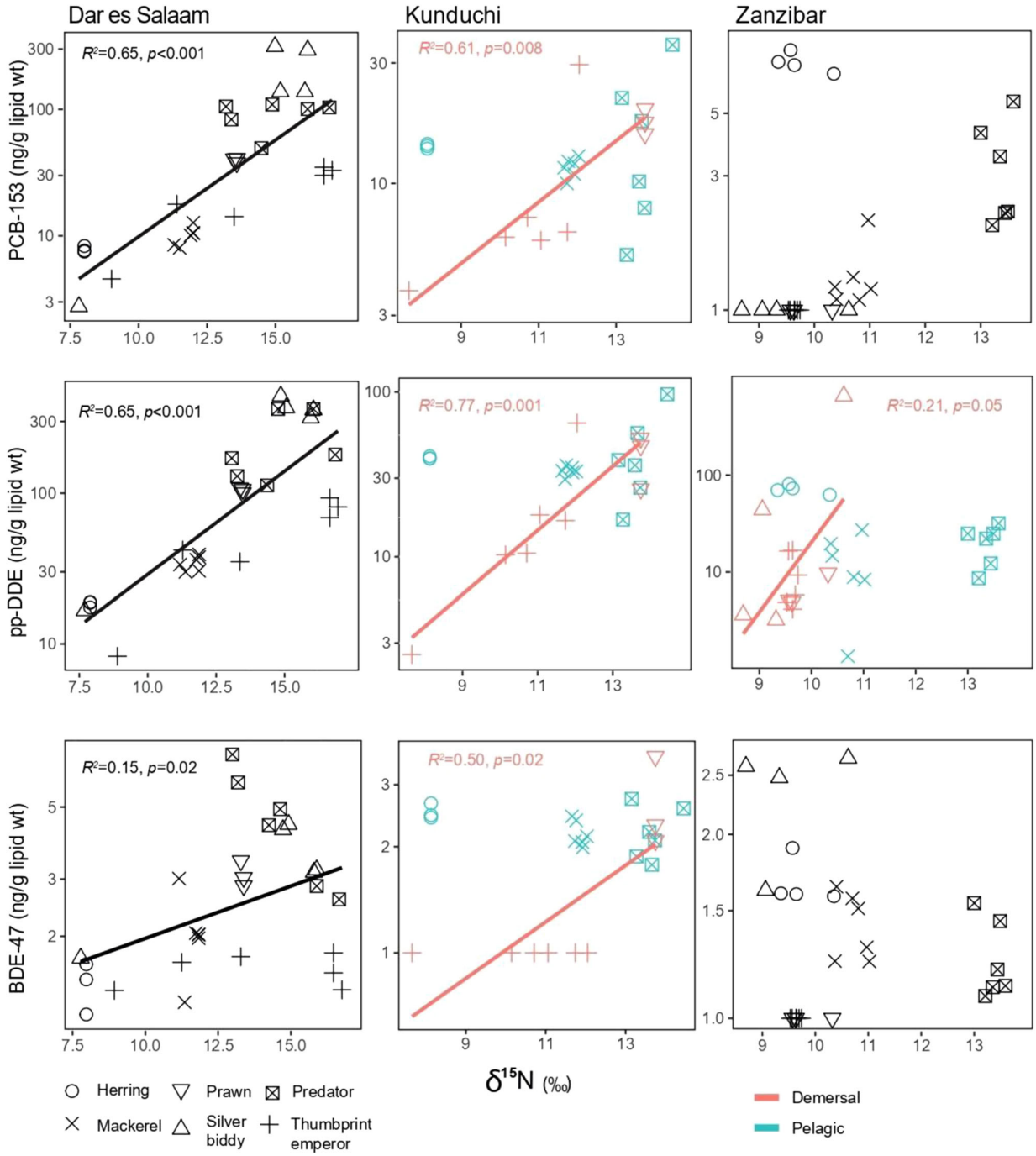


FIGURE 5: Associations between contaminants (PCB-153, pp-DDE, and BDE-47) and $\delta^{15}N$ in marine fishes and prawns collected from fish markets at the Dar es Salaam harbor, Kunduchi, and Zanzibar. Red and blue indicate significant interaction between habitat (pelagic/demersal) and $\delta^{15}N$, 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 $\delta^{15}N$ 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 $\delta^{15}N$; and this association did not differ between habitats. In Kunduchi, a positive association between contaminants and $\delta^{15}N$ was found for the demersal system but not the pelagic system. On Zanzibar, a positive association between *p, p*′-DDE and $\delta^{15}N$ 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 $\delta^{15}N$ 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 $\delta^{15}N$ values reflects individual feeding behavior or differences in baseline $\delta^{15}N$ values among locations. In general, contaminant concentrations were positively associated with $\delta^{15}N$ 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.

Acknowledgment—The present study was conducted as part of the University of Oslo: Life Science Convergence project ANTHROTOX, with the overall aim to combine social and natural sciences to understand and manage global toxicants. The project is funded through the University of Oslo: Life Science and the Norwegian Ministry of Education and Research. Acknowledgments are directed to local Tanzanian authorities for support, local fishermen for all help with sample collection, and everyone involved in fieldwork and fish dissection, especially the Brown-Mwakalapa household. We thank Z. Tairova and M. Mørk Larsen, Aarhus University Institute of Bioscience, State University of Zanzibar, and the "Building Stronger Universities programme Phase III project," Aarhus University's Work Package 2 "Marine and Coastal Ecosystem Health and Services," for facilitating the field campaign on Zanzibar. Thank you to M. Karimi, M. Hoorweg, and G. Haddad-Weiser at the Laboratory of Environmental Toxicology at the Norwegian University of Life Sciences for assistance with the analyses of organic contaminants.

Author Contributions Statement—Ane Haarr: conceptualization, investigation, writing–original draft preparation. Eliezer B. Mwakalapa: investigation, writing–review and editing. Jan L. Lyche: supervision, resources–analyses of organic contaminants, writing–review and editing. Aviti J. Mmochi: Resources–provision of laboratory facilities, writing–review and editing. Anuschka Polder: conceptualization, writing–review and editing. Anders Ruus: supervision, writing–review and editing. Katrine Borgå: supervision, conceptualization, writing–review and editing, project administration.

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

REFERENCES

Abbasi, G., Li, L., & Breivik, K. (2019). Global historical stocks and emissions of PBDEs. *Environmental Science & Technology*, *53*(11), 6330–6340. https://doi.org/10.1021/acs.est.8b07032

Adu-Kumi, S., Kawano, M., Shiki, Y., Yeboah, P., Carboo, D., Pwamang, J., Morita, M., & Suzuki, N. (2010). Organochlorine pesticides (OCPs), dioxin-like polychlorinated biphenyls (dl-PCBs), polychlorinated dibenzo-p-dioxins and polychlorinated dibenzo furans (PCDD/Fs) in edible fish from Lake Volta, Lake Bosumtwi and Weija Lake in Ghana. *Chemosphere*, *81*(6), 675–684. https://doi.org/10.1016/j.chemosphere.2010.08.018

Asante, K. A., Adu-Kumi, S., Nakahiro, K., Takahashi, S., Isobe, T., Sudaryanto, A., Devanathan, G., Clarke, E., Ansa-Asare, O. D., & Dapaah-Siakwan, S. (2010). Human exposure to PCBs, PBDEs and HBCDs in Ghana: Temporal variation, sources of exposure and estimation of daily intakes by infants. *Environment International*, *37*(5), 921–928. https://doi.org/10.1016/j.envint.2011.03.011

Asante, K. A., Takahashi, S., Itai, T., Isobe, T., Devanathan, G., Muto, M., Agyakwah, S. K., Adu-Kumi, S., Subramanian, A., & Tanabe, S. (2013). Occurrence of halogenated contaminants in inland and coastal fish from Ghana: Levels, dietary exposure assessment and human health implications. *Ecotoxicology and Environmental Safety*, *94*, 123–130. https://doi.org/10.1016/j.ecoenv.2013.05.008

Breivik, K., Armitage, J. M., Wania, F., Sweetman, A. J., & Jones, K. C. (2015). Tracking the global distribution of persistent organic pollutants accounting for e-waste exports to developing regions. *Environmental Science & Technology*, *50*(2), 798–805. https://doi.org/10.1021/acs.est.5b04226

Breivik, K., Gioia, R., Chakraborty, P., Zhang, G., & Jones, K. C. (2011). Are reductions in industrial organic contaminants emissions in rich countries achieved partly by export of toxic wastes? *Environmental Science & Technology*, *45*(21), 9154–9160. https://doi.org/10.1021/es202320c

Breivik, K., Sweetman, A., Pacyna, J. M., & Jones, K. C. (2002). Towards a global historical emission inventory for selected PCB congeners—A mass balance approach: 1. Global production and consumption. *Science of the Total Environment*, *290*(1–3), 181–198. https://doi.org/10.1016/S0048-9697(01)01075-0

Bustnes, J. O., Borgå, K., Dempster, T., Lie, E., Nygård, T., & Uglem, I. (2012). Latitudinal distribution of persistent organic pollutants in pelagic and demersal marine fish on the Norwegian coast. *Environmental Science & Technology*, *46*(14), 7836–7843. https://doi.org/10.1021/es301191t

European Commission (2005). Regulation (EC) No 396/2005 of the European Parliament and of the Council of 23rd February 2005 on maximum residue levels of pesticides in or on food and feed of plant and animal origin and amending Council Directive 91/414/EEC.

European Commission. (2011). Commission Regulation (EC) No 1259/2011 of 2 December 2011 amending Regulation (EC) No 1881/2006 as regards maximum levels for dioxins, dioxin-like PCBs and non dioxin-like PCBs in foodstuffs.

European Commission (2013). Directive 2013/39/EC of the European Parliament and of the Council of 12 August 2013 amending Directives 2000/60/EC and 2008/105/EC as regards priority substances in the field of water policy

Froese, R., & Pauly, D. (2020). FishBase. www.fishbase.org

Guo, L., Qiu, Y., Zhang, G., Zheng, G. J., Lam, P. K., & Li, X. (2008). Levels and bioaccumulation of organochlorine pesticides (OCPs) and polybrominated diphenyl ethers (PBDEs) in fishes from the Pearl River estuary and Daya Bay, South China. *Environmental Pollution*, *152*(3), 604–611. https://doi.org/10.1016/j.envpol.2007.06.067

Guzzo, M. M., Haffner, G. D., Sorge, S., Rush, S. A., & Fisk, A. T. (2011). Spatial and temporal variabilities of $\delta^{13}C$ and $\delta^{15}N$ within lower trophic levels of a large lake: Implications for estimating trophic relationships of consumers. *Hydrobiologia*, *675*(1), 41–53. https://doi.org/10.1007/s10750-011-0794-1

Haarr, A., Mwakalapa, E. B., Mmochi, A. J., Lyche, J. L., Ruus, A., Othman, H., Larsen, M. M., & Borgå, K. (2021). Seasonal rainfall affects occurrence of organohalogen contaminants in tropical marine fishes and prawns from Zanzibar, Tanzania. *Science of the Total Environment*, *774*, 145652. https://doi.org/10.1016/j.scitotenv.2021.145652

Jahnke, M., Gullström, M., Larsson, J., Asplund, M. E., Mgeleka, S., Silas, M. O., Hoamby, A., Mahafina, J., & Nordlund, L. M. (2019). Population genetic structure and connectivity of the seagrass *Thalassia hemprichii* in the western Indian Ocean is influenced by predominant ocean currents. *Ecology and Evolution*, *9*(16), 8953–8964. https://doi.org/10.1002/ece3.5420

Kidd, K. A., Bootsma, H. A., Hesslein, R. H., Muir, D. C., & Hecky, R. E. (2001). Biomagnification of DDT through the benthic and pelagic food webs of Lake Malawi, East Africa: Importance of trophic level and carbon source. *Environmental Science & Technology*, *35*(1), 14–20. https://doi.org/10.1021/es001119a

Machiwa, J. F. (1992). Anthropogenic pollution in the Dar es Salaam harbour area, Tanzania. *Marine Pollution Bulletin*, *24*(11), 562–567. https://doi.org/10.1016/0025-326X(92)90709-F

Munschy, C., Vigneau, E., Bely, N., Héas-Moisan, K., Olivier, N., Pollono, C., Hollanda, S., & Bodin, N. (2020). Legacy and emerging organic contaminants: Levels and profiles in top predator fish from the western Indian Ocean in relation to their trophic ecology. *Environmental Research*, *188*, 109761. https://doi.org/10.1016/j.envres.2020.109761

Mwakalapa, E. B., Mmochi, A. J., Müller, M. H. B., Mdegela, R. H., Lyche, J. L., & Polder, A. (2018). Occurrence and levels of persistent organic pollutants (POPs) in farmed and wild marine fish from Tanzania. A pilot study. *Chemosphere*, *191*, 438–449. https://doi.org/10.1016/j.chemosphere.2017.09.121

Mwevura, H., Amir, O. A., Kishimba, M., Berggren, P., & Kylin, H. (2010). Organohalogen compounds in blubber of Indo-Pacific bottlenose dolphin (*Tursiops aduncus*) and spinner dolphin (*Stenella longirostris*) from Zanzibar, Tanzania. *Environmental Pollution*, *158*(6), 2200–2207. https://doi.org/10.1016/j.envpol.2010.02.027

Mwevura, H., Bouwman, H., Kylin, H., Vogt, T., & Issa, M. A. (2020). Organochlorine pesticides and polycyclic aromatic hydrocarbons in marine sediments and polychaete worms from the west coast of Unguja Island, Tanzania. *Regional Studies in Marine Science*, *36*, 101287. https://doi.org/10.1016/j.rsma.2020.101287

Mwevura, H., Othman, O. C., & Mhehe, G. L. (2002). Organochlorine pesticide residues in sediments and biota from the coastal area of Dar es Salaam city, Tanzania. *Marine Pollution Bulletin*, *45*(1–12), 262–267. https://doi.org/10.1016/S0025-326X(01)00331-9

Naso, B., Perrone, D., Ferrante, M. C., Bilancione, M., & Lucisano, A. (2005). Persistent organic pollutants in edible marine species from the Gulf of Naples, southern Italy. *Science of the Total Environment*, *343*(1–3), 83–95. https://doi.org/10.1016/j.scitotenv.2004.10.007

Patil, I. (2018). *ggstatsplot: "ggplot2" based plots with statistical details*. CRAN. https://cran.r-project.org/web/packages/ggstatsplot/index.html

Polder, A., Müller, M., Lyche, J., Mdegela, R., Nonga, H., Mabiki, F., Mbise, T., Skaare, J., Sandvik, M., & Skjerve, E. (2014). Levels and patterns of persistent organic pollutants (POPs) in tilapia (*Oreochromis* sp.) from four different lakes in Tanzania: Geographical differences and implications for human health. *Science of the Total Environment*, *488*, 252–260. https://doi.org/10.1016/j.scitotenv.2014.04.085

Post, D. M., Layman, C. A., Arrington, D. A., Takimoto, G., Quattrochi, J., & Montana, C. G. (2007). Getting to the fat of the matter: Models, methods and assumptions for dealing with lipids in stable isotope analyses. *Oecologia*, *152*(1), 179–189. https://doi.org/10.1007/s00442-006-0630-x

R Foundation for Statistical Computing. (2017). *R: A language and environment for statistical computing*. https://www.R-project.org/

Richmond, M. D. (Ed.). (2011). *A field guide to the seashores of eastern Africa and the western Indian Ocean islands* (3rd ed.). Sida/WIOMSA.

Shang, X., Dong, G., Zhang, H., Zhang, L., Yu, X., Li, J., Wang, X., Yue, B., Zhao, Y., & Wu, Y. (2016). Polybrominated diphenyl ethers (PBDEs) and indicator polychlorinated biphenyls (PCBs) in various marine fish from Zhoushan fishery, China. *Food Control*, *67*, 240–246. https://doi.org/10.1016/j.foodcont.2016.03.008

Shi, J., Li, Y., Liang, H., Zheng, G. J., Wu, Y., & Liu, W. (2013). OCPs and PCBs in marine edible fish and human health risk assessment in the eastern Guangdong, China. *Archives of Environmental Contamination and Toxicology*, *64*(4), 632–642. https://doi.org/10.1007/s00244-012-9862-4

Ssebugere, P., Sillanpää, M., Wang, P., Li, Y., Kiremire, B. T., Kasozi, G. N., Zhu, C., Ren, D., Zhu, N., & Zhang, H. (2014). Polychlorinated biphenyls in sediments and fish species from the Murchison Bay of Lake Victoria, Uganda. *Science of the Total Environment*, *482*, 349–357. https://doi.org/10.1016/j.scitotenv.2014.03.009

Sun, R.-X., Sun, Y., Xie, X.-D., Yang, B.-Z., Cao, L.-Y., Luo, S., Wang, Y.-Y., & Mai, B.-X. (2020). Bioaccumulation and human health risk assessment of DDT and its metabolites (DDTs) in yellowfin tuna (*Thunnus albacares*) and their prey from the South China Sea. *Marine Pollution Bulletin*, *158*, 111396. https://doi.org/10.1016/j.marpolbul.2020.111396

Tanzania Ports Authority. (2016). *Environmental impact statement for the proposed improvement of Dar es Salaam port: Phase 1 of Dar es*

Supplementary Information

For:

Spatial variation in contaminant occurrence in marine fishes and prawns from coastal Tanzania

Contains:

Table A1. Detection frequency of all analyzed chemicals

Analyte	**Detection frequency (%)**
b-HCH	9.8
g-HCH	61
p,p´-DDE	96.3
o,p´-DDE	0
p,p´-DDD	37.8
o,p´-DDD	11
p,p´-DDT	84.1
o,p´-DDT	15.9
Oxychlordane	17.1
trans-chlordane	20.7
cis-chlordane	32.9
trans-nonachlor	57.3
cis-nonachlor	40.2
Mirex	11.0
HCB	96.3
CB-28	24.4
CB-52	34.1
CB-74	17.1
CB-99	47.6
CB-101	69.5
CB-105	78,0
CB-118	75.6
CB-138	90.2
CB-153	82.9
CB-156	64.9
CB-170	82.9
CB-180	90.2
CB-183	73.2
CB-187	80.5
BDE-47	90.2
BDE-99	24.4
BDE-100	59.8
BDE-153	37.8
BDE-154	52.4
BDE-183	1.2
BDE-196	11.0
BDE-202	35.4
BDE-206	6.1
BDE-207	6.1
BDE-208	4.9
BDE-209	36.6
HBCDD	26.8

Table A2. Contaminant concentrations (mean (median), ng/g wet weight) in fishes and prawns from three different locations in mainland Tanzania and Zanzibar

Species (common name)	Location	n	Lipid%	ΣDDTs[3]	HCB	ΣPCBs[4]	ΣPBDEs[5]	HBCDD
Silver-stripe herring	DAR	3[2]	0.97 (0.05) 0.98 (0.91-1.01)	0.21 (0.005) 0.21 (0.21-0.22)	0.01 (0.01) 0.01 (0.01-0.01)	0.21 (0.005) 0.21 (0.20-0.21)	0.02 (0.01) 0.02 (0.01-0.03)	nd
Spratelloides gracilis	KUN	3[2]	1.31 (0.18) 1.29-1.33	0.69 (0.03) 0.7 (0.66-0.72)	0.02 (0) 0.2 (0.02-0.02)	0.60 (0.02) 0.59 (0.59-0.62)	0.14 (0.006) 0.14 (0.13-0.14)	nd
(dagaa lumbunga)	ZNZ[1]	4[2]	1.32 (0.18) 1.32 (1.10-1.54)	1.16 (0.04) 1.15 (1.13-1.21)	0.02 (0.004) 0.02 (0.01-0.02)	0.25 (0.01) 0.26 (0.24-0.26)	0.05 (0.003) 0.05 (0.05-0.06)	nd
Indian mackerel *Rastrelliger kanagurta*, (kibua)	DAR	6	1.27 (0.36) 1.24 (0.74-1.88)	0.47 (0.11) 0.49 (0.28-0.61)	0.01 (0.01) 0.01(0.01-0.02)	0.35 (0.13) 0.36 (0.15-0.53)	0.03 (0.004) 0.03 (0.03-0.03)	nd
	KUN	6	1.30 (0.54) 1.17 (0.78-2.24)	0.49 (0.2) 0.41 (0.3-0.83)	0.003 (0.003) 0.003 (nd-0.01)	0.44 (0.19) 0.38 (0.27-0.78)	nd	nd
	ZNZ[1]	6	1.95 (0.90) 1.75 (1.08-3.35)	0.33 (0.27) 0.25 (0.06-0.80)	0.17 (0.04) 0.16 (0.14-0.25)	0.06 (0.03) 0.06 (0.03-0.12)	0.01 (0.01) 0.01 (0.01-0.03)	0.03 (0.03) 0.04 (0.01-0.07)
Pickhandle barracuda *Spyraena jello*, (mzia)	DAR	-	-	-	-	-	-	-
	KUN	6	0.37 (0.12) 0.35 (0.24-0.59)	0.17 (0.10) 0.14 (0.06-0.34)	0.01 (0.002) 0.01 (0.01-0.02)	0.15 (0.10) 0.14 (0.05-0.32)	0.004 (0.001) 0.004 (0.002-0.01)	nd
	ZNZ[1]	-	-	-	-	-	-	
Mackerel tuna *Euthynnus affinis*, (jodari)	DAR	-	-	-	-	-	-	-
	KUN	-	-	-	-	-	-	-
	ZNZ[1]	6	3.62 (3.45) 2.81 (0.91-10.3)	0.79 (0.53) 0.67 (0.30-1.62)	0.06 (0.05) 0.05 (0.01-0.16)	0.22 (0.19) 0.18 (0.05-0.54)	0.09 (0.04) 0.08 (0.04-0.14)	0.03 (0.01) 0.03 (0.02-0.05)
Prawn *Penaeus sp.* (kamba)	DAR	3[2]	0.78 (0.03) 0.77 (0.75-0.81)	0.81 (0.04) 0.81 (0.76-0.86)	0.01 (0.002) 0.01 (0.01-0.01)	0.99 (0.05) 0.98 (0.95-1.05)	0.07 (0.01) 0.07 (0.07-0.07)	nd
	KUN	3[2]	1.01 (0.48) 1.01 (0.53-1.49)	0.47 (0.29) 0.41 (0.3-0.83)	0.02 (0.01) 0.01 (0.01-0.02)	0.50 (0.19) 0.54 (0.30-0.67)	0.04 (0.01) 0.05 (0.03-0.06)	nd
	ZNZ[1]	3[2]	0.55 (0.1) 0.56 (0.45-0.65)	0.03 (0.01) 0.03 (0.02-0.04)	nd	nd	0.02 (0.01) 0.03 (0.01-0.03)	nd
Silver biddy *Gerres oyena*, (chaa)	DAR	5	1.28 (0.6) 1.27 (0.35-1.85)	6.15 (3.6) 7.66 (0.05-9.26)	0.01 (0.005) 0.01 (0.004-0.02)	9.39 (5.64) 9.85 (0.02-14.8)	0.11 (0.07) 0.13 (0.02-0.20)	0.13 (0.14) 0.07 (nd-0.29)
	KUN	-	-	-	-	-	-	-
	ZNZ[1]	4	0.87 (0.28) 0.94 (0.48-1.12)	0.01 (1.66) 0.28 (0.03-3.47)	0.01 (0.003) 0.01(0.004-0.01)	nd	0.06 (0.04) 0.05 (0.03-0.12)	nd
Thumbprint emperor *Lethrinus harak*, (changu)	DAR	6	0.81 (0.49) 0.6 (0.33-1.50)	0.53 (0.5) 0.35 (0.13-1.51)	0.01 (0.002) 0.01 (0.003-0.01)	0.37 (0.34) 0.24 (0.12-1.05)	0.01 (0.002) 0.01 (0.002-0.01)	nd
	KUN	6	0.63 (0.38) 0.48 (0.28-1.26)	0.19 (0.31) 0.06 (nd-0.80)	0.004 (0.003) 0.003 (nd-0.01)	0.19 (0.32) 0.07 (0.01-0.83)	nd	nd
	ZNZ[1]	6	0.36 (0.06) 0.34 (0.31-0.46)	0.03 (0.03) 0.02 (nd-0.07)	0.05 (0.003) 0.05 (0.05-0.06)	0.02 (0.002) 0.02 (0.02-0.02)	nd	nd
Epinephelus fasciatus, whitespotted grouper	DAR	4	0.55 (0.27) 0.47 (0.27-1.04)	1.62 (1.41) 1.14 (0.60-4.37)	0.01 (0.002) 0.01 (0.006-0.01)	1.51 (0.91) 1.24 (0.78-3.23)	0.05 (0.02) 0.05 (0.01-0.08)	nd
	KUN	-	-	-	-	-	-	-
	ZNZ[1]	-	-	-	-	-	-	-

[1] Data presented in Haarr (2021)

[2] Pooled samples

[3] ΣDDTs: *p,p′, o,p′*(DDE, DDD, DDT)

[4] PCBs: CB-28, -52, -74, -99, -101, -105, -118, -138, -153, -156, -170, -180, -183, -187

[5] PBDEs: BDE-47, -99, -100, -153, -154, -202, -209

Table A3. Concentrations (mean (median), range (ng/g lipid weight)) of organochlorine pesticides, PCBs, PBDEs and HBCDD in muscle tissues of fishes and prawns from coastal Tanzania and Zanzibar

	Herring ***S. gracilis***	**Mackerel** ***R. kanagurta***	**Prawn** ***Penaeus sp.***	**Silver biddy** ***G. oyena,***	**T. emperor** ***L. harak,***	**Predator**	**Location**
HCH	0.50 (0.49)	0.26 (0.28)	0.48 (0.51)	0.81 (0.75)	0.47 (0.38)	0.87 (0.72)	DAR
	0.40-0.61	nd-0.41	0.36-0.58	0.57-1.32	nd-1.0	nd-1.79	
	0.28	nd	1.8 (1.1)	-	nd	1.16 (1.03)	KUN
	0.20-0.35		0.33-4.0			0.75-1.76	
	nd	0.44 (0.33)	nd	nd	nd	nd	ZNZ[1)]
		0.19-1.05					
HCB	1.36 (1.35)	1.10 (1.06)	1.61 (1.62)	0.95 (0.93)	0.79 (0.85)	2.07 (1.60)	DAR
	1.29-1.44	0.96-1.35	1.51-1.69	0.87-1.05	0.62-0.93	1.6-3.93	
	1.17 (1.16)	1.04 (1.02)	2.0 (1.96)	-	0.54 (67)	2.61 (2.57)	KUN
	1.16-1.19	0.98-1.15	1.33-2.73		nd-0.73	1.80-3.60	
	1.34 (1.21)	10.7 (9.18)	nd	0.83 (0.81)	13.6	1.68 (1.72)	ZNZ[1)]
	0.92-2.01	5.34-21.5		0.63-1.09	10.1-16.7	(.13-2.12	
Σchlordanes	0.20 (0.29)	0.14 (0.18)	3.33 (3.22)	26.8 (32.9)	1.13 (1.29)	15.6 (14.1)	DAR
	nd-0.30	nd-0.24	3.08-3.69	nd-34.9	0.13-1.71	5.0-28.5	
	0.89 (0.83)	0.17 (0.18)	1.05 (0.98)	-	nd	nd	KUN
	0.81-1.04	nd-0.27	0.53-1.64				
	nd	nd	nd	nd	nd	0.92 (0.99)	ZNZ[1)]
						nd-1.64	
Mirex	nd	nd	nd	nd	nd	nd	DAR
	nd	nd	nd	nd	nd	nd	KUN
	nd	nd	nd	nd	nd	0.62 (0.59)	ZNZ[1)]
						0.18-1.15	
ΣDDE	16.4 (16.8)	31.7 (32)	98.5 (99.4)	284 (339)	50.8 (51.5)	208 (166)	DAR
	15.5-16.9	27.4-36.2	94.4-102	14.7-419	6.84-87.2	106-343	
	39 (38.8)	32.1 (32.3)	40.8 (46.0)	-	19.1 (12.4)	44.1 (36.3)	KUN
	38.5-39.7	28.5-34.9	24.8-51.5		nd-63.8	15.8-95.9	
	70.5 (70.3)	12.3 (10.8)	5.61 (4.12)	175 (22.8)	8.56	19.7 (22.5)	ZNZ[1)]
	61.5-79.8	nd-26.2	3.92-8.76	2.22-652	3.15-15.7	7.62-30.8	
ΣDDD	nd	nd	nd	81.3 (96.2)	11.6 (11.5)	36.6 (35.5)	DAR
				nd-114	nd-18.3	30.9-45.4	
	nd	nd	nd	-	nd	nd	KUN
	7.27 (7.19)	nd	nd	nd	nd	nd	ZNZ[1)]
	5.78-8.92						
ΣDDT	5.81 (5.53)	5.55 (5.48)	5.24 (5.52)	51.7 (44.2)	11.5 (11.5)	31.5 (27.2)	DAR
	5.45-6.47	4.52-6.59	4.62-5.6	nd-92.1	1.03-19.8	(21.4-49.8)	
	10.4 (10.3)	5.82 (5.71)	6.37 (6.82)	-	nd	2.95 (2.11)	KUN
	9.44-11.4	4.42-7.53	4.61-7.62			nd-7.67	
	11.3 (11.2)	3.42 (3.54)	nd	19.1 (1.9)	nd	7.76 (7.88)	ZNZ[1)]
	8.82-13.8	2.01-4.43		1.07-71.4		3.50-11.5	
ΣOCPs	24.3 (24.4)	38.7 (39.4)	109 (110)	446 (536)	76.3 (81.1)	294 (239)	DAR
	23.0-25.5	33.6-42.4	104-113	17.1-652	8.82-120	175-470	
	55.4 (54.6)	39.2 (39.2)	52.0 (55.9)	-	19.7 (13.0)	50.9 (41.1)	KUN
	53.8-58.0	34.1-43.7	33.5-66.6		nd-64.9	18.9-108	
	90.3 (89.8)	27.0 (30.4)	5.61 (4.12)	195 (25.5)	22.2	30.7 (33.2)	ZNZ[1)]
	77.2-105	14.9-37.1	3.92-8.76	4.07-725	17.4-32.2	13.4-46.5	

[1)] Data presented in Haarr (2021)

Table A3. Continued

	Herring	Mackerel	Prawn	Silver biddy	T. emperor	Predator	Location
CB-28	nd	nd	4.68 (4.76) 4.14-5.13	13.5 (14.5) nd-24.1	nd	5.54 (4.93) nd-11.8	DAR
	nd	nd	nd	-	nd	nd	KUN
	nd	nd	nd	0.78 (0.67) nd-1.82	nd	nd	ZNZ[1)]
CB-101	1.52 (1.57) 1.40-1.58	1.29 (1.49) nd-1.83	8.41 (8.45) 7.93-8.85	71.4 (49.4) nd-131	1.86 (2.4) nd-3.37	10.9 (7.89) 5.27-25.1	DAR
	2.85 (2.78) 2.57-3.19	1.76 (1.83) 1.34-2.07	2.78 (2.72) 2.35-3.27	-	nd	1.55 (0.98) nd-5.0	KUN
	0.97 (0.94) 0.65-1.35	0.34 (0.35) nd-0.60	nd	nd	nd	0.73 (0.74) 0.26-1.20	ZNZ[1)]
CB-118	1.20 (1.23) 1.09-1.28	1.43 (1.41) 1.15-1.83	10.8 (11.0) 10.4-11.1	42.3 (38.2) 0.79-70.5	4.52 (4.72) 0.45-7.95	21.6 (22.0) 14.7-26.8	DAR
	3.05 (2.99) 2.99-3.18	1.73 (1.74) 1.46-2.03	2.65 (2.58) 2.45-2.91	-	1.31 (0.73) nd-4.92	2.4 (2.03) nd-5.88	KUN
	1.21 (1.17) 1.08-1.41	nd	nd	nd	nd	0.48 (0.42) 0.22-0.96	ZNZ[1)]
CB-138	3.44 (3.39) 3.33-3.6	5.14 (5.21) 4.22-6.09	18.6 (18.8) 17.7-19.0	90.5 (73.3) nd-161	8.92 (8.7) 1.53-15.7	43.2 (44.4) 25.9-51.9	DAR
	8.01 (7.96) 7.82-8.29	6.69 (6.83) 5.57-7.17	9.28 (8.59) 8.40-10.8	-	4.24 (2.75) 1.61-12.6	8.78 (7.46) 2.28-18.4	KUN
	2.84 (2.78) 2.51-3.30	1.63 (1.70) 0.95-2.11	nd	nd	4.78 (4.88) 3.55-5.66	0.96 (0-78) 0.15-2.36	ZNZ[1)]
CB-153	6.06 (5.91) 5.78-6.50	8.08 (8.29) 6.25-10.5	34.9 (35.6) 33.1-35.9	159 (123) 1.49-283	19.2 (20.6) 3.12-30.5	82.0 (91.1) 43.5-97.6	DAR
	13.1 (13.1) 12.7-13.4	10.6 (10.8) 9.0-11.8	16.6 (16.5) 14.7-18.7	-	8.85 (5.23) 2.76-28.5	15.4 (12.9) 4.21-34.4	KUN
	6.58 (6.52) 5.91-7.39	0.30 (0.20) nd-1.09	nd	nd	nd	2.29 (1.88) 1.0-4.51	ZNZ[1)]
CB-180	2.87 (2.84) 2.81-2.97	3.86 (3.95) 2.96-4.95	14.5 (14.8) 13.8-15.0	60.2 (49.4) 1.05-102	5.84 (6.43) 1.12-9.07	31.5 (33.4) 15.7-42.0	DAR
	5.55 (5.44) 5.44-5.77	5.33 (5.49) 4.49-5.98	8.65 (9.25) 6.65-10.1	-	2.78 (1.76) nd-8.49	7.21 (5.63) 2.19-15.4	KUN
	2.51 (2.46) 2.29-2.82	0.69 (0.75) 0.36-0.86	nd	nd	0.90 (0.95) 0.43-1.52	1.38 (1.41) 0.55-2.67	ZNZ[1)]
BDE-47	0.34 (0.40) nd-0.61	1.22 (1.14) 0.12-2.66	2.81 (2.69) 2.41-3.33	3.31 (2.98) 0.71-5.14	0.53 (57) 0.26-0.80	5.79 (5.59) 2.0-11.0	DAR
	1.51 (1.46) 1.42-1.67	1.18 (1.11) 0.99-1.44	1.67 (1.31) 1.08-2.61	-	nd	1.21 (1.14) 0.78-1.73	KUN
	0.67 (0.60) 0.58-0.90	0.42 0.24-0.64	nd	1.34 (1.53) 0.62-1.67	nd	0.26 (0.17) 0.09-0.54	ZNZ[1)]
BDE-99	nd	nd	1.95 (1.67) 1.54-2.64	1.52 (1.50) nd-2.71	nd	2.26 (1.64) nd-6.07	DAR
	4.18 (3.75) 3.65-5.14	0.07 (0) nd-0.24	nd	-	nd	nd	KUN
	nd	nd	nd	nd	nd	nd	ZNZ[1)]
BDE-100	1.03 (0.93) 0.39-1.32	0.78 (0.75) 0.40-1.21	0.61 (0.58) 0.36-0.90	0.95 (1.14) nd-1.34	nd	1.03 (0.93) nd-2.86	DAR
	2.43 (2.43) 2.22-2.64	0.86 (0.80) 0.71-1.12	0.92 (1.37) nd-1.39	-	nd	nd	KUN
	2.04 (2.04) 1.75-2.33	nd	nd	nd	nd	1.42 (1.47) 0.37-2.17	ZNZ[1)]
BDE-153	nd	0.05 (0) nd.0.29	3.23 (3.21) 2.87-3.59	0.72 (0.66) nd-1.58	nd	2.13 (1.40) nd-2.43	DAR
	nd	0.16 (0.19) nd-0.33	nd	-	nd	nd	KUN
	0.95 (0.94) 0.82-1.08	nd	nd	3.50 (1.95) 0.80-9.31	nd	nd	ZNZ[1)]
BDE-154	nd	0.49 (0.53) 0.24-0.72	nd	1.30 (1.34) 0.69-1.66	nd	nd	DAR
	nd	0.60 (0.60) 0.55-0.65	0.65 (0.66) 0.20-1.09	-	nd	nd	KUN
	0.44 (0.45) 0.39-0.45	0.36 (0.35) nd-0.77	nd	0.55 (0.58) nd-1.04	nd	1.06 (1.10) 0.49-1.68	ZNZ[1)]
BDE-209	nd	nd	nd	nd	nd	1.81 (1.65) nd-4.39	DAR
	2.61 (3.18) 1.18-3.47	nd	nd	-	nd	nd	KUN
	nd	nd	4.15 (3.90) 1.68-6.85	2.10 (1.67) 1.04-4.04	nd	0.64 (0.48) nd-1.84	ZNZ[1)]
HBCDD	nd	nd	nd	9.12 (6.42) nd-19.6	nd	nd	DAR
	nd	nd	nd	-	nd	nd	KUN
	nd	1.69 (1.64) nd-4.71	nd	nd	nd	1.88 (1.02) 0.34-5.98	ZNZ[1)]

[1)] Data presented in Haarr (2021)

Paper III

III





Chemosphere 329 (2023) 138646

Contents lists available at ScienceDirect

Chemosphere

journal homepage: www.elsevier.com/locate/chemosphere

Chlorinated paraffins and dechloranes in free-range chicken eggs and soil around waste disposal sites in Tanzania

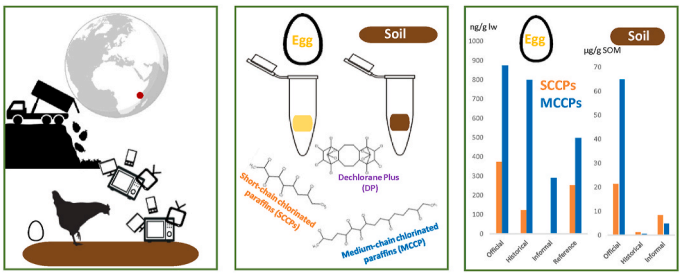
Ane Haarr [a,**], Maja Nipen [b], Eliezer B. Mwakalapa [c], Anders R. Borgen [b], Aviti J. Mmochi [d], Katrine Borga [a,e,*]

[a] Department of Biosciences, University of Oslo, P.O. Box 1066, 0316, Oslo, Norway
[b] Norwegian Institute for Air Research (NILU), P.O. Box 100, 2027, Kjeller, Norway
[c] Department of Natural Sciences, Mbeya University of Science and Technology, P.O. Box 131, Mbeya, Tanzania
[d] Institute of Marine Science, University of Dar es Salaam, P.O. Box 668, Zanzibar, Tanzania
[e] Center for Biogeochemistry in the Anthropocene, University of Oslo, PB 1066, 0316, Oslo, Norway

HIGHLIGHTS

- We quantify chlorinated paraffins and dechloranes in Tanzanian soil and chicken eggs.
- No clear association in contaminant levels or patterns between soil and eggs.
- Elevated contaminant levels in soil from e-waste location, but not in eggs.
- Risk assessment suggest concern for SCCP exposure via egg consumption at one site.

GRAPHICAL ABSTRACT



ARTICLE INFO

Handling Editor: J. de Boer

Keywords:
Flame retardants
Dumpsites
e-waste
Chicken eggs
Soil
Margin of exposure (MOE)

ABSTRACT

Electronic waste is a source of both legacy and emerging flame retardants to the environment, especially in regions where sufficient waste handling systems are lacking. In the present study, we quantified the occurrence of short- and medium chain chlorinated paraffins (SCCPs and MCCPs) and dechloranes in household chicken (*Gallus domesticus*) eggs and soil collected near waste disposal sites on Zanzibar and the Tanzanian mainland. Sampling locations included an e-waste facility and the active dumpsite of Dar es Salaam, a historical dumpsite in Dar es Salaam, and an informal dumpsite on Zanzibar. We compared concentrations and contaminant profiles between soil and eggs, as free-range chickens ingest a considerable amount of soil during foraging, with potential for maternal transfer to the eggs. We found no correlation between soil and egg concentrations or patterns of dechloranes or CPs. CPs with shorter chain lengths and higher chlorination degree were associated with soil, while longer chain lengths and lower chlorination degree were associated with eggs. MCCPs dominated the CP profile in eggs, with median concentrations ranging from 500 to 900 ng/g lipid weight (lw) among locations. SCCP concentrations in eggs ranged from below the detection limit (LOD) to 370 ng/g lw. Dechlorane Plus was the dominating dechlorane compound in all egg samples, with median concentrations ranging from 0.5 to 2.8 ng/g lw. SCCPs dominated in the soil samples (400–21300 ng/g soil organic matter, SOM), except at the official

* Corresponding author. Department of Biosciences, University of Oslo, P.O. Box 1066, 0316, Oslo, Norway.
** Corresponding author.
E-mail addresses: ane.haarr@ibv.uio.no (A. Haarr), maja.nipen@nilu.no (M. Nipen), mwakalapaeb@gmail.com (E.B. Mwakalapa), arb@nilu.no (A.R. Borgen), avitimmochi@gmail.com (A.J. Mmochi), katrine.borga@ibv.uio.no (K. Borga).

https://doi.org/10.1016/j.chemosphere.2023.138646
Received 13 January 2023; Received in revised form 2 April 2023; Accepted 7 April 2023
Available online 8 April 2023

dumpsite where MCCPs were highest (65000 ng/g SOM). Concentrations of dechloranes in soil ranged from below LOD to 240 ng/g SOM, and the dominating compounds were Dechlorane Plus and Dechlorane 603. Risk assessment of CP levels gave margins of exposure (MOE) close to or below 1000 for SCCPs at one location.

1. Introduction

Chemical flame retardants are added to a wide range of consumer goods, such as electronic products, that eventually become obsolete. Lack of sound handling and recycling of waste in developing regions in Africa represents an emerging environmental concern (White et al., 2020; Nipen et al., 2022b). Electronic waste (e-waste) is a source of both legacy, e.g. polychlorinated biphenyls (PCBs) and polybrominated diphenyl ethers (PBDEs), as well as emerging flame retardants, e.g. chlorinated paraffins (CPs) and dechloranes, that are replacing regulated contaminants (Abbasi et al., 2019; Nevondo and Okonkwo, 2021; White et al., 2020). Apart from some production of short chain chlorinated paraffins (SCCPs) in South Africa (10,000 tons year-1 as per 2015) and Egypt, other production of these contaminants has not been reported for Africa (Breivik et al., 2002; Abbasi et al., 2019; Hansen et al., 2020; Nevondo and Okonkwo, 2021). Thus, their increased environmental occurrence in this region is largely attributed to increased import and use of consumer goods along with lack of sound treatment of end-of-life products and waste (Breivik et al., 2015; Nipen et al., 2022b).

Declining levels of atmospheric legacy contaminants seen in industrialized regions as a result of reduced emissions is generally not found in developing countries (Gioia et al., 2011; Wong et al., 2021). Temporal trend studies using dated sediment cores show that concentrations of both legacy and emerging contaminants often peak in surface sediment layers in developing and newly industrialized regions, suggesting recent sources, compared to regions that have remained industrialized for some time, where peak concentrations in sediment layers correlate with peak in global production (Nipen et al., 2022b). A spatial shift in contaminant sources is thus occurring, from source regions where production and use have taken place historically, to source regions at the receiving end of obsolete products and waste (Breivik et al., 2011; Abbasi et al., 2015; Nipen et al., 2022b). This spatial shift can be attributed to increasing economic development and industrialization as well as a range of socio-economic factors (Nipen et al., 2022b), and stresses the need for expanding the research efforts on environmental contamination to developing regions in the Global South. Tanzania is experiencing rapid economic and population growth, expanding industry, and increasing import of consumer goods and potentially also waste. With limited capacity to safely manage an increasing stream of waste, both industrial and domestic, Tanzania faces a challenge regarding environmental contamination (Yhdego, 2017; Mahenge et al., 2018).

CPs are a large group of polychlorinated alkanes that encompass thousands of different congeners that are used in metal-working fluids, paints and sealants, and added to a wide range of products as plasticizers and flame retardants (Glüge et al., 2016, 2018). CPs have become ubiquitous in the environment since their introduction in the 1930s, and annual production is today estimated to exceed 1 million tons (Glüge et al., 2016, 2018; Wei et al., 2016; Vorkamp et al., 2019). Based on the carbon chain length, CPs can be classified into three groups: short chain CPs (C10–C13, SCCPs), medium chain CPs (C14–C17, MCCPs), and long chain CPs (>C18, LCCPs). They cover a wide range of physical-chemical properties, with octanol-water partitioning coefficients (Kow) varying from log Kow 5 to 7.5 for SCCPs and 6 to 10 for MCCPs (Glüge et al., 2013). Since the listing of SCCPs under Annex A of the Stockholm Convention on Persistent Organic Pollutants in 2017 (UNEP, 2017), MCCPs have increasingly been introduced as replacement substances (Glüge et al., 2018). However, studies indicate that MCCPs have similar Persistent, Bioaccumulative and Toxic (PBT) properties as the regulated SCCPs (Castro et al., 2019), and are currently listed for consideration under the European Chemicals Agency (ECHA, 2021). Dechloranes are another group of high production volume chlorinated flame retardants that includes Dechlorane Plus (DP) and Dechlorane 602 and 603. DP is used in plastic and electronic products and is currently listed for consideration under the Stockholm Convention due to its bioaccumulative properties and ability to undergo long-range transport (Hansen et al., 2020; Schuster et al., 2020). Estimated annual production of DP is around 750–6000 tons, while less is known about the chemical properties and production volumes for Dechlorane 602 and 603 (Wang et al., 2016; Hansen et al., 2020). The EFSA Panel on Contaminants in the Food Chain (CONTAM) recently performed benchmark dose (BMD) modelling on data from various studies on the effects of CPs on rodents, and selected as reference points a BMDL10 of 2.3 × 106 ng/kg body weight/day and 36 × 106 ng/kg body weight/day for SCCPs and MCCPs, respectively (EFSA CONTAM Panel et al., 2020). Similar reference points for risk assessment of dechloranes in food for human consumption are not available.

Most studies reporting occurrence of CPs and dechloranes in the environment are from the Northern hemisphere, and China is the main producer of these chemicals (Houde et al., 2008; Clement et al., 2012; Wei et al., 2016; Zhang et al., 2019). Very few studies have investigated CPs and dechloranes in the sub-Saharan African environment (Moeckel et al., 2020; Nevondo and Okonkwo, 2021; Nipen et al., 2022a). Concentrations and composition profiles of CPs and dechloranes in various environmental matrices are governed by complex processes and mechanisms including distance from primary sources, temperature and season, and distance from urban and industrialized areas (Wang et al., 2013; Nipen et al., 2022a). Due to relatively high Kow, these substances will partition to hydrophobic matrices such as organic matter in soil and lipids in organisms, which also affect their environmental distribution and accumulation (Castro et al., 2019; Hansen et al., 2020). In a tropical climate, semi-volatile organic substances, like PCBs, dechloranes and CPs, will more readily evaporate and could be transported and deposited at higher latitudes (Breivik et al., 2011; Vorkamp et al., 2019). Recently, elevated concentrations of CPs and dechloranes were found in air and soil in urban areas and around waste and e-waste disposal sites in Dar es Salaam, Tanzania (Nipen et al., 2022a). CPs were also detected in human milk samples from Africa, with concentrations exceeding concentrations of PCBs, and with a dominance of MCCPs over SCCPs suggesting a shift towards non-regulated compounds (Krätschmer et al., 2021). Prior to our study, no peer-reviewed literature exists on CPs and dechloranes in African biota, but SCCP concentrations in chicken eggs from selected sites in Africa, including Tanzania, are reported by the International Pollutants Elimination Network (IPEN) showing elevated levels around waste and e-waste disposal sites (Petrlik et al., 2021). Eggs from free-range household chickens (*Gallus domesticus*) are ideal indicators for contaminated soils, and eggs are a commonly included matrix in environmental monitoring programs globally (Polder et al., 2016; Petrlik et al., 2022). In addition to commercial feed, invertebrates, seeds and household leftovers, free-range chickens ingest considerable amounts of soil during foraging, which can lead to accumulation of contaminants in the tissues of the hen and further transfer into the egg (De Vries et al., 2006; Waegeneers et al., 2009; Polder et al., 2016; Petrlik et al., 2022). Chickens also accumulate CPs from feed (Ueberschär et al., 2007), and mixtures of both short- and medium chained CPs were found to be transferred from chicken to egg with increasing accumulation ratios with increasing carbon chain length and chlorine content (Mézière et al., 2021).

Few studies have previously quantified emerging contaminants in tropical regions far from production and use, where occurrence of these contaminants is expected to increase due to economic development and

industrialization. The aim of the present study was to assess the occurrence of CPs and dechloranes in soil and chicken eggs around waste disposal sites on mainland Tanzania and Zanzibar and to investigate if and how concentrations and congener group profiles vary among locations and between matrices. Collection of soil samples were conducted during an initial screening study of contaminant occurrence around waste disposal sites. As these initial analyses of soil samples showed elevated concentrations of CPs and dechloranes, this motivated collection of eggs from free-range, household chickens from the same locations and one presumed reference site in later fieldwork campaigns. Using the reference dose levels set for CPs and a margin of exposure (MOE) approach, risk assessment of CP exposure via egg consumption was conducted.

2. Materials and methods

2.1. Sample sites

In our initial screening sampling in January 2018, soil from waste disposal sites on the Tanzanian mainland and Zanzibar were specifically targeted to assess CP and dechlorane contamination from waste in general, and from electronic waste in particular (Fig. 1). The two sample sites in Dar es Salaam consisted of one historical dumpsite and the current official dumpsite. The historic Mtoni dumpsite in Dar es Salaam (−6.8730°, 39.2832°) was closed in 2009 after operating for around six years and is located around 8 km from Dar es Salaam city center. This historical dumpsite is in close vicinity of the Kizinga River flowing into Kurasini Creek, surrounded by residential areas. The currently used, official dumpsite at Pugu Kinyamwezi (−6.9278°, 39.12785°) is located around 25 km from the city center. Despite initial plans for building a sanitary landfill for Dar es Salaam, Pugu is today receiving industrial, agricultural, domestic, commercial, medical and other hazardous wastes (e.g. e-waste) without restrictions or regulations regarding leachate and gas management, fencing, soil covering and worker safety measures (Yhdego, 2017). A licensed e-waste dealer is located next to the official dumpsite, where e-waste collection, sorting and dismantling take place. On Zanzibar, the Maruhubi dumpsite (−6.1442°, 39.2101°) is an informal dumpsite receiving solid wastes from Zanzibar municipality. It is located around 3 km from Zanzibar city center, and close to mangrove forests. From these same sites, chicken eggs were sampled in August 2018 (Zanzibar) and February 2019 (Tanzanian mainland/Dar es Salaam). In addition, eggs were collected from an area away from any known waste disposal sites, a residential area (Bumbwini Makoba, −5.9500°, 39.2000°) around 25 km North of Zanzibar city. Soil samples were not obtained from the residential area on Zanzibar. However, soil samples collected at rural locations around Dar es Salaam as part of a parallel study by Nipen et al. (2022a) are used for reference.

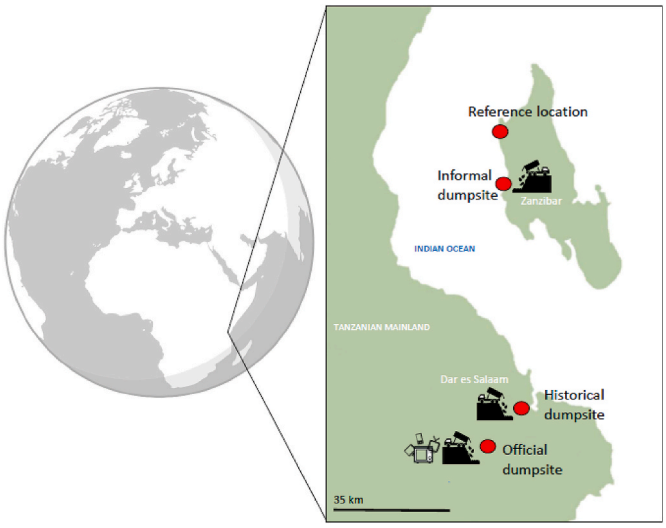


Fig. 1. Sampling locations on mainland Tanzania and Unguja island of Zanzibar. Free-range household chicken eggs and soil were collected at three waste disposal sites: The official dumpsite of Dar es Salaam located next to a small e-waste handling facility, a historical dumpsite in Dar es Salaam, no longer in use, and an informal dumpsite on Zanzibar, where unofficial waste disposal takes place. Only egg samples were collected from the residential area, located away from any waste disposal sites.

2.2. Sample collection and preparation

At each location, three soil cores (approximately 5 cm deep) were collected within a range of three to five square meters using a stainless-steel hand-held corer. The three cores from each site were pooled into one composite sample. Soil organic matter content (SOM) was estimated by loss on ignition using dried sample material. SOM in the soil samples varied among locations, from dry, sandy soil at the official dumpsite (SOM 3.2%), to 12% SOM at the informal dumpsite on Zanzibar. Two composite soil samples were collected at the historical dumpsite, one from a location influenced by run-off from the old dumpsite and one around 200 m away from the run-off location. The latter had lower SOM content (4.1%) compared to the area influenced by run-off from the old dumpsite (8.3% SOM).

The chickens were kept outside picking food from the local grounds, but owners also reported giving them additional feed such as food scraps and industrial feed. Six eggs were collected from each location, in total 24 eggs. The egg-laying sequence of the sampled eggs is not known. The whole egg content was collected in falcon tubes and kept frozen (−20 °C) until laboratory analyses.

2.3. Sample extraction

Composite soil samples (around 7 g) were dried and homogenized together with sodium sulphate with a mortar and pestle to a fine powder. Fresh samples of whole eggs (around 1 g) were used for extraction after homogenization with sodium sulphate. Soil samples were extracted using accelerated solvent extraction, while egg samples were extracted using a cold column technique, both extractions using acetone and n-hexane (1:1) after the addition of internal standards (13C - labelled 1,5,5,6,6,10 – hexachlorodecane, DP syn and Dec 602). Extracts were concentrated using a Turbovap system (Zymark) and sulphuric acid was used for removal of lipids. Extracts were cleaned with 4 g activated silica and a 1 cm layer of sodium sulphate packed in a column eluted with diethyl ether in n-hexane (1:9). Solvent was changed to isooctane, volume concentrated to 100 μL, and 1,2,3,4-tetrachloronaphtalene (TCN) was added as recovery standard. Extracts were analyzed for CPs, DP (syn- and anti-isomers), Dec 602, and Dec 603 using gas chromatography quadrupole time-of-flight high-resolution mass spectrometry (GC/Q-TOF) (Agilent, Santa Clara, USA) in electron capturenegative ionization (ECNI) mode. 34 SCCPs and 28 MCCPs congener groups (groups of CPs sharing the same molecular formula) were identified and quantified. A total of 22 egg samples were analyzed for CPs (two samples were lost during sample preparation), and 24 egg samples were analyzed for dechloranes. Lipid content in the egg samples was determined gravimetrically.

2.4. Quantification of chlorinated paraffins

The quantification of CPs in soil in the initial screening study was conducted using the method described by Tomy et al. (1997), where the technical mixture with the closest matching chlorination degree and congener group pattern is used for quantification of each sample. As the chemical analysis of chlorinated paraffins was a field in rapid development at the time of our study, the laboratory further improved its methods and updated to the deconvolution method developed by Bogdal

et al. (2015), by the time of the follow up study with egg analysis. The congener group profiles of CPs are comparable across the two methods of quantification as the same instrument was used for both soil and egg samples. Unfortunately we were not able to re-quantify the initial screening samples based on the Bogdal method, but our soil data from the official dumpsite/e-waste facility of Dar es Salaam were comparable to CPs quantified in soil samples collected from the same location the following year using the deconvolution method (Nipen et al., 2022a) (See quality assurance below).

The deconvolution method by Bogdal et al. (2015) involves quantification of CPs using technical mixtures with different chlorination degree (SCCPs: C10 – C13 standards with 51%, 55% and 63% Cl; MCCPs: C14 – C17 standards of 42%, 52%, and 57% Cl, Dr. Ehrenstorfer, Germany) and standards with single carbon chain lengths as calibration standards (SCCPs: C10, C11, C12, and C13 (50% and 65% Cl); MCCPs: C14 with 52% Cl, Dr. Ehrenstorfer, Germany). A mathematical deconvolution method reconstructs the congener group pattern in the samples using a linear combination of the congener group patterns in the calibration standards. Match between sample and standard pattern was assessed with the goodness of fit where R2 above 0.5 was considered acceptable. Average R2 for SCCPs were 0.91 while average R2 for MCCPs was 0.89 (Table A1).

2.5. Quality assurance & quality control

Despite using different quantification methods of soil CP occurrence in the present study and the soil transect study by Nipen et al. (2022a), the concentrations are comparable in the two soil samples from same site (the official dumpsite/e-waste facility in Dar es Salaam: present study: 670 ng/g dw SCCPs, 2100 ng/g MCCPs, and 7.6 ng/g dw DP. Sample W-23 from Nipen et al., 2022a: 480 ng/g dw SCCPs, 2700 ng/g dw MCCPs, and 6 ng/g dw DP). This confirms the quality and representativeness of the quantification methods used in the present study.

Substantial measures were done to avoid contamination during sample preparation and extraction, including use of high-quality grade solvents, glassware heat-treated at 450 °C and rinsed with solvents immediately prior to use, and sample preparation procedures performed in a laminar flow cabinet when possible. For every sample batch (samples were run in five batches), 1–2 blank samples were included to account for any background signal or contamination of CPs during the extraction. In total, eight blank samples were run to assure representative values of the background signal. The limit of detection (LOD) was defined as three times standard deviation of the blanks. The LODs for CPs in eggs were 16.2 ng/g ww and 65.0 ng/g ww for SCCPs and MCCPs, respectively, and 11 ng/g dw and 19 ng/g dw for SCCPs and MCCPs, respectively, in soil. LODs for dechloranes in eggs were 0.01 ng/g ww for Dechlorane 602 and 603, and 0.02 for Dechlorane Plus. The LODs for dechloranes in soil were 0.01 and 0.001 ng/g dw for Dechlorane Plus, and Dechlorane 602 and 603, respectively. The ratios of TCN and 13C labelled internal standards were compared against the ratios of TCN and internal standards in quantification mixtures (which were not subject to extraction and clean-up) to determine the recovery of internal standards. Recovery of CPs in soil samples were between 54% and 57% and between 71% and 87% for dechloranes. Recovery of CPs in eggs were between 83% and 130% (Table A1). Average recoveries of dechloranes in eggs were between 84% and 147% (calculated from two radiolabeled congeners used for recovery calculations, Table A2). Quantification of both CPs and dechloranes involves addition of internal standards prior to sample extraction and clean up, and thus recovery percentage is automatically corrected for.

2.6. Risk assessment of CP intake via egg consumption

Human risk assessment of CP intake via eggs was assessed using the margin of exposure (MOE) approach. Egg consumption for the Tanzanian population is estimated to be 106 eggs per person per year (Ringo and Lekule, 2020), and average weight for all eggs sampled in the present study (46.3 g) was used to calculate daily egg intake in grams per day. Daily intake of CPs via consumption of eggs was calculated using the following formula:

$$\text{Egg intake} = (\text{ConcCP x ecd x lipidegg}) \ / \ \text{BW}$$

where ConcCP is SCCP or MCCP concentration in the egg samples (ng/g lw); ecd is the daily egg consumption (g/day); lipidegg the average lipid content (%) of eggs at each location in the present study; and average human body weight (BW), set at 60 kg. The MOE was calculated using the following formula:

$$\text{MOE} = \text{BMDL10} \ / \ \text{Egg intake}$$

where BMDL10 is 2.3 × 106 ng/kg bw/day and 36 × 106 ng/kg bw/day for SCCPs and MCCPs, respectively. The CONTAM Panel conclude that a MOE higher than 1000 indicate no health concern.

2.7. Data treatment and statistical analyses

All statistical analyses were conducted using R Studio. Compounds detected below LOD in more than 85% of the samples were removed from further data treatment. This included SCCPs in eggs from the informal dumpsite on Zanzibar (Table A1). Otherwise, values that were quantified but below LOD were included for data treatment and statistical analyses. This included three egg samples for SCCPs and five samples for MCCPs (Table A1). Due to low sample size, and because assumptions of normality and equal variance were not met, assessment of spatial variation of contaminant concentrations in eggs was conducted using the non-parametric Kruskal Wallis test.

Assessment of CP congener group profiles among sites and between matrices was done using principal component analyses (PCA) with the vegan package for R (Jari Oksanen et al., 2019). All variables were scaled to zero mean and unit standard deviation. For the multivariate assessment of CP congener group profiles, individual congener groups (e.g. C10H17Cl5) were normalized to total CP concentrations (SCCPs + MCCPs). For visualization of CP congener group profiles in Fig. 4, individual congener groups were normalized to total SCCP and MCCP concentrations, respectively.

3. Results

CPs was the dominant contaminant group in both egg and soil samples from the present study, with concentrations up to three orders of magnitude higher than for dechloranes (Table 1). Elevated concentrations of SCCPs, MCCPs and DP in soil were found at the official dumpsite compared to the other locations (Fig. 2), but there was no clear association between concentrations or congener group profiles between egg and soil samples. Contaminant concentrations in eggs did not differ between dumpsites and the reference location, and the standard deviation within sites was relatively large (Table 2). Contaminant concentrations on a wet weight/dry weight basis with LODs annotated can be found in the Appendix (Figure A1).

Mean lipid content in eggs ranged from 8.9% from the official dumpsite to 14.2% in eggs from the residential area. Due to the high lipid solubility of CPs and dechloranes and differences in lipid content among eggs from the different locations, the assessment of spatial variation in contaminant concentrations was conducted on a lipid weight basis to ensure that spatial variability in CP and dechloranes reflects variation beyond lipid content. SOM content in the soil samples ranged from 3% to 12%. Due to low sample size, statistical testing of differences in SOM content or contaminant concentrations among locations could not be performed, but data are presented normalized to SOM, also to allow comparison to the egg lipid normalized concentrations.

Table 1
Concentrations of short and medium chained chlorinated paraffins (SCCP, MCCP) and dechloranes (mean (median), range (standard deviation, SD), ng/g) in chicken eggs from four locations in Dar es Salaam (Tanzanian mainland) and Zanzibar (Unguja island).

Location	n	Lipid%	SCCP	MCCP	MCCP/SCCP	Dech Plus (DP)	Dech 602	Dech 603
		mean	**ng/g wet weight** *mean (median)* *min-max (SD)*					
Official, DAR	5/6[a]	8.9	38 (37) 32–49 (6)	81 (76) <LOD–110 (18)	2.7	0.04 (0.04) 0.03–0.05 (0)	<LOD	<LOD
Historical, DAR	6	10	40 (17) <LOD–115 (41)	130 (78) <LOD–305 (93)	4.4	0.3 (0.1) 0.1–0.6 (0.2)	<LOD	0.2 (0.1) 0.1–0.3 (0.1)
Informal, ZNZ	6	11.6	<LOD	64 (67) <LOD–75 (10)	6.6	0.3 (0.3) 0.2–0.5 (0.1)	0.02 (0.02) 0.01–0.02 (0.002)	0.2 (0.3) 0.2–0.3 (0.03)
Reference, ZNZ	5/6[a]	14.2	33 (28) 19–57 (13)	86 (70) <LOD–127 (28)	2.9	0.1 (0.1) 0.1–0.3 (0.1)	<LOD	0.2 (0.1) 0.03–0.9 (0.3)
		mean	**ng/g lipid weight** *mean (median)* *min-max*					
Official, DAR	5/6[a]	8.9	480 (370) 290–790 (185)	1100 (870) <LOD–2400 (655)	2.7	0.5 (0.5) 0.3–0.8 (0.2)	<LOD	<LOD
Historical, DAR	6	10	380 (220) <LOD–940 (325)	1200 (900) <LOD–2500 (683)	4.4	2.5 (1.5) 0.7–5 (1.8)	<LOD	1.6 (1.4) 0.9–2.7 (0.7)
Informal, ZNZ	6	11.6	<LOD	560 (560) <LOD–670 (91)	6.6	2.9 (2.8) 1.6–4.1 (1)	0.2 (0.2) 0.1–0.2 (0.02)	2.1 (2.2) 1.8–2.4 (0.2)
Reference, ZNZ	5/6[a]	14.2	240 (250) 130–390 (94)	650 (500) <LOD–1230 (323)	2.9	0.8 (0.7) 0.4–2 (0.6)	<LOD	1.5 (0.4) 0.3–6.2 (2.1)

[a] Five eggs analyzed for chlorinated paraffins, six eggs analyzed for dechloranes.

3.1. Concentrations of chlorinated paraffins in eggs and soil

SCCPs were detected above LOD in 13 of 22 egg samples, while MCCPs were above LOD in 16 of 22 egg samples. Median concentrations of SCCPs in eggs ranged from <LOD to 370 ng/g lipid weight (lw) among locations. SCCPs were below LOD in all eggs from the informal dumpsite on Zanzibar while highest concentrations were found at the official dumpsite ($p = 0.02$, $\chi 2$ Kruskal Wallis = 14.5, Fig. 2, Table 1). MCCP concentrations in eggs were higher compared to SCCPs at all locations (SCCP/MCCP ratio ranging from 0.3 to 0.5, Table 1). Concentrations ranged from 500 to 900 ng/g lw with no significant difference among locations ($p = 0.09$, $\chi 2$ Kruskal Wallis = 6.43, Fig. 2). The average MOE values ranged from 2400 to 3100 for SCCPs and 13,500 to 24,900 for MCCPs among locations (Table A3). At all locations, MOE for MCCPs exceeded 1,000, while MOE for SCCPs was below or close to 1000 (890 and 1300, Fig. 3) in two egg samples collected at the historical dumpsite in Dar es Salaam.

In the soil samples, CP concentrations were highest at the official dumpsite next to the e-waste handling facility in Dar es Salaam. MCCPs was the dominating CP group at the official dumpsite (SCCP/MCCP ratio 0.3, Table 1), while SCCPs dominated at the other locations. Concentrations among locations ranged from 400 to 21300 ng/g SOM for SCCPs and <LOD to 65000 ng/g SOM for MCCPs (Table 1).

3.2. Concentrations of dechloranes in eggs and soil

Dechlorane Plus (combining both syn- and anti-isomers) was detected in eggs from all locations, and was the dominant dechlorane group. Median DP concentrations ranged from 0.5 ng/g lw to 2.8 ng/g lw among locations, and highest concentrations were found at the informal dumpsite on Zanzibar ($p = 0.006$, $\chi 2$ Kruskal Wallis = 12.3, Fig. 2). Dechlorane 603 was detected above LOD in 18 of 24 samples, with median concentrations ranging from <LOD to 2.2 ng/g lw. Dechlorane 602 was only detected above LOD in eggs from the informal dumpsite on Zanzibar (Table 1).

Dechlorane 602 was detected at relatively low concentrations (below or close to LOD) in soil from all sites. However, relatively high concentration of Dechlorane 603 was found at the historical dumpsite, at the location away from the area affected by run-off from the old dumpsite. Here, concentration of Dech 603 was up to two orders of magnitude higher compared to the other locations, and higher than DP from the same site (230 and 12 ng/g SOM, respectively). Except from at this site, DP dominated the dechlorane pattern in soil, and the concentration was highest at the official dumpsite of Dar es Salaam and the informal dumpsite on Zanzibar, 240 and 70 ng/g SOM, respectively (Table 2).

3.3. CP congener group profiles in egg and soil

C10 and C14 homologues were evenly distributed among samples and locations, while C15–C17 homologues were generally more present in eggs compared to soil. SCCPs with higher chlorination degree dominated in the soil samples, while MCCPs with lower chlorination degree dominated in the egg samples (Fig. 4). The multivariate analyses of CP congener group profiles showed a gradient from high to low chlorination degree along the first principal component (PC1 28%) of the PCA plot (Fig. 5). Congener groups with more chlorine atoms pointed in the direction of the soil samples, and groups with fewer chlorine atoms point in the direction of the egg samples. There is a gradient from low to high carbon chain length along the second principal component (PC2, 21%), with increasing relative contribution of CPs with shorter carbon chain lengths in soil samples and longer carbon chain lengths in egg samples with increasing PC2 values. PC1 and PC2 together account for 49% of the total variation in the dataset, meaning that there might be other drivers of variation in the data not captured in this study. Some CPs had higher absolute loadings on the third axis, PC3 (C10Cl5, C10Cl6,

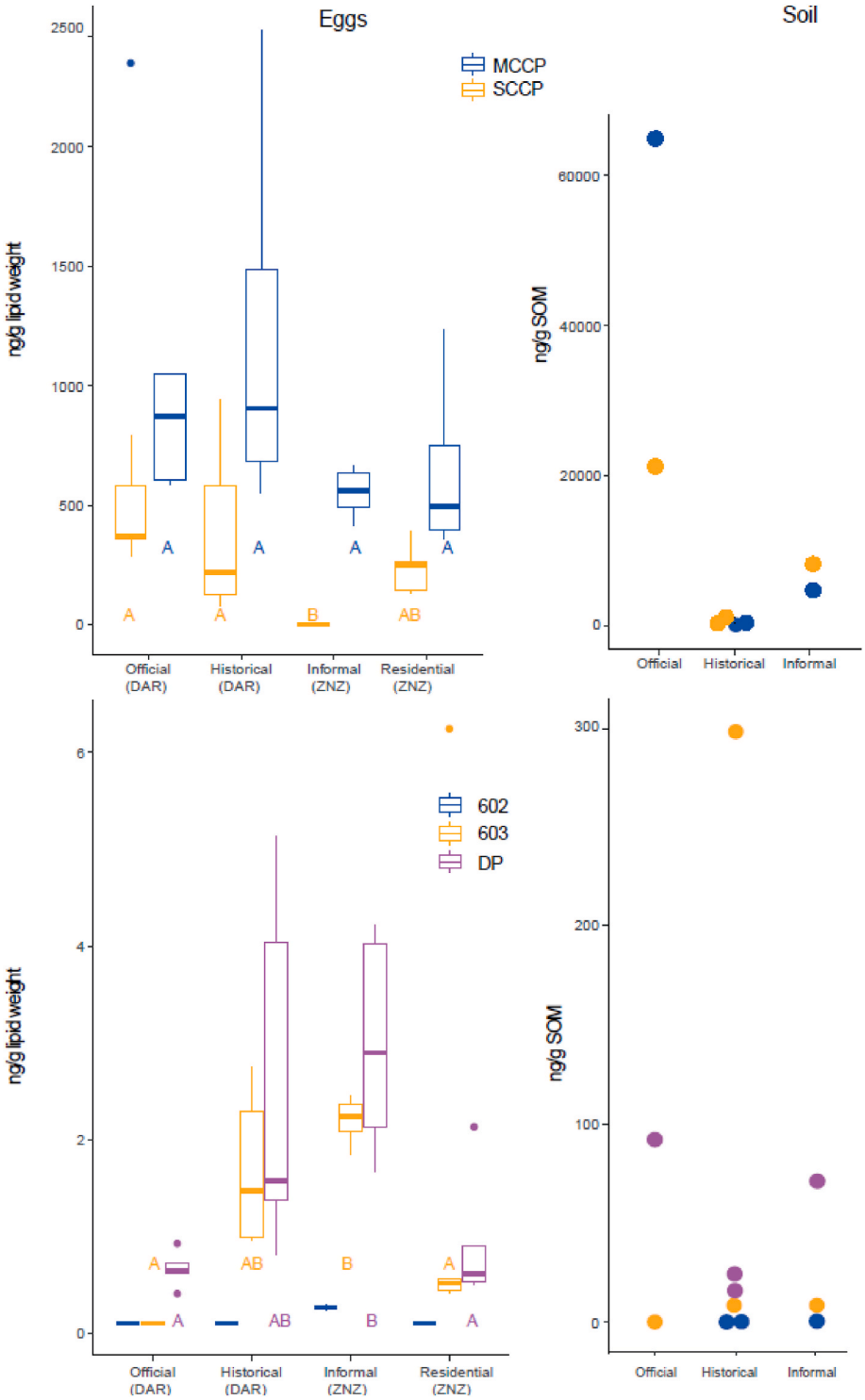


Fig. 2. Concentrations of chlorinated paraffins and dechloranes in eggs (ng/g lipid weight) and soil (ng/g SOM) from three dumpsites on mainland Tanzania and Zanzibar. Groups not significantly different from each other (Kruskal Wallis test) share letter code (A, B).

C13Cl5, C13Cl6, C14Cl5, C16Cl7, C16Cl8), but due to the low amount of variation explained by this axis (12%), PC3 was not considered important for interpretations of the main trends in the results.

4. Discussion

Elevated concentrations of SCCPs, MCCPs and DP in soil at the official dumpsite next to the e-waste facility in Dar es Salaam could indicate that waste and e-waste represent a contaminant source to the Tanzanian environment. However, due to the low spatial variation of CP concentrations in eggs and the large variation within sites, no clear conclusions regarding a waste signal can be made. Unknown contaminant sources at the residential area cannot be ruled out. Particularly high concentrations of SCCPs in two eggs from the historical dumpsite resulted in MOE close to or below the 1000-limit set by the CONTAM panel. This suggests that consumption of eggs from this location represent a health concern, which warrants further research on human exposure to CPs and their health effects.

Table 2
Concentrations of chlorinated paraffins and dechloranes in soil samples collected at three locations around Dar es Salaam and Zanzibar, Tanzania. Each soil sample consists of three composite samples.

Location	%SOM	SCCP	MCCP	SCCP/MCCP	Dech Plus (DP)	Dech 602	Dech 603
ng/g dry weight							
Official (DAR)	3.2	670	2100	0.3	7.6	0.004	<LOD
Historical (DAR)	8.3	100	40	2.5	0.7	0.01	0.4
Historical II (DAR)	4.1	17	<LOD	1.9	0.5	<LOD	10
Informal (ZNZ)	12.1	1010	580	1.7	8.4	0.1	0.3
ng/g SOM							
Official (DAR)	3.2	21300	65000	0.3	240	0.1	<LOD
Historical (DAR)	8.3	1250	500	2.5	9	0.1	4
Historical II (DAR)	4.1	400	<LOD	1.9	12	<LOD	230
Informal (ZNZ)	12.1	8300	4800	1.7	70	0.6	2.5

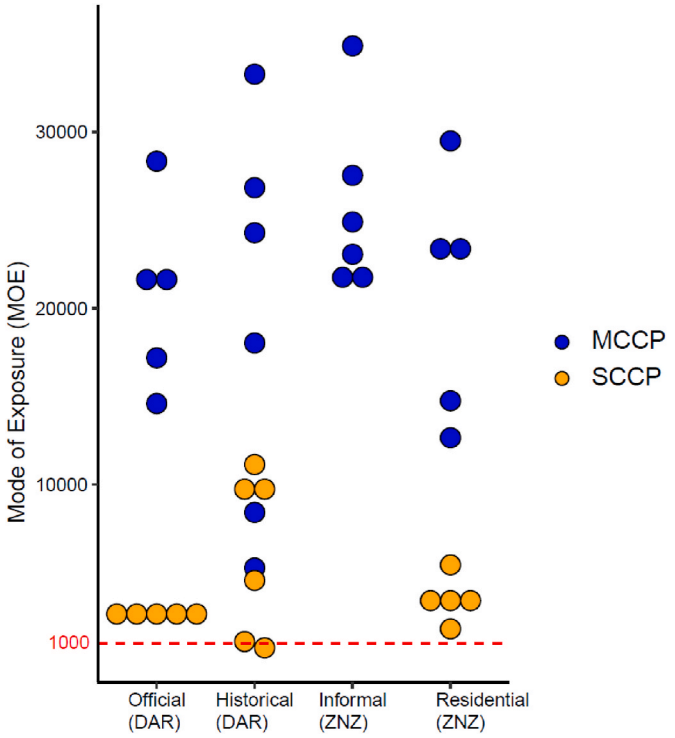


Fig. 3. Mode of Exposure (MOE) calculated for SCCPs and MCCPs in individual egg samples collected from locations on mainland Tanzania and Zanzibar. The MOE limit at 1000 (indicated by the red, dotted line) is set by the CONTAM panel for human exposure to CPs and MOE above this limit indicate no risk. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

Despite limited amount of soil samples collected in the present study, differences in CP congener group patterns between soil and eggs were observed. While MCCPs was the dominant CP group in the egg samples, SCCPs dominated in two out of three soil samples. This may be due to higher Kow and affinity to lipid-rich tissues such as eggs for CPs with longer carbon chain lengths. Elevated concentrations of Dechlorane Plus in soil from the official and informal dumpsites, compared to the historical dumpsite, could also indicate a waste signal. As opposed to the historical dumpsite, the informal and official dumpsites are active and currently receiving household wastes, which might be represented by more modern products and consumer goods compared to what has been discarded at the historical dumpsite that was shut down in 2009. Elevated concentrations of Dechlorane 603 at the historical dumpsite but not at the official dumpsite suggest local use of pesticides rather than a waste signal, as this compound can occur as an impurity in the pesticides aldrin and dieldrin (Shen et al., 2011). Higher concentrations of MCCPs compared to SCCPs in soil samples at the official dumpsite next to the e-waste handling facility could be a consequence of more modern products containing MCCPs being disposed of at this location, as regulatory efforts have led to a dominance of MCCPs over SCCPs in environmental compartments (Gluge et al., 2018). The calculated MOEs for the CP exposure via consumption of eggs from the present study do not suggest a health concern for MCCPs (all MOEs >1000), whereas one egg from the historic waste site had SCCPs levels of consern with MOE values below 1000. However, this limited scenario does not account for differences in egg consumption or CP exposure from other sources, and further investigations should be made regarding human exposure to CPs and dechloranes and potential health risks.

In general, chlorinated paraffin and dechlorane concentrations in eggs and soil in the present study were higher compared to what has been reported from Europe, but several orders of magnitude lower compared to e-waste locations in China (Table 3). Due to few replicates in the present study, comparisons between other studies must be done with care. In addition, analytical challenges and uncertainties, especially regarding quantification of CPs, make comparisons among different laboratories challenging. Concentrations of CPs and DP in home-produced eggs from e-waste locations in China were up to three orders of magnitude higher compared to the present study (Zheng et al., 2012; Zeng et al., 2018). Concentrations of DP in eggs from the present study were in the same range as in eggs from Latvia (Zacs et al., 2021). SCCPs could not be detected in chicken eggs purchased from different suppliers from Belgium (LOQ: 0.5 ng/g ww), while MCCP concentrations were around ten times lower than concentrations in eggs from the present study (McGrath et al., 2021, Table 3). Concentrations of CPs in soil from an e-waste dismantling location in China were 200 times higher than the highest SCCP concentration found in the present study, and over three orders of magnitude higher compared to the highest MCCP concentration measured in the present study (Xu et al., 2019). In background soil samples collected from Norway and the UK in 2008, average concentrations of SCCPs were more than ten times lower than the lowest concentration measured in the present study (Halse et al., 2015). Soil samples collected at rural sites outside Dar es Salaam (Table 3) showed CP concentrations in the same range as samples collected in the present study from the historical dumpsite, and lower concentrations of DP (Nipen et al., 2022a). This supports the hypothesis that wastes containing CPs and dechloranes have not readily been discarded at the historical dumpsite, and that soil from this location does not represent a major contaminant source to household chickens foraging in the area.

Avian maternal transfer of CPs is lower compared to other POPs, such as PCBs and DDT (Choo et al., 2020; Guan et al., 2020; Knudtzon et al., 2021). However, the effects of CP chain length, chlorine content and chlorine distribution/positioning on bioaccumulation and maternal transfer are still not well understood (Bettina et al., 2011; Mézière et al., 2021). After dietary exposure to five technical mixtures, the maternal transfer of CPs with both low and high chlorine content from feed to chicken eggs was favored for SCCPs and MCCPs, whereas lower

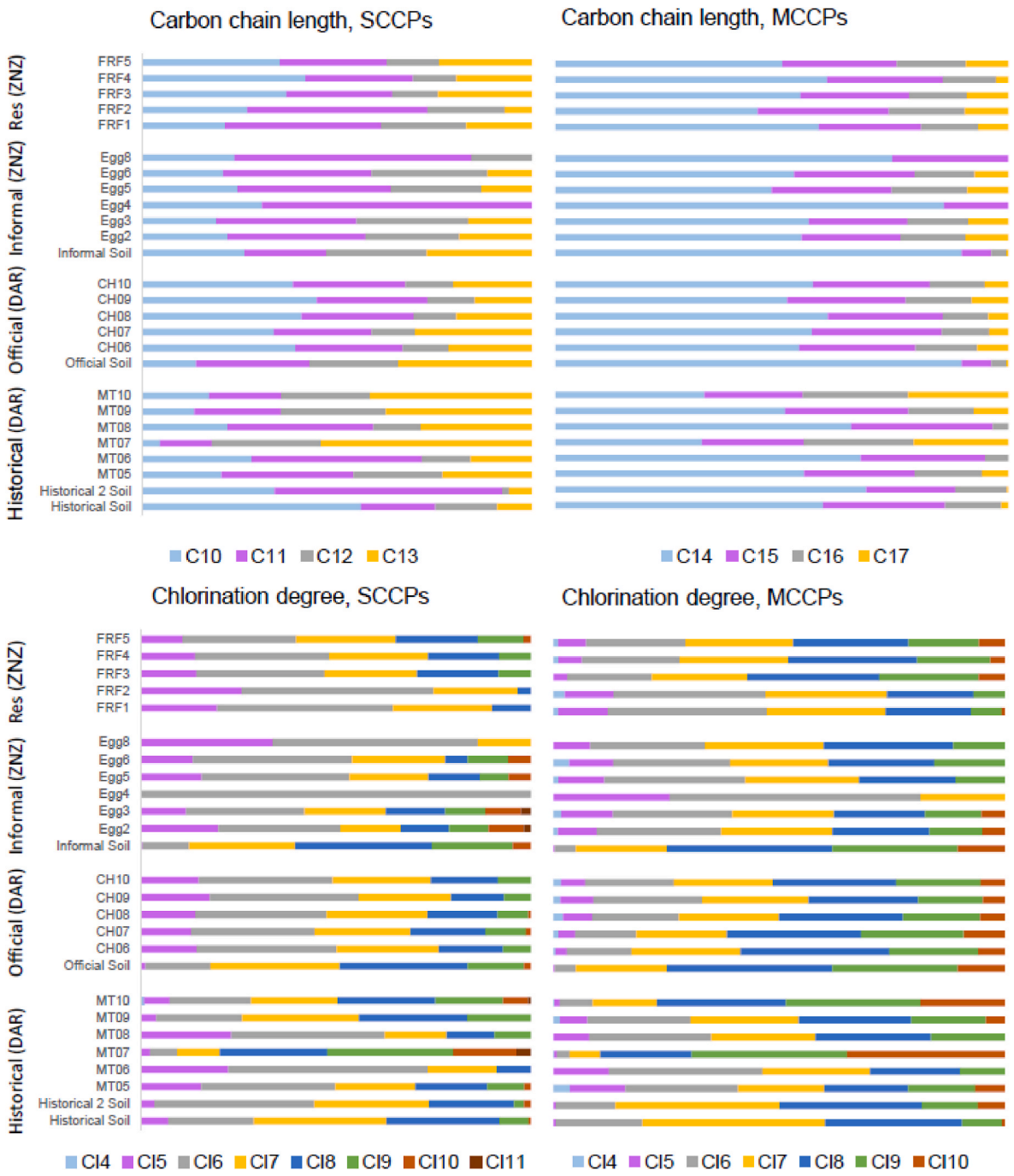


Fig. 4. Congener group profiles for chlorinated paraffins (SCCPs and MCCPs) in chicken egg and soil collected around various waste disposal sites on mainland Tanzania and Zanzibar.

chlorinated congeners were transferred for long chained chlorinated paraffins (LCCPs) (Mézière et al., 2021). Maternal transfer of CPs in watersnake, Enhydris chinensis, (egg to muscle ratio) decreased with increasing log Kow (ranging from log Kow 5 to 10) (Guan et al., 2020). Contrary, egg to liver ratio of CPs in the black-spotted frog, Pelophylax nigromaculatus, increased with log Kow and carbon chain length, suggesting that maternal transfer of CPs is related to the lipophilicity of the chemical. In addition, maternal transfer and chlorine content had a parabolic relationship, where egg to liver ratio increased up to chlorine content of 6–7, and then decreased as chlorine content increased further (Du et al., 2019). This parabolic relationship was also found for PBDEs in the rice frog, Rana limnocharis, with maternal transfer ratio peaking at 6–7 bromine atoms and log Kow at around 8 (Wu et al., 2009). A study by Zheng et al. (2018) also suggests highest maternal transfer rate for mid-halogenated POPs with log Kow of 6.5–7 in the Kentish Plover, Charadrius alexandrines. This seems in accordance with the present study, where CPs with 5–8 chlorine atoms and log Kow values around 6.5–9 were dominating in the egg samples. Dechlorane Plus is a relatively large molecule with a log Kow value above 9, and a high potential for maternal transfer in chickens compared to other organohalogen contaminants, including PBDEs and PCBs. However, the mechanisms for maternal transfer of DP are not clear (Li et al., 2021), and cannot be assessed further in the present study.

The different sampling times of egg and soil pairs (collected up to one year apart) from each location could affect the lack of correspondence in egg and soil concentrations in the present study. However, this is

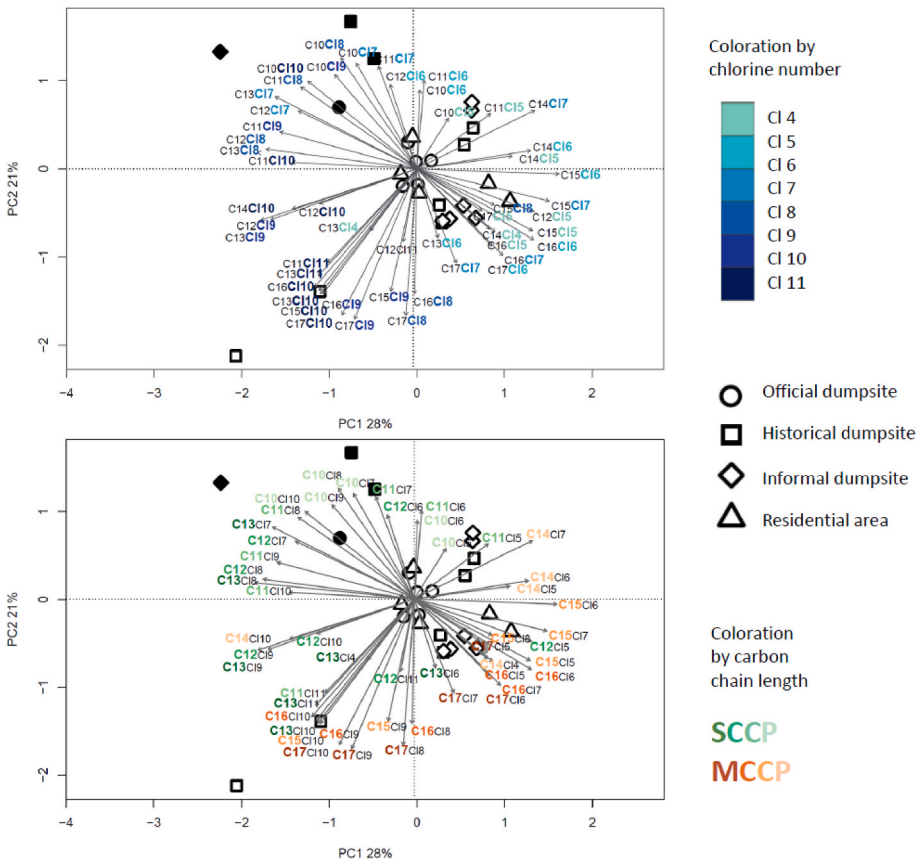


Fig. 5. Biplots of congener group profiles of chlorinated paraffins in eggs and soil from dumpsites on mainland Tanzania and Zanzibar. The different locations are shown as different symbols. Open symbols represent eggs, while filled symbols represent soil. Carbon chain length and chlorination degree of the different congener groups are highlighted in the two plots, respectively. Coloration of congener groups with light to darker blue with increasing chlorine number (above), and by carbon chain length (below) with darker green or orange for increasing chain length of short-chain and medium-chain CPs, respectively. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

Table 3
Concentrations of CPs and dechloranes in chicken egg and soil reported from other studies. Concentrations are given in ng/g (different units used for normalization (lipid weight, SOM) is annotated). Mean (median) and range is reported when given in reference.

Location	n	Matrix	SCCP	MCCP	Dech Plus (DP)	Unit	Reference
Tanzania	1 (pooled)	egg	599 (–)	NA		lw	Petrlik et al. (2021)
China (e-waste)	30	egg	10,100 (1490) 611-111,000	6830 (999) 297-91,100		lw	Zeng et al. (2018)
Belgium	10	egg	ND	- (10) 3.3–16		ww	McGrath et al. (2021)
China (e-waste)	33	egg			- (–) 665–3290	lw	Zheng et al. (2012)
Latvia	8	egg			0.03	ww	Zacs et al. (2021)
China (e-waste)	7	soil	43000 (2300) 69-2,2x105	841000 (22600) 507-4,4x106		dw	Xu et al. (2019)
Norway & UK	58	soil	35 (–) <0.6–570	NA		ng/g SOM	Halse et al. (2015)
Tanzania (waste transect)	9	soil	670 (40) 11–5300	966 (160) <19-5100	0.9 (0.1) <0.01–6	dw	Nipen et al. (2022a)
Tanzania (rural transect)	6	soil	31 (58) <11-120	19 (19) <19-210	0,04 (0,04) 0,03–0,07	dw	Nipen et al. (2022a)

probably more relevant in regions where organisms experience large seasonal variation in lipid content (Warner et al., 2019). In addition to soil, industrial feed is also considered a significant source of CPs to domestic chickens (Dong et al., 2019). Future studies should include analyses of industrial feed, in addition to soil, to assess its importance as a source of CPs and dechloranes to domestic animals.

5. Conclusion

This study is one of very few studies to quantify levels of CPs and dechloranes in biota from Sub-Saharan Africa. A spatial shift in contaminant sources is expected and stresses the need for increasing the monitoring- and research efforts to developing regions in the Global South. Our results show higher levels of MCCPs compared to SCCPs in eggs, while SCCPs were dominating in two out of three soil samples. The detected levels of dechloranes were up to three orders of magnitude lower compared to CPs with DP as the dominant compound. Further, our results indicate that MCCPs congeners with lower chlorine content (5–8 chlorine atoms) seem to accumulate more readily in eggs compared to soil. This might be due to dechlorination of MCCPs in the hen or that larger congeners with higher log Kow are less bioavailable or more readily eliminated prior to maternal transfer. This would need to be confirmed by comparing the congener profiles in the eggs with profiles in other tissues from the mother, which is beyond the scope of the present study. Based on results from the present study, no clear conclusion can be made regarding waste and e-waste as a source of CPs

and dechloranes to the Tanzanian environment. Rather, results indicate that CPs and dechloranes are ubiquitous in the environment as a result of large-scale production and lack of regulations. Risk assessment of CPs shows that consumption of eggs from free-range chickens could represent a health concern regarding exposure to SCCPs, which warrants further research on the occurrence and sources of these chemicals in this region.

Credit author statement

Ane Haarr: Conceptualization, Investigation, Formal analysis, Visualization, writing – original draft preparation. **Maja Nipen:** Investigation, Resources – contaminant analyses, Writing - review and editing. **Eliezer B. Mwakalapa:** Investigation, Writing - review and editing. **Anders R. Borgen:** Resources – contaminant analyses, Writing - review and editing **Aviti J. Mmochi:** Resources – provision of laboratory facilities in Tanzania, Writing - review and editing **Katrine Borgå:** Supervision, Project administration, Conceptualization, Writing - review and editing.

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.

Data availability

Data will be made available on request.

Acknowledgements

The present study was conducted as part of the University of Oslo: Life Science convergence project ANTHROTOX, with the overall aim to combine social and natural sciences to understand and manage global toxicants. The project is funded through the University of Oslo:Life Science and the Norwegian Ministry of Education and Research. Acknowledgements are directed to the Institute of Marine Science (IMS, Zanzibar), the National Institute for Medical Research (NIMR, Tanzania), and the Tanzanian Commission for Science and Technology (COSTECH, permit no. 2019-016-NA-2018-251) for ethical clearance, research permit and support. The authors wish to thank local farmers and landowners for help with egg and soil collection.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.chemosphere.2023.138646.

References

Abbasi, G., Buser, A.M., Soehl, A., Murray, M.W., Diamond, M.L., 2015. Stocks and flows of PBDEs in products from use to waste in the US and Canada from 1970 to 2020. Environ. Sci. Technol. 49 (3), 1521–1528. https://doi.org/10.1021/es504007v.

Abbasi, G., Li, L., Breivik, K., 2019. Global historical stocks and emissions of PBDEs. Environ. Sci. Technol. 53 (11), 6330–6340. https://doi.org/10.1021/acs.est.8b07032.

Bettina, H., Hermann, F., Wolfgang, V.l., Mehmet, C., 2011. Effects of chain length, chlorination degree, and structure on the octanol– water partition coefficients of polychlorinated n-alkanes. Environ. Sci. Technol. 45 (7), 2842–2849. https://doi.org/10.1021/es103098b.

Bogdal, C., Alsberg, T., Diefenbacher, P.S., MacLeod, M., Berger, U., 2015. Fast quantification of chlorinated paraffins in environmental samples by direct injection high-resolution mass spectrometry with pattern deconvolution. Anal. Chem. 87 (5), 2852–2860. https://doi.org/10.1021/ac504444d.

Breivik, K., Sweetman, A., Pacyna, J.M., Jones, K.C., 2002. Towards a global historical emission inventory for selected PCB congeners - a mass balance approach: 1. Global production and consumption. Sci. Total Environ. 290, 181–198. https://doi:10.1016/S0048-9697(01)01075-0.

Breivik, K., Gioia, R., Chakraborty, P., Zhang, G., Jones, K.C., 2011. Are reductions in industrial organic contaminants emissions in rich countries achieved partly by export of toxic wastes? Environ. Sci. Technol. 45 (21), 9154–9160. https://doi.org/10.1021/es202320c.

Breivik, K., Armitage, J.M., Wania, F., Sweetman, A.J., Jones, K.C., 2015. Tracking the global distribution of persistent organic pollutants accounting for e-waste exports to developing regions. Environ. Sci. Technol. 50 (2), 798–805. https://doi.org/10.1021/acs.est.5b04226.

Castro, M., Sobek, A., Yuan, B., Breitholtz, M., 2019. Bioaccumulation potential of CPs in aquatic organisms: uptake and depuration in daphnia magna. Environ. Sci. Technol. 53 (16), 9533–9541. https://doi:/10.1021/acs.est.9b01751.

Choo, G., Wang, W., Cho, H.-S., Kim, K., Park, K., Oh, J.-E., 2020. Legacy and emerging persistent organic pollutants in the freshwater system: relative distribution, contamination trends, and bioaccumulation. Environ. Int. 135, 105377 https://doi.org/10.1016/j.envint.2019.105377.

Clement, R.E., Reiner, E.J., Bhavsar, S.P., 2012. Organohalogen contaminants of emerging concern in Great Lakes fish: a review. Anal. Bioanal. Chem. 404 (9), 2639–2658. https://doi.org/10.1007/s00216-012-6300-4.

De Vries, M., Kwakkel, R.P., Kijlstra, A., 2006. Dioxins in organic eggs: a review. NJAS - Wageningen J. Life Sci. 54 (2), 207–221. https://doi.org/10.1016/S1573-5214(06)80023-0.

Dong, S., Li, X., Su, X., Wang, P., 2019. Concentrations and congener group profiles of short-and medium-chain chlorinated paraffins in animal feed materials. Sci. Total Environ. 647, 676–681. https://doi.org/10.1016/j.scitotenv.2018.08.017.

Du, X., Yuan, B., Zhou, Y., Zheng, Z., Wu, Y., Qiu, Y., Zhao, J., Yin, G., 2019. Tissue-specific accumulation, sexual difference, and maternal transfer of chlorinated paraffins in black-spotted frogs. Environ. Sci. Technol. 53 (9), 4739–4746. https://doi.org/10.1021/acs.est.8b06350.

ECHA, 2021. Inclusion of substances of very high concern in the candidate list for eventual inclusion in Annex XIV (decision of the European chemicals agency). https://echa.europa.eu/documents/10162/af3efea2-1518-3bbe-0bf5-3867131c2f4c. October 2022.

EFSA CONTAM Panel, Schrenk, D., Bignami, M., Bodin, L., Chipman, J.K., del Mazo, J., Grasl Kraupp, B., Hogstrand, C., Hoogenboom, L.R., Leblanc, J.C., Nebbia, C.S., Ntzani, E., Petersen, A., Sand, S., Schwerdtle, T., Vleminckx, C., Wallace, H., Bruschweiler, B., Leonards, P., Rose, M., Binaglia, M., Horvath, Z., Bordajandi, L.R., Nielsen, E., 2020. Scientific Opinion – risk assessment of chlorinated paraffins in feed and food, 2020 EFSA J. 18 (3), 220. https://doi.org/10.2903/j.efsa.2020.5991, 5991.

Gioia, R., Eckhardt, S., Breivik, K., Jaward, F.M., Prieto, A., Nizzetto, L., Jones, K.C., 2011. Evidence for major emissions of PCBs in the West African region. Environ. Sci. Technol. 45 (4), 1349–1355.

Glüge, J., Bogdal, C., Scheringer, M., Buser, A.M., Hungerbühler, K., 2013. Calculation of physicochemical properties for short-and medium-chain chlorinated paraffins. J. Phys. Chem. Ref. Data 42 (2), 023103. https://doi.org/10.1063/1.4802693.

Glüge, J., Wang, Z., Bogdal, C., Scheringer, M., Hungerbühler, K., 2016. Global production, use, and emission volumes of short-chain chlorinated paraffins–A minimum scenario. Sci. Total Environ. 573, 1132–1146. https://doi.org/10.1016/j.scitotenv.2016.08.105.

Glüge, J., Schinkel, L., Hungerbühler, K., Cariou, R., Bogdal, C., 2018. Environmental risks of medium-chain chlorinated paraffins (MCCPs): a review. Environ. Sci. Technol. 52 (12), 6743–6760. https://doi.org/10.1021/acs.est.7b06459.

Guan, K.-L., Liu, Y., Luo, X.-J., Zeng, Y.-H., Mai, B.-X., 2020. Short-and medium-chain chlorinated paraffins in aquatic organisms from an e-waste site: biomagnification and maternal transfer. Sci. Total Environ. 708, 134840 https://doi.org/10.1016/j.scitotenv.2019.134840.

Halse, A.K., Schlabach, M., Schuster, J.K., Jones, K.C., Steinnes, E., Breivik, K., 2015. Endosulfan, pentachlorobenzene and short-chain chlorinated paraffins in background soils from Western Europe. Environ. Pollut. 196, 21–28. https://doi.org/10.1016/j.envpol.2014.09.009.

Hansen, K.M., Fauser, P., Vorkamp, K., Christensen, J.H., 2020. Global emissions of dechlorane Plus. Sci. Total Environ. 742, 140677 https://doi.org/10.1016/j.scitotenv.2020.140677.

Houde, M., Muir, D.C., Tomy, G.T., Whittle, D.M., Teixeira, C., Moore, S., 2008. Bioaccumulation and trophic magnification of short-and medium-chain chlorinated paraffins in food webs from Lake Ontario and Lake Michigan. Environ. Sci. Technol. 42 (10), 3893–3899. https://doi.org/10.1021/es703184s.

Jari Oksanen, F.G.B., Friendly, Michael, Kindt, Roeland, Legendre, Pierre, McGlinn, Dan, Peter, R., Minchin, O'Hara, R.B., Simpson, Gavin L., Peter Solymos, M., Henry, H., Szoecs, Stevens Eduard, Wagner, Helene, 2019. Community ecology. Package. Version 2.5-6.

Knudtzon, N.C., Thorstensen, H., Ruus, A., Helberg, M., Bæk, K., Enge, E.K., Borgå, K., 2021. Maternal transfer and occurrence of siloxanes, chlorinated paraffins, metals, PFAS and legacy POPs in herring gulls (Larus argentatus) of different urban influence. Environ. Int. 152, 106478 https://doi.org/10.1016/j.envint.2021.106478.

Krätschmer, K., Malisch, R., Vetter, W., 2021. Chlorinated paraffin levels in relation to other persistent organic pollutants found in pooled human milk samples from primiparous mothers in 53 countries. Environ. Health Perspect. 129 (8), 087004 https://doi.org/10.1289/EHP7696.

Li, Z.-R., Luo, X.-J., Lin, L., Zeng, Y.-H., Mai, B.-X., 2021. Effect of laying sequence and selection of maternal tissues in assessment of maternal transfer of organohalogenated contaminants during chicken egg formation: a pilot study. Environ. Pollut. 270, 116157 https://doi.org/10.1016/j.envpol.2020.116157.

Mahenge, R.S., Ulanga, P., Malabeja, M., Pilly, J., Massoy, W., 2018. Performance Audit Report on Electronic Waste Management. URL: https://www.nao.go.tz/uploads/ELECTRONIC-WASTE-MANAGEMENT-2018.pdf. October 2022.

McGrath, T.J., Limonier, F., Poma, G., Bombeke, J., Winand, R., Vanneste, K., Andjelkovic, M., Van Hoeck, E., Joly, L., Covaci, A., 2021. Concentrations and distribution of chlorinated paraffins in Belgian foods. Environ. Pollut. 291, 118236 https://doi.org/10.1016/j.envpol.2021.118236.

Mézière, M., Marchand, P., Hutinet, S., Larvor, F., Baéza, E., Le Bizec, B., Dervilly, G., Cariou, R., 2021. Transfer of short-, medium-, and long-chain chlorinated paraffins to eggs of laying hens after dietary exposure. Food Chem., 128491 https://doi.org/10.1016/j.foodchem.2020.128491.

Moeckel, C., Breivik, K., Nøst, T.H., Sankoh, A., Jones, K.C., Sweetman, A., 2020. Soil pollution at a major West African E-waste recycling site: contamination pathways and implications for potential mitigation strategies. Environ. Int. 137, 105563 https://doi.org/10.1016/j.envint.2020.105563.

Nipen, M., Vogt, R.D., Bohlin-Nizzetto, P., Borgå, K., Mwakalapa, E.B., Borgen, A.R., Jørgensen, S.J., Ntapanta, S.M., Mmochi, A.J., Schlabach, M., 2022a. Spatial trends of chlorinated paraffins and dechloranes in air and soil in a tropical urban, suburban, and rural environment. Environ. Pollut. 292, 118298 https://doi.org/10.1016/j.envpol.2021.118298.

Nipen, M., Vogt, R.D., Bohlin-Nizzetto, P., Borgå, K., Mwakalapa, E.B., Borgen, A.R., Schlabach, M., Christensen, G., Mmochi, A.J., Breivik, K., 2022b. Increasing trends of legacy and emerging organic contaminants in a dated sediment core from East-Africa. Front. Environ. Sci. 711 https://doi.org/10.3389/fenvs.2021.805544.

Nevondo, V., Okonkwo, O.J., 2021. Status of short-chain chlorinated paraffins in matrices and research gap priorities in Afria: a review. Environ. Sci. Pollut. Control Ser. 28, 52844–52861. https://doi.org/10.1007/s11356-021-15924-w.

Petrlik, J., Bell, L., Beeler, B., Møller, M., Jopkova, M., Arisandi, P., Brabcova, K., Carcamo, M., Chávez Arce, S.C., Dizon, T., Ismawati Driwiega, Y., Kuenpouo, G., Mngànya, S., Ochieng Ochola, G., Skalsky, M., 2021. Plastic Waste Disposal Leads to Contamination of the Food Chain. Retrieved from International Pollutants Elimination Network (IPEN). URL: https://ipen.org/sites/default/files/documents/ipen-plastic-waste-contamination-full-en.pdf. October 2022.

Petrlik, J., Bell, L., Digangi, J., Allo'o, S.M.a.O., Kuepouo, G., Ochola, G.O., Grechko, V., Jelinek, N., Strakova, J., Skalsky, M., 2022. Monitoring dioxins and PCBs in eggs as sensitive indicators for environmental pollution and global contaminated sites and recommendations for reducing and controlling releases and exposure. Emerging Contaminants 8, 254–279. https://doi.org/10.1016/j.emcon.2022.05.001.

Polder, A., Müller, M., Brynildsrud, O., De Boer, J., Hamers, T., Kamstra, J., Lie, E., Mdegela, R., Moberg, H., Nonga, H., 2016. Dioxins, PCBs, chlorinated pesticides and brominated flame retardants in free-range chicken eggs from peri-urban areas in Arusha, Tanzania: levels and implications for human health. Sci. Total Environ. 551, 656–667. https://doi.org/10.1016/j.scitotenv.2016.02.021.

Ringo, E.J., Lekule, F.P., 2020. Market trends and consumer behaviors and preferences in the Tanzanian poultry subsector. https://dutchafricapoultry.com/wp-content/uploads/2021/10/Poultry-Study-Tanzania-report-8-July-2020.pdf. December 2022.

Schuster, J.K., Harner, T., Sverko, E., 2020. Dechlorane Plus in the global atmosphere. Environ. Sci. Technol. Lett. 8 (1), 39–45. https://doi.org/10.1021/acs.estlett.0c00758.

Shen, L., Reiner, E.J., MacPherson, K.A., Kolic, T.M., Helm, P.A., Richman, L.A., Marvin, C.H., Burniston, D.A., Hill, B., Brindle, I.D., 2011. Dechloranes 602, 603, 604, dechlorane Plus, and chlordene Plus, a newly detected analogue, in tributary sediments of the laurentian great lakes. Environ. Sci. Technol. 45 (2), 693–699. https://doi.org/10.1021/es1027844.

Tomy, G.T., Stern, G.A., Muir, D.C., Fisk, A.T., Cymbalisty, C.D., Westmore, J.B., 1997. Quantifying C10− C13 polychloroalkanes in environmental samples by high-resolution gas chromatography/electron capture negative ion high-resolution mass spectrometry. Anal. Chem. 69 (14), 2762–2771. https://doi.org/10.1021/ac961244y.

Ueberschär, K.H., Dänicke, S., Matthes, S., 2007. Dose–response feeding study of short chain chlorinated paraffins (SCCPs) in laying hens: effects on laying performance and tissue distribution, accumulation and elimination kinetics. Mol. Nutr. Food Res. 51 (2), 248–254. https://doi.org/10.1002/mnfr.200600168.

UNEP, 2017. Stockholm convention on persistent organic pollutants. Annex A, decision SC-8/11. http://chm.pops.int/Implementation/Alternatives/AlternativestoPOPs/ChemicalslistedinAnnexA/ShortchainchlorinatedparaffinsSCCPs/tabid/5986/Default.aspx. January 2021.

Vorkamp, K., Balmer, J., Hung, H., Letcher, R.J., Rigét, F.F., 2019. A review of chlorinated paraffin contamination in Arctic ecosystems. Emerging Contaminants 5, 219–231. https://doi.org/10.1016/j.emcon.2019.06.001.

Waegeneers, N., De Steur, H., De Temmerman, L., Van Steenwinkel, S., Gellynck, X., Viaene, J., 2009. Transfer of soil contaminants to home-produced eggs and preventive measures to reduce contamination. Sci. Total Environ. 407 (15), 4438–4446. https://doi.org/10.1016/j.scitotenv.2008.12.041.

Wang, Y., Li, J., Cheng, Z., Li, Q., Pan, X., Zhang, R., Liu, D., Luo, C., Liu, X., Katsoyiannis, A., 2013. Short-and medium-chain chlorinated paraffins in air and soil of subtropical terrestrial environment in the Pearl River Delta, South China: distribution, composition, atmospheric deposition fluxes, and environmental fate. Environ. Sci. Technol. 47 (6), 2679–2687. https://doi.org/10.1021/es304425r.

Wang, P., Zhang, Q., Zhang, H., Wang, T., Sun, H., Zheng, S., Li, Y., Liang, Y., Jiang, G., 2016. Sources and environmental behaviors of Dechlorane Plus and related compounds—a review. Environ. Int. 88, 206–220. https://doi.org/10.1016/j.envint.2015.12.026.

Warner, N.A., Sagerup, K., Kristoffersen, S., Herzke, D., Gabrielsen, G.W., Jenssen, B.M., 2019. Snow buntings (Plectrophenax nivealis) as bio-indicators for exposure differences to legacy and emerging persistent organic pollutants from the Arctic terrestrial environment on Svalbard. Sci. Total Environ. 667, 638–647. https://doi.org/10.1016/j.scitotenv.2019.02.351.

Wei, G.-L., Liang, X.-L., Li, D.-Q., Zhuo, M.-N., Zhang, S.-Y., Huang, Q.-X., Liao, Y.-S., Xie, Z.-Y., Guo, T.-L., Yuan, Z.-J., 2016. Occurrence, fate and ecological risk of chlorinated paraffins in Asia: a review. Environ. Int. 92, 373–387. https://doi.org/10.1016/j.envint.2016.04.002.

White, K.B., Kalina, J.Í., Scheringer, M., PrIbylová, P., KukučKa, P., Kohoutek, J.Í., Prokes, R., KláNová, J., 2020. Temporal trends of persistent organic pollutants across Africa after a decade of MONET passive air sampling. Environ. Sci. Technol. 55 (14), 9413–9424. https://doi.org/10.1021/acs.est.0c03575.

Wong, F., Hung, H., Dryfhout-Clark, H., Aas, W., Bohlin-Nizzetto, P., Breivik, K., Mastromonaco, M.N., Lundén, E.B., Ólafsdóttir, K., Sigurðsson, Á., 2021. Time trends of persistent organic pollutants (POPs) and Chemicals of Emerging Arctic Concern (CEAC) in Arctic air from 25 years of monitoring. Sci. Total Environ. 775, 145109 https://doi.org/10.1016/j.scitotenv.2021.145109.

Wu, J.P., Luo, X.J., Zhang, Y., Chen, S.J., Mai, B.X., Guan, Y.-T., Yang, Z.Y., 2009. Residues of polybrominated diphenyl ethers in frogs (Rana limnocharis) from a contaminated site, South China: tissue distribution, biomagnification, and maternal transfer. Environ. Sci. Technol. 43 (14), 5212–5217. https://doi.org/10.1021/es901103y.

Xu, C., Zhang, Q., Gao, L., Zheng, M., Qiao, L., Cui, L., Wang, R., Cheng, J., 2019. Spatial distributions and transport implications of short-and medium-chain chlorinated paraffins in soils and sediments from an e-waste dismantling area in China. Sci. Total Environ. 649, 821–828. https://doi.org/10.1016/j.scitotenv.2018.08.355.

Yhdego, Michael, 2017. From a sanitary landfill to a dump site: Pugu Kinyamwezi community curse in Dar es Salaam, Tanzania. ERC—Environmental Resources Consultancy. https://doi.org/10.13140/RG.2.2.27828.04483.

Zacs, D., Perkons, I., Abdulajeva, E., Pasecnaja, E., Bartkiene, E., Bartkevics, V., 2021. Polybrominated diphenyl ethers (PBDEs), hexabromocyclododecanes (HBCDD), dechlorane-related compounds (DRCs), and emerging brominated flame retardants (EBFRs) in foods: the levels, profiles, and dietary intake in Latvia. Sci. Total Environ. 752, 141996 https://doi.org/10.1016/j.scitotenv.2020.141996.

Zeng, Y.H., Huang, C.C., Luo, X.J., Liu, Y.N., Ren, Z.H., Mai, B.X., 2018. Polychlorinated biphenyls and chlorinated paraffins in home-produced eggs from an e-waste polluted area in South China: occurrence and human dietary exposure. Environ. Int. 116, 52–59. https://doi.org/10.1016/j.envint.2018.04.006.

Zhang, C., Chang, H., Wang, H., Zhu, Y., Zhao, X., He, Y., Sun, F., Wu, F., 2019. Spatial and temporal distributions of short-, medium-, and long-chain chlorinated paraffins in sediment cores from nine lakes in China. Environ. Sci. Technol. 53 (16), 9462–9471. https://doi.org/10.1021/acs.est.8b07296.

Zheng, S., Wang, P., Sun, H., Matsiko, J., Hao, Y., Meng, D., Li, Y., Zhang, G., Zhang, Q., Jiang, G., 2018. Tissue distribution and maternal transfer of persistent organic pollutants in Kentish Plovers (Charadrius alexandrines) from Cangzhou Wetland, Bohai Bay, China. Sci. Total Environ. 612, 1105–1113. https://doi.org/10.1016/j.scitotenv.2017.08.323.

Zheng, X.B., Wu, J.P., Luo, X.J., Zeng, Y.H., She, Y.Z., Mai, B.X., 2012. Halogenated flame retardants in home-produced eggs from an electronic waste recycling region in South China: levels, composition profiles, and human dietary exposure assessment. Environ. Int. 45, 122–128. https://doi.org/10.1016/j.envint.2012.04.006.

Supplementary information

For

Chlorinated paraffins and dechloranes in free-range chicken eggs and soil around waste disposal sites in Tanzania

Contains:

Table S2. Concentrations, chlorination degree (Cl%), MCCP/SCCP ratio and recovery % of chlorinated paraffins in egg samples. Match between sample and standard pattern used in deconvolution method for CP quantification is assessed by goodness of fit (R^2).

ID	Location	Lipid %	SCCPs ng/g ww	SCCP Cl%	SCCP R^2	MCCPs ng/g ww	MCCP Cl%	MCCP R^2	MCCP / SCCP	Recovery (%)
CH05	Official	8.3	NA	NA	NA	NA	NA	NA	NA	NA
CH06	DAR	4.7	37.2	60.6	0.94	110	57.2	0.96	3.0	74
CH07		8.9	32	60.7	0.91	93.4	57.5	0.96	2.9	73
CH08		8.4	49	60.9	0.94	73	56.2	0.98	1.5	81
CH09		9.3	34.7	60.5	0.95	56.7*	55.4	0.96	1.6	85
CH10		13.3	38.7	60.7	0.94	75.7	56.3	0.96	2.0	74
MT05	Historical	9.2	22.5	59.6	0.95	89.1	54.7	0.93	4.0	67
MT06	DAR	5.7	11.1*	58.3	0.89	48.3*	54.6	0.99	4.3	72
MT07		12.3	115.4	63.8	0.96	305	59.9	0.13**	2.6	61
MT08		9.5	10*	58.3	0.78	59.8	55.5	0.98	6.0	70
MT09		12.0	9.2*	59.8	0.83	66.1	55.5	0.95	7.2	97
MT10		11.5	79.5	61.6	0.93	191	58.3	0.33**	2.4	68
EGG2	Informal	11.2	16.1*	60.2	0.92	74.8	54.8	0.97	4.6	60
EGG3	ZNZ	12.6	13.3*	60.8	0.94	72.9	54.5	0.97	5.5	58
EGG4		10.6	6.3*	59.4	0.71	69.7	51.8	0.94	11.1	68
EGG5		12.2	9.8*	59.8	0.95	58.3*	53.8	0.95	6.0	130
EGG6		11.9	11.2*	59.3	0.82	64.5*	54.4	0.92	5.8	64
EGG8		11.1	10.3*	57.4	0.89	46*	55.3	0.96	4.5	75
FRF1	Reference	10.3	27.5	58.5	0.96	127	53.9	0.98	4.6	71
FRF2	ZNZ	14.1	18.9	58.2	0.96	70.1	53.6	0.95	3.7	68
FRF3		14.5	56.8	60.6	0.93	109	56.9	0.91	1.9	75
FRF4		13.8	34.5	60.6	0.95	54.5*	55.7	0.97	1.6	79
FRF5		18.5	26.7	61.0	0.93	67.4	55.5	0.95	2.5	76
FRF6		12.0	NA	NA	NA	NA	NA	NA	NA	NA

NA indicates that sample was lost during sample preparation and thus not quantified for CPs. * indicates that sample has been quantified, but is below the limit of detection, set at 16.2 and 65 ng/g ww for SCCPs and MCCPs, respectively. ** Goodness of fit (R^2) between sample and standard pattern below 0.5 meaning that these values are associated with higher uncertainty than the rest of the samples

Table S3. Concentrations of dechloranes in egg samples among the different locations. Two radiolabeled congeners were used for recovery calculations: Dechlorane 602 and Dechlorane Plus syn

ID	**Location**	**Lipid %**	**Dech 602 ng/g ww**	**Dech 603**	**DP *syn***	**DP *anti***	**Recovery (%)** Dech 602, DP *syn*
CH05	Official	8.3	0.01*	0.01*	0.018*	0.03	117,99
CH06	DAR	4.7	0.01*	0.01*	0.018*	0.04	121,101
CH07		8.9	0.01*	0.01*	0.018*	0.05	107, 93
CH08		8.4	0.01*	0.01*	0.018*	0.05	153, 113
CH09		9.3	0.01*	0.01*	0.021	0.03	132, 105
CH10		13.3	0.01*	0.01*	0.018*	0.04	123, 120
MT05	Historical	9.2	0.01*	0.16	0.03	0.08	108, 101
MT06	DAR	5.7	0.01*	0.13	0.02	0.06	102, 95
MT07		12.3	0.01*	0.12	0.19	0.39	103, 95
MT08		9.5	0.01*	0.25	0.03	0.11	120, 99
MT09		12.0	0.01*	0.10	0.02	0.06	136,111
MT10		11.5	0.01*	0.10	0.13	0.45	114, 100
EGG2	Informal	11.2	0.022	0.26	0.09	0.30	99, 93
EGG3	ZNZ	12.6	0.020	0.26	0.06	0.21	103, 95
EGG4		10.6	0.015	0.19	0.04	0.13	86, 82
EGG5		12.2	0.020	0.24	0.06	0.19	95, 86
EGG6		11.9	0.019	0.27	0.10	0.39	113, 97
EGG8		11.1	0.021	0.25	0.10	0.35	131, 106
FRF1	Reference	10.3	0.01*	0.03	0.03	0.06	120, 116
FRF2	ZNZ	14.1	0.01*	0.06	0.02	0.04	131, 109
FRF3		14.5	0.01*	0.89	0.06	0.24	143. 110
FRF4		13.8	0.01*	0.05	0.018*	0.60	154, 122
FRF5		18.5	0.01*	0.09	0.02	0.07	145, 113
FRF6		12.0	0.01*	0.13	0.02	0.07	165, 128

*Indicates that sample is not detected above LOD

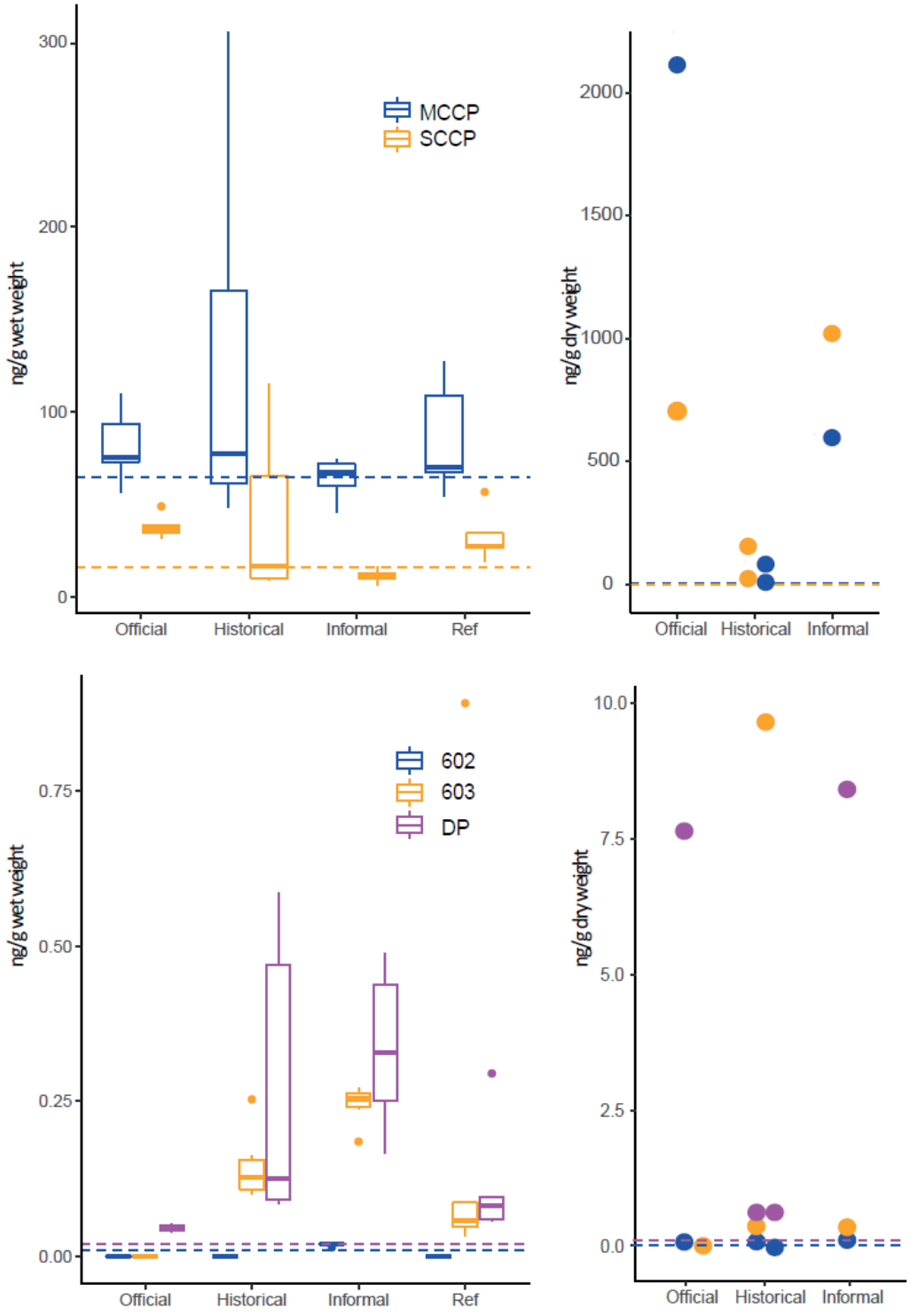


Figure S1. Concentrations of chlorinated paraffins (above) and dechloranes (below) in eggs (ng/g ww) and soil (ng/g dw). Detection limits (LODs)are indicated with dashed lines.