OCCURRENCE AND POTENTIAL HEALTH RISKS OF INDICATOR POLYCHLORINATED BIPHENYLS IN SELECTED MARINE FISH SPECIES: A CASE STUDY OF DAR ES SALAAM

A Dissertation Submitted in Partial Fulfilment of the Requirements for the Degree of Master of Science in Food Quality and Safety Assurance

By

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EXTENDED ABSTRACT

Polychlorinated biphenyls (PCBs) are industrial chemicals in a group of persistent organic pollutants (POPs) that are ubiquitous in environments including water bodies. Ocean sediments are one of the major reservoirs of PCBs, contaminated through runoff water, atmospheric transfer, and sewer discharge of industrial and municipal wastes, risking contaminating aquatic organisms. Fish are vulnerable to contamination because PCBs are highly lipophilic and can easily be absorbed and stored in fish fatty tissues. However, there are limited studies on the marine fish contamination status in Tanzania's coastal waters.

The overall objective of the present study overall objective was to assess the occurrence and levels of indicator PCBs in selected species of marine fish in Dar es Salaam, and the specific objectives were to determine indicator PCBs levels in mackerel fish (*Rastrelliger kanagutra*) and rabbitfish (*Siganus sotur*) and to assess the dietary exposure and health risks of PCBs to artisanal fishermen and fish vendors.

Fish samples were randomly collected from fishermen and fish vendors at Kivukoni Feri fish market where a total of 60 fresh fish samples were collected from two different species, *Rastrelliger kanagutra,* and *Siganus sotur*, chosen based on the prior conducted food consumption survey. A food frequency survey was conducted among fishermen and fish vendors to assess the fish consumption habit of the respondents. PCBs extraction and sample matrix clean-up were done using the QuEChERS method and quantification analysis of PCBs was done using a Gas Chromatograph tandem Mass Spectrometer (GC-MS/MS) at the Government Chemist Laboratory Authority (GCLA) laboratories.

Results showed the presence of indicator PCBs in both fish species, with 73.3% of *R. kanagutra* samples and 20% of *S.sutor* samples showing PCBs contamination. The levels of the six indicator PCBs were relatively low in both fish species with the highest mean concentration ($\sum_6$PCBs), 3.71 ngg^{-1} w/w observed in *R. kanagutra*. The individual mean levels of detected indicator PCBs ranged from <LOD to 1.15 ngg^{-1} w/w, while the sum of the six indicator PCBs ($\sum_6$PCBs) levels ranged from 0.12 to 3.71 ngg^{-1} w/w. PCBs contamination was dominated by the three congeners PCB 138, 153, and 180, while PCBs 28, 52, and 101 levels were below the limit of quantitation (<LOD).

The finding of this study indicates PCBs contamination of fish in the study area due to the historical release of PCBs. *R. kanagutra* were found to have significantly higher PCBs contamination levels than the *S. sutor* fish species due to differences in trophic levels and feeding habits. However, detected PCBs levels in both fish species were below the maximum residue limits (MRL) of 75 ngg^{-1} w/w set by the European Commission (UC).

The potential human health risks as a result of PCBs contaminated fish consumption were assessed for the general population for both children and adults. Estimated lifetime cancer risks were very low ranging from 5.14E-09 to 6.14E-07 for both children and adults, below the recommended health guidelines of <10E-6 for adults and <10E-04 for children. Estimated non-carcinogenic risks hazard indices were far less than the critical value of 0.1, ranging from 1.95E-06 to 3.26E-05 for both children and adults, indicating that consumption of the *R.kanagutra* and *S. sutor* fish species from the Kivukoni Feri fish market would have negligible adverse health effects to the general population.

The results of the fish consumption survey showed that *R. kanagutra* and *S. sutor* were the most preferred fish for consumption among fishermen and fish vendors due to their availability in the daily catch and lower prices and therefore more affordable to be used as a meal. Moreover, fishermen and fish vendors were found to consume an average amount of 400.5 gday^{-1} of fish which were higher than the national per capita fish consumption of 8.22 kg (23 gday^{-1}) and global per capita consumption of 20.5 kg (56 gday^{-1}).

Dietary exposure and potential health risk of PCBs to artisanal fishermen and fish vendors were estimated using the fish consumption rate obtained in the initial food frequency survey and compared to that of the general population estimated using national per capita fish consumption. The average intake of total PCBs by fishermen and fish vendors was higher in comparison to that of the general population ranging between 5.10 and 27.56 ngkg^{-1} bw.day^{-1} for normal and higher fish consumers depending on the estimation scenario, while that of the general population ranged between 0.14 and 0.31 ngkg^{-1} bw.day^{-1} in lower and upper estimation scenario respectively. The average daily intake of total PCBs by fishermen and fish vendors exceeded the recommended tolerable daily intake, (TDI) of 20 ngkg^{-1} bw.day^{-1} in high fish consumers while the estimated lifetime carcinogenic and non-carcinogenic hazard indices exceeded the recommended health guidelines of <10E-6 and 0.1 respectively, raising serious concerns for potential adverse health effects in long run. However, the estimated average daily intake of total PCBs for the general population was far below the recommended guidelines for an average intake of total PCBs and the carcinogenic and non-carcinogenic hazard indices estimation was also far below the recommended health guideline of <10E-6 and 0.1 respectively.

PCBs exposure through the intake of fish was observed to be a course of serious concern for fishermen and fish vendors, due to exposure to high levels of PCBs that exceeded recommended health guidelines for tolerable daily intake.

To safeguard the most vulnerable fish consumers from PCBs exposure, the development of cost-effective strategies to minimize the accumulation of PCBs in aquatic ecosystems and environments is required. Creation of awareness among fish consumers on the potential adverse health risks of PCBs and the different ways to minimize exposure such as species diversification to reducing consumption of fish species that are more vulnerable to PCBs contamination such as fatty fish, carnivorous and omnivorous fish while consuming more of herbivorous and low-fat fish species.

DECLARATION

I, Gabriel Jacob Gabriel, do hereby declare to the Senate of Sokoine University of Agriculture that this dissertation is my own original work done within the period of registration and that it has neither been submitted nor being concurrently submitted in any other institution.

Gabriel Jacob Gabriel Date
(MSc. Food Quality and Safety Assurance)

The above declaration is confirmed;

Dr. Beatrice M. Kilima Date
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DEDICATION

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LIST OF ABREVIATIONS

AT	Average time per year
ATSDR	Agency for Toxic Substances and Disease Registry
BW	Body weight
C	Contaminant concentration
CADI	Chronic average daily intake
CDI	Chronic Daily Intake
C_R	Lifetime cancer risks
Cm	Centimeter
EC	European Commission
ED	Exposure duration
EF	Exposure frequency
EFSA	European Food Safety Authority
EU	European Union
FAO	Food and Agriculture organization of the United Nation
$FeCl_2$	Iron (II) Chloride
g	Grams
GCLA	Government Chemist Laboratory Authority
GC-MS/MS	Gas Chromatography Mass Spectrometer
HI	Hazard index
HQ	Hazard quotient
iPCBs	Indicator polychlorinated biphenyls
IPEP	The International POPs Elimination Project
IR	Fish consumption rate
Kg	Kilogram
LB	Lower bound
LOD	Limit of detection
LOD	Limit of quantitation
M	Meter
$MgSO_4$	Magnesium Sulphate
Min	Minutes
Ml	Milliliter
MLF	Ministry of Livestock and Fisheries
Mm	Millimeter
MRL	Maximum Residue Limit
MRM	Multiple reaction monitoring
NEMC	National Environmental Management Council
Ng	Nanogram
PCBs	Polychlorinated biphenyls
POPs	Persistent organic pollutants
Ppm	Parts per million
PSA	Primary secondary amine
QuEChERS	Quick, Easy, Cheap, Effective, Rugged, Safe
rpm	Revolution per minute
RfD	Reference dose
SF	Oral cancer slope
SPSS	Statistical package for social science
TDI	Tolerable daily intake
UB	Upper bound

URT/NIP	United Republic of Tanzania - National Implementation Plan
USEPA	United State Environmental Protection Agency
WHO	World Health Organization
WIO	West Indian Ocean
w/w	Wet weight
^{0}C	Degree Centigrade
µL	Microliter
µm	Micrometer

LIST OF MANUSCRIPTS

The dissertation is based on two manuscripts:

1. Gabriel J.G.M, and Kilima B.M. (2022). Occurrence and potential health risks of indicator polychlorinated biphenyls in selected marine fish species (*Rastrelliger kanagutra*) and rabbitfish (*Siganus sotur*). A case of Dar es Salaam. *Accepted for publication in International Journal of Agriculture, Environment and Bioresearch*

2. Gabriel J.G.M, and Kilima B.M. (2022). Dietary exposure and risk assessment of polychlorinated biphenyls to artisanal fishermen and fish vendors. A case study of Dar es Salaam. (*In press*).

CHAPTER ONE

1.0 General introduction

1.1 Background Information

1.1.1 Polychlorinated biphenyls (PCBs)

Polychlorinated biphenyls (PCBs) are synthetic chlorinated hydrocarbon compounds that were widely used worldwide until their commercial manufacture was banned by most countries from the late 1970s to 1980s (Montano *et al.,* 2022; Shi *et al.* 2019; Wu *et al.,* 2020). PCBs are odourless, tasteless, bright yellow, and can be in oil or solid form (Munawar *et al.,* 2021). They are characterized by their persistence in the environment, resistance to degradation, long-range atmospheric transport (Mwakalapa *et al.,* 2018; Romanic *et al.,* 2018; Aslam *et al.,* 2019), and high lipophilic nature that gives them the ability to accumulate in fatty tissues of animals and into the environment over time (Lin *et al.,* 2020; Bandow *et al.,* 2020; Umasangaji *et al.,* 2020; Nardelli *et al.,* 2019). PCBs are aromatic compounds made of two benzene rings joined together by a single carbon bond (Figure 1.1) (Wenaty *et al.,* 2019a).

Polychlorinated biphenyls (PCBs) are manufactured from the direct chlorination of biphenyls with $FeCl_2$ used as a catalyst and can also be produced as a by-product of other chemical processes (Munawar *et al.,* 2021; Montano *et al.,* 2022). There are possible 209 derivatives of PCBs known as congeners depending on the number of chlorine atoms they have, as each congener contains one to ten chlorine atoms (Mwakalapa *et al.,* 2018; Wenaty *et al.,* 2019a; WHO, 2016) and are identified by the number of chlorine atoms attached to the benzene rings and the position at which they are attached (Munawar *et al.,* 2021; WHO, 2016).

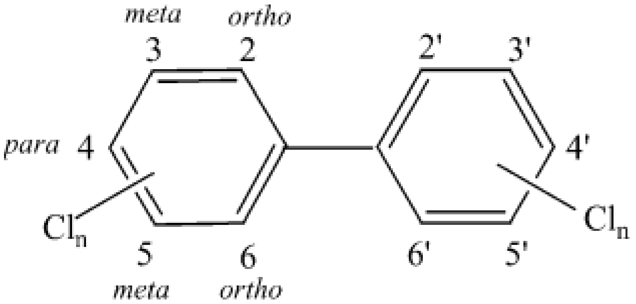

Figure 1.1: Basic structure of polychlorinated biphenyls
(Source: Munawar *et al.* (2021)

The Stockholm convention recommended six out of the 209 PCB congeners (PCB 28, PCB 52, PCB 101, PCB 138, PCB 153, and PCB 180) to be used as indicators in the monitoring of PCBs contamination in environments due to their abundance of the environment (Nardelli *et al.,* 2019; WHO, 2016; Bandow *et al.,* 2020). Before the ban on production and use, PCBs were widely used in various products such as transformer oils, lubricants, heat exchangers, capacitors, flame retardants, and as an additive in sealants, paints, adhesives, and other products (Obanya *et al.,* 2019; Mwakalapa *et al.,* 2018; Munawar *et al.,* 2021; Hulin *et al.,* 2020; Wenaty *et al.,* 2019a; Montano *et al.,* 2022; Shi *et al.* 2019; URT/NIP, 2018).

1.1.2 Presence and source of polychlorinated biphenyl (PCBs) in Tanzania

Tanzania imported and used products containing polychlorinated biphenyls such as transformers oil, capacitors, coolants, fire retardants, sealants, paints, and other products before they were prohibited (URT/NIP, 2018). Despite the importation ban of PCBs contained products, studies (Mahugija, 2015; Hellar-Kihampa, *et al.,* 2013; Polder *et al.,* 2014, *et al.,* 2019a; Mwakalapa *et al.,* 2018) have documented the presence of PCBs and other persistent organic pollutants (POPs) contamination in various location in Tanzania more than a decade after the ban. Furthermore, URT/NIP (2018) and IPEP (2005) reported that the highly contaminated area were sites with electrical transformers installations, storage, and maintenance sites (Table 1.1). IPEP (2005), found 30 highly contaminated sites (hot spots) across various locations in Tanzania, with 11 of the sites identified as heavily contaminated and located close to water bodies, residences, and other human activities (URT/NIP, 2018; IPEP, 2005).

Table 1.1: Sites heavily contaminated with PCBs in Tanzania

No.	Site	Priority	No.	Site	Priority
1	Dar es Salaam – DIA	1	16	Ubungo Thermal Power Station	2
2	DAWASA Lower Ruvu – Bagamoyo	1	17	Zanzibar – Mtoni Sub Station	2
3	DAWASA Upper Ruvu – Mlandizi intake	1	18	Zanzibar – Wesha Power Station	2
4	Kikuletwa Mini Hydro	1	19	Zanzibar – Tibirizi Power Station	2
5	Mbalizi Minihydro Station – Mbeya	1	20	Dar es Salaam City Centre substation	3
6	Morogoro Tanneries Industry	1	21	Gongo la Mboto Substation	3
7	Nyumba ya Mungu	1	22	Kidatu Switch Yard	3
8	Old Pangani Hydro	1	23	Mlandizi substation (TANESCO)	3
9	TANESCO Electrical Workshop Ubungo	1	24	Musoma – Power Station	3
10	Tosamaganga Mini Hydro Iringa	1	25	MUTEX - Musoma	3
11	Water Pump – Kibaha District	1	26	MWATEX – Mwanza	3
12	Bukoba - Sadelmi Store	2	27	SPM Mills CO.Ltd Mgololo	3
13	Kidatu Hydro Plant	2	28	TAZARA Workshop – Mbeya	3
15	Kilombero 2	2	29	Tumbi Sub-station (TANESCO)	3
15	Morogoro Msamvu Office sub – Station	2	30	Ubungo Control Centre 33/11 S/yard	3

Source: International POPs Elimination Project, IPEP, (2005).

However, Montano *et al.* (2022), reported that air from industrialized urban areas and indoor air in buildings constructed using PCB-containing sealants, paints, and other PCBs containing building materials have been often overlooked as another source of PCBs. The reported presence of PCBs contamination around the country indicates that the importation ban of equipment such as electrical transformers and other products containing PCBs has not affected the release of PCBs into the environment as prior imported PCBs-containing products are still being used and others are in storage around the country (IPEP, 2005; URT/NIP, 2005, 2018).

1.1.3 Contamination pathway of polychlorinated biphenyl (PCBs) in the food chain ecosystem

Polychlorinated biphenyls (PCBs) contamination pathways into environments involve improper handling of the PCB-containing products during use, accidental releases such as spillage during equipment maintenance and leakage following wear and tear or corrosion of the equipment at installation and storage sites, unintentional production as by-products during the manufacture of other products such as dyes and paint (Munawar *et al.,* 2021; IPEP, 2005; Mahugija, 2015; Umasangaji *et al.,* 2020). Improper disposal methods of PCBs containing wastes in landfills and other waste dump sites, contribute to the spread of contamination of PCBs into underground water due to leaching, and emissions of volatile and semi-volatile PCB compounds into the atmosphere from uncontrolled open burning of PCBs containing wastes (Umasangaji *et al.,* 2020). Furthermore, PCBs contaminations in environments are contributed by the misuse and inappropriate use of PCB-containing oils such as used in oil in welding machines and as burning oil in boilers, and also its use in cooking/frying oil for profit maximization due to its thermal stability properties (IPEP, 2005; Wenaty *et al.,* 2019a).

Polychlorinated biphenyls (PCBs) from contaminated areas are slowly and continuously released into the environment (Jafarabadi *et al.,* 2019; Obanya *et al.,* 2019; Montano *et al.,* 2022). From contaminated areas, PCBs reach vital areas of food production such as rivers, lakes, and oceans where they end up accumulating mostly in sediments, water, and other fish feeding grounds via runoff water and sewage discharges (Ibeto *et al.,* 2019; Obanya *et al.,* 2019). Activities such as shipping and ship dismantling can also lead to PCB contamination through the leakage of PCBs contaminated oil directly into the water (Jafarabadi *et al.,* 2019). In the ocean, sediment accumulates a large proportion of PCBs and eventually becomes a source of PCBs exposure to aquatic organisms as it is slowly released into water over time (Jafarabadi *et al.,* 2019). Living organisms such as fish in the ocean and other water bodies have a higher chance of accumulating high levels of PCBs by bioconcentration and/or biomagnification through water, feeding on phytoplankton, seagrasses and consumption of other small lifeforms such as anchovies, ostracods and cladocerans (Fishbase, 2017; Hakimelahi *et al.,* 2020; Jafarabadi *et al.,* 2019).

1.1.4 Exposure of fish to PCBs contamination

Fish become exposed to PCBs either directly from water or through feeding on other smaller life forms and plants (Umasangaji *et al.,* 2020). Lower trophic level fish in polluted water bodies bioconcentrate PCBs through absorption of PCBs in water through their dermal tissues or gills and feeding on other contaminated feed (Jafarabadi *et al.,* 2019; Umasangaji *et al.,* 2020) while high trophic levels fish accumulate PCBs by feeding on other contaminated smaller fish and other life-forms which make high trophic fish more exposed to PCBs because of biomagnification (Panseri *et al.,* 2019; Montano *et al.,* 2022; Fishbase, 2017). In fish, PCBs accumulate in fatty tissues, mainly in the livers and kidneys (Jafarabadi *et al.,* 2019). Because of the lipophilic nature of PCBs, fatty fish such as salmon, have a higher concentration of PCBs (Duedahl-Olesen *et al.,* 2020; Montano *et al.,* 2022; Mwakalapa *et al.,* 2018). Habitation of the fish also contributes to the exposure of fish to PCBs as most of the fish residing in coastal shallow waters tend to have a higher amount of PCBs contamination due to the high contamination of coastal areas closer to waste and sewage outlets (Umasangaji *et al.,* 2020; FAO, 2014).

1.1.5 Management and regulation of PCBs in Tanzania

Control, management, and regulation of PCBs in Tanzania are done through various laws and regulations including the Industrial and Consumer Chemical (Management and Control) Act No. 3 of 2003. The Act is implemented under the Ministry of Health, through Government Chemist Laboratory Authority (GCLA). The Act control importation, production, transportation, distribution, storage, use, disposal, and all aspects related to industrial chemicals. The Act also includes provisions for strict restriction and phasing out of PCBs, decommissioning of facilities, and remediation of the contaminated area. The Environmental Management Act: EMA of 2004 implemented under the National Environmental Management Council (NEMC) under the Vice President Office (VPO), provides principles for environmental management of impacts, risk assessments, pollution prevention, waste management, and regulation of persistent organic pollutants (POPs) (URT/NIP, 2018).

Tanzania has also entered International agreements in efforts to manage and eventually eliminate hazardous chemicals and persistent organic pollutants (POPs) including PCBs. Tanzania ratified the Stockholm Convention on persistent organic pollutants in 2004 and began its national implementation plan with inventory surveys in 2003 and 2004, then follow-up inventory surveys in 2014 through the Division of Environment, Vice Presidents Office (Polder *et al.,* 2014; URT/NIP, 2018; Senyangwa and Persson, 2014). Other agreements entered include the Rotterdam Convention on the prior informed procedure for certain hazardous chemicals and pesticides in International trade ratified in 2002, the Basel Convention on the control of the Transboundary Movements of Hazardous Waste and their Disposal ratified in 1993 and the Montreal Protocol for the Protection of the Ozone Layer also ratified in 1993 (Senyangwa and Persson, 2014).

1.1.6 Human exposure and adverse health effects associated with polychlorinated biphenyl (PCBs)

The major source of human exposure to PCBs is through food consumption (Ravenscroft and Schell, 2018; WHO, 2016), while other sources of exposure include volatile airborne PCBs in the surrounding atmosphere, dust in industrial complexes and homes or office rooms decorated with PCBs containing paint and sealants (Montano *et al.,* 2022). Exposure to PCBs through food intake is higher through consumption of food of animal origin than other types of food (Duedahl-Olesen *et al.,* 2020; Obanya *et al.,* 2019). Fish and other marine food products contribute more to human exposure to PCBs as they are highly contaminated with PCBs compared to other animal-source food with higher fat content (Jafarabadi *et al.,* 2019).

Once in the human body, PCBs accumulate in the fatty tissue (adipose tissues) because of their high lipophilic nature (WHO, 2016). A working group categorized PCBs as carcinogenic to humans (Group 1) (Hulin *et al.,* 2020), and studies by Montano *et al.* (2022), Lin *et al.* (2020), Muller *et al.* (2017), and Panseri *et al.* (2019) reported that PCBs are the probable cause of the increased risk of prostate, liver and breast cancer, endocrine system disruption, chloracne, dermal lesion, and developmental effects in breastfed infants. Furthermore, Munawar *et al.,* (2021) reported that PCBs have been shown to have effects on genes such as gene mutation and chromosomal loss when they interact with other chemicals, and also effects on reproductive system impairment such as poor sperm quality and miscarriage (Quijano *et al.,* 2018; Hulin *et al.,* 2020).

1.1.7 Dietary exposure assessment and risk characterization

Exposure assessment refers to the qualitative and/or quantitative evaluation of the likely intake of the hazards (chemical, biological and physical) through food or any other relevant sources (IPCS, 2020). This involves analysis of the daily intake of the biological, chemical, and physical hazards in food by consumers by combining data on consumption and concentration of the hazardous agents (IPCS, 2020). Furthermore, IPCS (2020) defines risk characterization as the processes that involve the determination of the quantitative and/or qualitative estimation of the probability of the occurrence and severity of the potential or known adverse health effects in a given population based on the exposure assessment.

1.1.7.1 Dietary exposure assessment

Exposure assessment involves the estimation of the average intake of chemical contaminants by the combination of consumption data and concentration of the chemical in food. This can be estimated using Equation 1 (Sirot *et al.,* 2012).

$$E = \frac{\mathrm{IR} * \mathrm{C}}{\mathrm{BW}} \quad \text{.........} \quad (1)$$

Where E is the exposure of the subject to the contaminant, IR is the fish consumption rate by the subject, C is the mean concentration level of the contaminant in fish and BW is the body weight of the subject.

1.1.7.2 Risk characterization

Carcinogenic health risks (C_R)

Lifetime cancer risk (C_R) represents the possibility of occurrence of any type of cancer by an individual over a lifetime and is estimated by combining exposure data and the cancer slope as illustrated in Equation 2 (Ibeto *et al.,* 2019; Wenaty *et al.,* 2019a).

$$C_R = \mathrm{CDI} * \mathrm{SF} \quad \text{.........} \quad (2)$$

Where CDI is the chronic average daily intake of PCBs and SF is the cancer slope factor/potency) and CDI is estimated using Equation 3. Where C is the mean concentration level of a chemical contaminant in food, IR is the consumption rate of a particular food.

$$\mathrm{CDI} = \frac{\mathrm{C} * \mathrm{IR}}{\mathrm{BW}} \quad \text{.........} \quad (3)$$

Description of lifetime cancer risks are based on standards as follows; very low when the estimated value is $C_R \leq$ 10E-06, low when 10E-06<value≤10E-04, moderate when 10e-04<value≤10E-03, high when 10E-03<value≤10E-01 and very high when estimated value is above 10E-01 (ATSDR, 2018).

Non-carcinogenic health risks (HI)

Non-carcinogenic risks are risks that are estimated as the probability that a person would develop health risks other than cancer over a long time of exposure and are estimated using the hazard index quotients (HQs) of chemical contaminant, which is the ratio of average dietary exposure to the chemical contaminant and the reference dose which represent a level at which no adverse effects can occur (Lin *et al.,* 2020), as illustrated by using Equation 4.

$$\mathrm{HQ} = \frac{CADI}{RfD} \quad \text{.........} \quad (4)$$

Where RfD is the Reference dose and the overall non-carcinogenic risk is estimated using equation 5 (Lin *et al.,* 2020) for multiple chemical contaminants.

$$\mathrm{HI} = \sum \mathrm{HQs} \quad \text{.........} \quad (5)$$

Where ∑HQs is the sum of the hazard quotients (HQs) of each indicator PCBs measured

Non-carcinogenic risks are using guidelines where; no carcinogenic health risk will occur when HI < 0.1, low non-carcinogenic risks might occur when HI value range from 0.1 to 1, and when HI > 1 there is high potential for non-carcinogenic risks to occur (ATSDR, 2018; Lin *et al.,* 2020).

1.2 Fish Consumption Status in Tanzania

The fishing industry is one of the main sources of food in Tanzania and contributes about 10% of the national export earnings (URT, 2019; WorldFish, 2017). Fisheries are dominated by inland/freshwater fishing which contributes about 85% of all the fish in national-level production (URT, 2019), while fishing from marine water (Indian Ocean) contributes 10% - 15% of the total national fish production. (WorldFish, 2017; Polder *et al.,* 2014). Consumer awareness of the health benefits associated with the consumption of fish and other white meat has increased fish consumption rates, reaching 8.22 kgs per capita fish consumption in 2020 (URT, 2021). Although a majority of fish consumed in Tanzania are from freshwater bodies, the majority of fish consumed in the coastal regions are from marine water (WorldFish, 2017). The majority of the fish consumed is from small-scale fishermen (artisanal fishermen) (FAO, 2014) which make up the largest percentage of the fish caught in the country (WorldFish, 2017; URT, 2019). The fish consumption rate of coastal communities is much higher than that of other communities (Cisneros-Montemayor *et al.,* 2016; Soselisa *et al.,* 2021) with artisanal fishermen and local fish vendors communities consuming a higher amount of fish and fishery products mainly from their catches to minimize living cost as most of them are poor and fish is their main source of food, income, and employment (Cisneros-Montemayor *et al.,* 2016).

1.3 Problem statement and justification

Tanzania imported polychlorinated biphenyls (PCBs) in various products such as transformer oils, capacitors, coolants, fire retardants, adhesive additives, paints, and other products up until they were banned (URT/NIP, 2005, 2018; IPEP, 2005). Despite the import ban, the release of PCBs into the environment has not stopped because of the large amount of PCB-containing equipment that is still in use and/or in storage facilities where accidental release and spills occur due to poor management and storage (IPEP, 2005), while runoff water and some of the volatile and semi-volatile PCBs are transported to other areas far from release/source sites (Arinaitwe *et al.,* 2018; Wenaty, *et al.,* 2019b; Mwakalapa *et al.,* 2018; Wu *et al.,* 2020).

Studies by (Mahugija, 2015; Hellar-Kihampa, *et al.,* 2013; Polder *et al.,* 2014, Wenaty *et al.,* 2019a; Mwakalapa *et al.,* 2018; Muller *et al.,* 2017) have documented the presence of PCBs and other persistent organic pollutants (POPs) contamination in various locations in Tanzania. An inventory report by Vice President's Office in 2005 as an implementation plan for compliance with the Stockholm Convention ratified by Tanzania in 2004, found that there were 418 pieces of equipment containing 273 tons of oil suspected to contain PCBs with more than 500ppm across 25 regions in Tanzania (URT/NIP, 2005).

The follow-up inventory in 2014 in six regions, found that most of the equipment identified to contain PCBs with more than 500ppm in a previous survey was still in place (in use and others in storage facilities), some were leaking and some had been replaced with new PCBs free equipment (URT/NIP, 2018). More than half of the highly contaminated sites identified are located in Dar es Salaam and the neighboring Coastal region and are in an area close to water bodies (rivers) (IPEP, 2005; URT/NIP, 2018) that directly pour water into the Indian ocean likely contaminating fishing grounds and fish which is one of the main source of food in coastal regions (URT, 2019; FAO, 2014). Runoff water and leaching from these sites, ultimately end up in the ocean accumulating in sediments and fish feeding grounds. Furthermore, FAO, (2014) reported that raw sewage from Dar es Salaam city residents, industrial wastes such as paint wastes, tars, oils, alkalis, and organic wastes oils are connected directly to the sewer systems that discharge directly into the sea without pre-treatment.

Marine food (particularly fish) is more highly contaminated with PCBs than other types of food since PCBs are highly lipophilic and therefore bioaccumulate in the fatty tissues of the fish (Ravenscroft and Schell, 2018, WHO, 2016; Jafarabadi *et al.,* 2019). Studies show that human exposure to PCBs from food consumption is a cause of great concern due to their toxicity nature (Hites and Holsen*,* 2019; WHO, 2016; Duedahl-Olesen *et al.* 2020; Ravenscroft and Schell, 2018; Li *et al.,* 2018). Chronic human exposure to PCBs can result to serious health effects such as hormonal (endocrine system) disruption, cancer, fertility, developmental effects, and immunological effects (Wolf *et al.,* 2019; Quijano *et al.,* 2018; Hulin *et al.,* 2020).

Studies (Cisneros-Montemayor *et al.,* 2016; Soselisa *et al.,* 2021) show that the fish consumption rate of coastal communities is estimated to be 15 times that of other communities as communities consume a higher amount of fish and fishery products which makes the communities vulnerable to high exposure to chemical contaminants in case of pollution. Furthermore, for small-scale fishermen and fish vendors, fish is the main dish they consume compared to the general population, therefore studying the status of exposure and possible effects that may be a result of the consumption of contaminated fish is warranted.

The study aimed at providing information on the status of PCBs exposure to fish which is one of the major sources of food in coastal areas (Dar es Salaam), establish the levels of PCBs in selected marine fish species, *Ratrelliger kanagutra* (Indian mackerel) and *Siganus sutor* (spine-foot rabbitfish). The study also assessed the potential health risk for the fishermen and fish vendors who are among high fish consumers and most vulnerable to unacceptable levels of PCBs exposure while also estimating the potential health risks for the general population. The study involved two of the mostly preferred fish species among the study community and widely used in coastal regions due to their availability and affordable price (FAO, 2014; Mziray and Kimirei, 2016; Breuil and Bodiguel, 2015; Sekadende *et al.,* 2020; Ebrahimu, *et al.,* 2020).

1.4 Objectives

1.4.1 Overall objective

The overall objective of this study was the assessment of the occurrence and levels of indicator polychlorinated biphenyls in selected species of marine fish in Dar es Salaam.

1.4.2 Specific objectives

i. To determine the levels of indicator polychlorinated biphenyls in Mackerel fish (*Rastrelliger kanagutra*) and Rabbitfish (*Siganus sotur*).
ii. To assess dietary exposure and health risk of polychlorinated biphenyls to artisanal fishermen and fish vendors.

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CHAPTER TWO

Paper 1: Occurrence and potential health risks of indicator polychlorinated biphenyls (PCBs) in selected marine fish species *Rastrelliger kanagutra* and *Siganus sotur*. A case of Dar es Salaam[*]

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Abstract

The occurrence of polychlorinated biphenyls (PCBs) in marine fish and their potential health risks were assessed in Dar es Salaam region. Six indicatory PCBs were assessed in selected marine fish species, *Rastrelliger kanagutra* and *Siganus sotur* collected from Kivukoni-Feri, the major fish market in Dar es Salaam. The sample was extracted using the QuEChERS method and quantified by GC-MS/MS. The concentration of detected PCBs congeners in the analysed fish species was relatively low, ranging from <LOD to 3.71 ng/g w/w in *R. kanagutra* and from <LOD to 0.58 ng/g w/w in *S. sotur*. However, the sum of mean PCBs levels was below the maximum residue limit (MRL) of 75 ngg^{-1}w/w set by the European Commission (EC) in the muscles of fish and fishery products. The estimated lifetime cancer risks were categorised as very low in both children and adults, with indices values ranging from 1.24E-08 to 6.14E-07 for children and 5.14E-09 to 2.55E-07 for adults, while non-carcinogenic risks were insignificant as hazard indices were less than 0.1 for both children and adults.

Keywords: Polychlorinated biphenyls (PCBs), marine fish, persistent organic pollutants (POPs), risk assessment, biomagnification

2.1 Introduction

Polychlorinated biphenyls (PCBs) are synthetic industrial chemicals widely used worldwide from the 1920s until the ban on their commercial production was imposed in the late 1970s (Montano *et al.,* 2022; Shi *et al.,* 2019; Munawar *et al.,* 2021). PCBs are characterized by their resistance to degradation (Romanic *et al.,* 2018; Wenaty *et al.,* 2019a), high lipophilicity (Mwakalapa *et al.,* 2018; Lin, *et al.,* 2020; Munawar *et al.,* 2021), and ability to transport over a long distance far from the point of production or release (Umasangaji *et al.,* 2020) remaining in the environment for a long time and accumulate along the food chains (Lin, *et al.,* 2020). PCBs had many uses in applications such as flame retardants, heat-resistant lubricants, paints, electrical transformers, and electronic appliances (Shen *et al.,* 2017; URT/NIP, 2018; Wenaty *et al.,* 2019a).

Improper handling of the PCBs containing products during use, storage, and maintenance (Umasangaji *et al.,* 2020) leads to the release of PCBs into the environment and then distributed to other locations far from the site of release through runoff water, underground water from contaminated areas and emissions from open-burning municipal waste landfills (Jafarabadi *et al.,* 2019; Hellar-Kihampa *et al.,* 2013; Arinaitwe *et al.,* 2018; Aslam *et al.,* 2019). Most PCBs end up in oceans sediments which is the stable reservoir for PCBs, remaining there for a long time and continuously being released in small quantities into the water and other fish feeding grounds over time (Ibeto *et al.,* 2019; Obanya *et al.,* 2019; Umasangaji *et al.,* 2020). Human exposure to PCBs is primarily through the consumption of food of animal origin namely fish, meat, and dairy products (WHO, 2016; Duedahl-Olesen *et al.,* 2020; Ravenscroft and Schell, 2018). Exposure to PCBs can result in adverse health effects including endocrine system disruption, cancer, infertility, developmental effects in breastfed infants, and immunological effects (Hulin *et al.,* 2020; Lin *et al.,* 2020; Muller *et al.,* 2017; Quijano *et al.* 2018; WHO, 2016; Panseri *et al.,* 2019; Montano *et al.,* 2022).

Tanzania imported and used products containing PCBs including electrical transformers, oil circuit breakers, and switch gears before they were banned, with some of them still in use and others in storage across the country (URT/NIP, 2005, 2018). Studies (Hellar-Kihampa *et al.,* 2013; Mahugija, 2015; Polder, 2014; Wenaty *et al.,* 2019a; Mwakalapa *et al.,* 2018; Muller *et al.,* 2017) have documented the presence of PCBs contamination in various locations in Tanzania. As most of the PCBs from contaminated sited end up in the ocean, marine organisms in particular fish are vulnerable to contamination by PCBs (Duedahl-Olesen *et al.,* 2020; Montano *et el.,* 2022).

Fish is one of the main sources of food in Tanzania, and is an important source of essential compounds such as iodine, vitamin D, and essential fatty acids, making up to 19.7% of the country's animal protein intake (URT 2019; WorldFish, 2017; Duedahl-Olesen *et al.,* 2020). However, there is limited information on PCBs contamination status in the marine environment and seafood in Tanzania. This study aimed to assess the occurrence of indicator PCBs (PCB 28, PCB 52, PCB 101, PCB 138, PCB 153, and PCB 180) in selected marine fish species *Rastrelliger kanagutra* (Indian mackerel) and *Siganus sutor* (spine-foot rabbitfish) and assess their probable health effects in Dar es Salaam city, Tanzania.

2.2 Materials and Methods

2.2.1 Study area description

This study was conducted in Dar es Salaam city located along the coast of mainland Tanzania. Dar es Salaam is the most populous and industrialized region in Tanzania with

5.4 million people according to the 2022 census. The city is the major hotspot for the discharge of industrial and urban wastes in the Indian Ocean.

2.2.2 Choice of analysed fish species

Fish samples used in this study were collected from two species, *Rastrelliger kanagutra* (Indian mackerel) and *Siganus sutor* (spine-foot rabbitfish) chosen because of their popularity in the Dar es Salaam region. Studies (Mziray and Kimirei, 2016; Breuil and Bodiguel, 2015; Sekadende *et al.,* 2020; Ebrahimu, *et al.,* 2020) have reported the significant contribution of the two species (>40%) to the total daily catches in most of the Western Indian Ocean (WIO) regions throughout the season, therefore, taking a significant part in the daily meal in coastal communities due to their lower prices. Furthermore, both of the species habitats, are in the shallow coastal waters that are considered to be most likely contaminated by the chemical toxicants in waste effluents and contaminated river water discharged into the ocean (Umasangaji *et al.,* 2020).

2.2.3 Sample collection and preparation

Fish samples were collected from fishermen and fish vendors at the Kivukon Feri fish market in Dar es Salaam. Sixty fish samples from two different species (30 fish samples from each species) *R. kanagutra* and *S.sutor* were randomly collected. The collected samples were placed in the container, marked appropriately, immediately transferred into a cool ice box, and transported to the Government Chemist Laboratory Authority (GCLA) laboratory where they were held at -18℃ until the time of analysis. During sample preparation, fish were thawed and skin-free fillets were cut and placed on clean aluminum foil, then homogenized using a laboratory blender. The homogenized sample was placed in sample containers and stored at -18℃ until the time of analysis. Contamination during sample preparation was avoided by cleaning the laboratory blender, knives, spatula, and forceps thoroughly with soap and water then wiping with ethanol between samples.

2.2.4 Sample extraction and clean-up

Fish samples extraction was conducted by using a slightly modified Quick, Easy, Cheap, Efficient, Rugged, and Safe (QuEChERS) extraction method described in detail by Norli *et al.* (2011), where five grams of fish fillets were weighed into 50 ml centrifuge tubes marked M for *R. kanagutra* and R for *S. sotur* samples. Five milliliters of water were added and vortexed for one minute to homogenize the mixture. Ten milliliters of acetonitrile were then added to the mixture and vortexed for two minutes then sonicated for 30 minutes to enhance extraction. The mixture was then centrifuged for five minutes at 4000rpm and placed into a freezer and kept at – 20℃ for one hour, then immediately centrifuged for five minutes at 4000rpm. The supernatant was then transferred into a tube containing 300 mg $MgSO_4$ and 50 mg PSA and the mixture was vortexed and centrifuged for five minutes, then the supernatant was transferred into the vials for GC analysis.

2.2.5 Instrumental analysis

Detection and quantification of indicator PCBs were carried out using a gas chromatography-tandem mass spectrometer (GC-MS/MS – Agilent 7890B – 7000D) operating in multiple reaction monitoring (MRM/SRM) with HP-5MS capillary column (30 m x 0.25 mm x 025µm). The carrier gas used was helium flowing at a constant flow rate of 1.2 ml/min. The injection volume was 1 µL in splitless mode into the GC column. The inlet temperature was set at 280℃ and the program for oven temperature was, 80℃ hold for 1 min, 80 – 170℃ at 50℃ min^{-1}, then 170 – 240℃at 5℃ min^{-1}, hold for 1 minute, finally 240 – 280℃ at 20℃ min^{-1}.

2.2.6 Quality control and assurance

Cross-contamination and contamination from outside environments were controlled by conducting analyses in a controlled environment. In-house quality control samples (spiked sample, blank sample, and blank solvent) which were processed after a run of each 10 samples were used for monitoring and recovery analyses. Recoveries were estimated from a spiked sample of the six indicator PCBs standards (PCB 28, 52, 101, 138, 153, 180) and analysed with the same analytical method used to analyse the samples. The concentration (ngg^{-1}) obtained in samples was corrected by using the blank values obtained for each batch.

2.2.7 Risk characterisation

The potential human health risks related to human exposure to PCBs through the consumption of contaminated fish were estimated by using a risk assessment model adopted from (Wenaty *et al.,* 2019a) in which lifetime cancer risk (C_R) was calculated using Equation 1;

$$C_R = SF \times CDI \quad \ldots\ldots(1)$$

where SF is the cancer oral slope factor (2 per mg/kg/day) used for food ingestion and early life exposures (ATSDR, 2018), CDI is the estimated chronic daily intake, and the CDI is estimated using equation 2;

$$\mathrm{CDI} = \frac{\mathrm{C \times IR \times EF \times ED}}{\mathrm{BW \times AT}} \quad \ldots\ldots(2)$$

Where C is the measured concentration of contaminant in fish (mg/kg), IR is the fish consumption rate 23 gday^{-1} (URT 2021), EF is the exposure frequency (365 day/year), ED is the exposure duration (60 years for adults and 12 years for children (Wenaty *et al.,* 2019a), BW is the hypothetical average body weight ((70kgs for adults and 29kgs for children for Africans and Europeans consumers (Yu *et al.,* 2010) and AT is the average time per year (For carcinogenic risk for both adults and children, AT was set at 70 hrs × 365 days and for non-carcinogenic risks AT was set at 30hrs × 365 days for adults and 8 × 365 days for children, (Lin *et al.,* 2020).

Non-carcinogenic risks were estimated using the hazard quotients (HQs) for each indicator PCBs by using Equation 3;

$$\mathrm{HQ} = \frac{\mathrm{CDI}}{RfD} \quad \ldots\ldots(3)$$

Where RfD is the Reference dose (estimate of daily oral exposure, 0.02 mg/kg-day (Lin *et al.,* 2020) and the overall non-carcinogenic risk was were estimated using Equation 4;

$$\mathrm{HI} = \sum \mathrm{HQs} \quad \ldots\ldots(4)$$

Where ∑HQs is the sum of the hazard quotients (HQs) estimated for each of the indicator PCBs.

Description of lifetime cancer risks were based on (ATSDR, 2018) standards as follows; very low when the estimated value is $Cr \leq 10E\text{-}06$, low when $10E\text{-}06<\text{value}\leq 10E\text{-}04$, moderate when $10e\text{-}04<\text{value}\leq 10E\text{-}03$, high when $10E\text{-}03<\text{value}\leq 10E\text{-}01$ and very high when estimated value is $Cr >10E\text{-}01$. For non-carcinogenic risks, no carcinogenic health risk will occur when HI < 0.1, low potential for non-carcinogenic risks to occur when HI value range from 0.1 to 1, and when HI > 1 there is a high potential for non-carcinogenic risks to occur (Wenaty *et al.,* 2019a; Lin *et al.,* 2020).

2.2.8 Data analysis

Statistical analysis of data was conducted using IBM SPSS software v.20 in which data were presented in mean. One sample t-test was used to compare the sum of the estimated PCBs ($\sum_6$PCBs) levels to the set maximum residue level (MRL) and the independent t-test was used to compare concentration between species. A level of $p < 0.05$ was chosen as the criterion for statistical significance.

2.3 Results

2.3.1 Characteristics of analysed fish samples

The mean length for *R. kanagutra* was 25.79 cm with lengths ranging from a minimum length of 23.5 cm to a maximum length of 27.5 cm while the mean weight was 300.54 grams with weights ranging from a minimum weight of 214.74 grams to a maximum weight of 389.79 grams. The mean length for *S. sutor* was 28.07 cm with lengths ranging from a minimum length of 24.0 cm to a maximum length of 33.0 cm, while the mean weight was 448.22 grams with weights ranging from a minimum weight of 280.41grams to a maximum weight of 717.77 grams.

2.3.2 Occurrence and levels of PCBs in fish muscles

Sixty fish samples were analysed for PCBs, 30 samples from each species, *R. kanagutra* and *S. sutor*. The results showed the presence of indicator PCBs in both fish species with 73.3% of *R. kanagutra* samples and 20% of *S. sutor* samples contaminated with PCBs (Table 2.1).

Table 2.1: Mean concentration of PCBs contaminants in ngg-1 wet weight (ngg^{-1} w/w) in fish species *R. kanagutra* and *S. sutor* from Kivukoni-Feri fish market.

		Mean Concentration (ngg^{-1} w/w)				
PCB Congener	N	*R. kanagutra*	Sample detected (%)	*S. sutor*	Sample detected (%)	EU-MRL
PCB 28	30	<LOD	0	<LOD	0	-
PCB 52	30	<LOD	0	<LOD	0	-
PCB 101	30	0.008	7	<LOD	0	-
PCB 138	30	0.220	56	0.008	7	-
PCB 153	30	0.396	73	0.026	13	-
PCB 180	30	0.216	63	0.016	13	-
$\sum_6$PCBs	**30**	**0.840**		**0.050**		75

Key: N: Total number of samples analyzed for each species, EU-MRL: Maximum residue limit set by European Commission (EC, 2011), w/w: Wet weight, LOD: Limit of Detection

The levels of the six indicator PCBs were relatively low in both fish species with the highest mean concentration ($\sum_6$PCBs) of 3.71 ngg^{-1} w/w observed in *R. kanagutra.* For *R. kanagutra* analysed samples, the individual levels of congeners, PCBs 28 and 52 were below detection level (<LOD), PCB 101 concentration ranged from 0.11 to 0.12 ngg^{-1} w/w, PCB 138 ranged from 0.12 to 1.07 ngg^{-1} w/w, PCB 153 ranged from 0.1 to 1.55 ngg^{-1} w/w and PCB 180 concentration ranged from 0.11 to 1.09 ngg^{-1} w/w, and the sum of the six indicator PCBs ($\sum_6$PCBs) levels ranged from 0.12 to 3.71 ngg^{-1} w/w (Appendix 1.2). For *S. sutor* analysed

samples, the individual levels of congeners, PCBs 28, 52 and 101 were below detection level (<LOD), PCB 138 concentration ranged from 0.1 to 0.14 ngg^{-1} w/w, PCB 153 ranged from 0.13 to 0.28 ngg^{-1} w/w and PCB 180 ranged from 0.12 to 0.13 ngg^{-1} w/w and the sum of six indicator PCBs ($\sum_6$PCBs) levels ranged from 0.12 to 0.54 ngg^{-1} w/w (Appendix 1.2).

2.3.3 Risk characterization

The potential human health risks to the general population as a result of PCBs contaminated fish consumption were assessed using equations 1, 2, 3, and 4 for both children and adults. The estimated lifetime cancer risks, C_R, were between 1.24E-08 and 6.14E-07 for children and between 5.14E-09 and 2.55E-07 for adults, while the estimate non-carcinogenic health risks (HI), were between 1.95E-06 and 3.26E-05 for children and 8.07E-07 and 1.35E-05 for adults (Table 2.2).

Table 2.2. Estimated lifetime cancer risks for children and adults as a result of intake of detected PCBs in *R. kanagutra* and *S. sutor* from Kivukoni-Feri fish market.

	Estimated lifetime cancer risks (C_R)			
	R. kanagutra		*S. sutor*	
PCB Congener	Adults	Children	Adults	Children
PCB 28	-	-	-	-
PCB 52	-	-	-	-
PCB 101	5.14E-09	1.24E-08	-	-
PCB 138	1.41E-07	3.41E-07	5.14E-09	1.24E-08
PCB 153	2.55E-07	6.14E-07	1.68E-08	4.07E-08
PCB 180	1.39E-07	3.35E-07	1.03E-08	2.48E-08

Table 2.3. Non-carcinogenic risks for children and adults as a result of intake of detected PCBs in *R. kanagutra* and *S. sutor* from Kivukoni-Feri fish market

	Estimated Non-carcinogenic risks (HI)			
	R. kanagutra		*S. sutor*	
PCB Congener	Adults	Children	Adults	Children
PCB 28	-	-	-	-
PCB 52	-	-	-	-
PCB 101	1.29E-07	3.10E-07	-	-
PCB 138	3.54E-06	8.53E-06	1.29E-07	3.10E-07
PCB 153	6.36E-06	1.54E-05	4.21E-07	1.02E-06
PCB 180	3.47E-06	8.38E-06	2.57E-07	6.21E-07
Hazard Indices (∑HQs)	**1.35E-05**	**3.26E-05**	**8.07E-07**	**1.95E-06**

Key: ∑HQs: Sum of hazard quotients, HI: Hazard indices

2.4 Discussion

2.4.1 Occurrence and levels of PCBs in fish muscles

The estimated PCBs levels in the analysed fish species were dominated by the three congeners PCB 138, 153, and 180. PCB 153 was the most prevalent congener in the present study for both fish species followed by PCB 138 and PCB 180 congeners. The prevalence of the three congeners is explained by their higher degree of chlorination which makes them more resistant to degradation in the environment, ability to bioaccumulate due to their higher lipophilicity and low biodegradation rate in organisms (Lin *et al.,* 2020; Wenaty *et al.,* 2019a; Mwakalapa *et al.,* 2018). This trend is similar to that observed by (Mwakalapa *et al.,* 2018, Haar *et al.,* 2021; Shang *et al.,* 2016) where PCB 153 congener was more dominant but slightly different from studies from other locations in Tanzania (Wenaty *et al.,* 2019a; Polder *et al.,* 2014) where PCB 138 congener was more dominant congener, which suggests contamination originated from different sources of PCBs (Mwakalapa *et al.,* 2018). The absence of PCB 28 and 52 in both species and the very small contamination level of PCB 101 only in *R. kanagutra* species suggest that contamination is not from the fresh release of PCBs into the environment. This can be explained since PCB 28, 58, and 101 have relatively higher volatility, water solubility, and biodegradability than PCB 138, 153, and 180 (Lin *et al.,* 2020; Wenaty *et al.,* 2019a; Hellar-Kihampa *et al.,* 2013).

Statistical analysis showed a significant difference in the mean PCBs levels between the analysed species samples at ($p< 0.05$). The mean PCBs concentration of indicator PCBs congener in *R. kanagutra* was significantly higher than *S. sutor* with a sum of indicator PCBs mean concentration ($\sum PCB_6$) ranging from 0.12 to 3.71 ngg^{-1} w/w for *R. kanagutra* and 0.12 to 0.54 ngg^{-1} w/w for *S. sutor*. However, the PCBs contamination levels observed were below the maximum residue limits (MRL) of 75 ngg^{-1}w/w set by the European Commission, for fish meat and fishery products (EC, 2011). The higher concentration of PCBs in *R. kanagutra* fish species than in *S. sutor* species was probably due to the difference between their feeding habit and that *R. kanagutra* have higher trophic levels than *S. sutor* (Fishbase, 2017). *R. kanagutra* are omnivorous feeding on seagrasses and phytoplankton (diatom/algae) but also feed on zooplanktons and other small organisms such as polychaetes, ostracods, cladocerans, and small fishes such as anchovy and sardines which results to higher biomagnifacation (Mziray and Kimirei, 2016; Fishbase, 2017) while the *S. sutor* are herbivorous fish feeding on seaweed and seagrasses benthic algae and associated flora (Mziray and Kimirei, 2016; Fishbase, 2017). These findings have a similar trend to results obtained by other studies (Shen *et al.,* 2017; Panseri *et al.,* 2019) that showed omnivorous fish and other predator fish (carnivorous) have more contamination levels of PCBs than herbivorous fish because of biomagnification.

Fish habitation may have also influenced the difference in PCBs contamination levels in *R. kanagutra* and *S. sutor* fish species. *R. kanagutra* are oceanodromous fish and habit may have contributed to its higher PCBs levels compared to *S. sutor* which lives in shallow water but are not oceanodromous. This finding is in line with those obtained in a study by Shang *et al.* (2016) where oceanodromous fish living in shallow water were found to have higher PCBs contamination than non-oceanodromous species that live in shallow water. Fish age is also one of the factors influencing the concentration level of PCBs in fish (Wenaty *et al.,* 2019a; Polder *et al.,* 2014). The age of the collected fish sample in the present study was not recorded but the maximum age of *R. kanagutra* is reported to be 4 years while the reported maximum age of *S. sutor* is 2.5 years (Fishbase, 2017) which may explain why *R. kanagutra* have higher PCBs levels than *S. sutor* due to difference in time of accumulation of PCBs in corresponding fish species taking into account the average size of the analysed samples (Fishbase, 2017).

The findings in the present study showed that the concentration of PCBs levels obtained in marine fish in this study was relatively higher than that of fish from freshwater in Tanzania, where Polder *et al.* (2014) reported that tilapia from Lake Tanganyika (0.57 ngg^{-1} w/w), Lake Nyasa (0.02 ngg^{-1} w/w) and Lake Victoria (0.08 ngg^{-1} w/w) had lower PCBs levels than that obtained in the present study (0.84 ngg^{-1} w/w) with the same feeding habits (*R. kanagutra*). This trend is similar to other reported studies where PCBs contamination levels in marine fish were found to be higher than that obtained from freshwater fish (Ahmed *et al.,* 2016; Shang *et al.,* 2016) probably due to the vast amount of waste discharged into the ocean from multiple source channels and rivers from different parts of the country far from the ocean. However, the PCBs levels obtained in this study were lower than that obtained by other studies for marine fish with the same feeding habits in India (65.8 ngg^{-1} w/w) by Ahmed *et al.* (2016) and China (3.82 ngg^{-1} w/w) by Shang *et al.* (2016) involving *R. kanagutra* fish species which suggest higher level of contamination because to higher level of industrialization of the areas which may lead to higher environment pollution.

2.4.2 Risks characterization

The estimated lifetime cancer risks estimated for adults and children were observed to be below the reference critical values of 10E-6 for adults and 10E-04 for children (Lin *et al.,* 2020), ranging between from 5.14E-09 to 6.14E-07 for both children and adults which indicate that consumption of the *R.kanagutra* and *S. sutor* fish species from the Kivukoni Feri fish market will have negligible adverse health effects. The hazard indices (HI) values used to estimate the non-carcinogenic health risks were lower than 0.1, ranging from 1.95E-06 to3.26E-05 for both children and adults, also indicating that there are no non-carcinogenic risks that are likely to occur as a result of consumption of the *R.kanagutra* and *S. sutor* fish species from the study area.

2.5 Conclusion

Based on the findings of this study, there was PCBs contamination in the study area. However, the detected PCBs concentration levels were below the maximum residue limits (MRL) set by European Commission, while the estimated values for potential health effects using the detected contamination levels were observed to have insignificant adverse health effects for both children and adults in the general population. Despite the very low-risk estimates of potential health risks in the present study, further studies and regular monitoring of PCBs contamination of marine fish and fisheries products are recommended.

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CHAPTER THREE

Paper 2: Dietary exposure and risk assessment of polychlorinated biphenyls to artisanal fishermen and fish vendors. A case study of Dar es Salaam[†]

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Abstract

Dietary exposure and potential health risks to artisanal fishermen and fish vendors were assessed in Dar es Salaam city, Tanzania. Fish consumption and preference were estimated using a consumption survey among fishermen and fish vendors. Polychlorinated biphenyls (PCBs) levels were estimated from the most preferred fish species. The estimated overall mean PCBs levels were 0.45 ngg^{-1} w/w and the mean fish consumption rate among fishermen was 400.5 $gday^{-1}$. The mean intake of total PCBs by fishermen and fish vendors was higher in comparison to that of the general population ranging between 5.10 and 27.56 $ngkg^{-1}$ $bw.day^{-1}$ for normal and higher fish consumers, depending on the estimation scenario, while for general population exposure to total PCBs were 0.14 and 0.31 $ngkg^{-1}$ $bw.day^{-1}$ in lower and upper estimation scenario respectively. The average daily intake of total PCBs by fishermen and fish vendors exceeded the recommended tolerable daily intake, (TDI) of total PCBs of 20 $ngkg^{-1}$ $bw.day^{-1}$ in high fish consumers while the estimated lifetime carcinogenic and non-carcinogenic hazard indices for fishermen and fish vendors exceeded the recommended health guidelines raising serious concerns for potential adverse health effects. For the general population, the potential for adverse health effects was insignificant as the estimated hazard indices meet the recommended average lifetime intake of PCBs. However, regular monitoring of fish contamination with PCBs in the study area is recommended for the timely mitigation of risks.

Keywords: Dietary exposure, risk assessment, polychlorinated biphenyl (PCBs), artisanal fishermen, fish vendors.

3.1 Introduction

Polychlorinated biphenyls (PCBs) are one of the persistent organic pollutants, industrial chemical pollutants that were extensively used before their commercial production was banned in the late 1970s (Kampire *et al.,* 2017; Montano *et al.,* 2022). PCBs contamination is still widespread in the environment due to their resistance to degradation (Ravenscroft and Schell, 2018; Wenaty *et al.,* 2019a, Dos Santos *et al.,* 2018), resulting in the bioconcentration and biomagnification in the food chain (Quijano *et al.,* 2018; Shi *et al.,* 2019; Munawar *et al.,* 2021). The presence of PCBs in the food chain is a cause of great concern due to their toxicity nature (Hites and Holsen, 2019; Quijano *et al.,* 2018; WHO, 2016).

Chronic human exposure to PCBs can result in serious health effects such as endocrine system (hormonal) disruption, cancer, fertility, developmental effects, and immunological effects (Montano *et al.,* 2022; Quijano *et al.,* 2018; WHO, 2016; Muller *et al.* (2017)). Studies by Raffetti *et al.* (2018) and Wolf *et al.* (2019) have reported the relationship between diabetes and other hormonal imbalances due to PCBs exposure in humans. Most of the exposure to PCBs is through the consumption of food of animal origin, meat, fish, and dairy products (WHO, 2016; Duedahl-Olesen *et al.,* 2020; Sirot *et al.,* 2012). Previous dietary studies conducted in Africa (Vaccher *et al.,* 2020), similar to other studies in Europe and Asia (Quijano *et al.,* 2018; Sirot *et al.,* 2012; Zang e*t al.,* 2015) showed that fish and other marine products had a higher level of contamination with PCBs compared to other animal source food with higher fat content.

Fish is one of the main sources of food and income in coastal area communities in Tanzania, and is also an important source of iodine, vitamin D, essential fatty acids, and protein (Polder *et al.,* 2014; WorldFish, 2017; Duedahl-Olesen *et al.,* 2020). Consumer awareness of the health benefits associated with the consumption of fish and other white meat has increased fish consumption rates, reaching 8.22 kg per capita fish consumption in 2020 (URT, 2021). However, fish consumption in coastal communities is estimated to be 15 times higher than the general population (Cisneros-Montemayor *et al.,* 2016; Soselisa *et al.,* 2021). Artisanal fishermen and local fish vendors communities in particular, consume a higher amount of fish and fishery products mainly from their daily catches to minimize living costs (Cisneros-Montemayor *et al.,* 2016). Consumption frequency and amount of contaminated food contribute to the risk of exposure to toxic chemicals (ATSDR, 2018). Artisanal fishermen and local fish vendors communities are at much higher risk in case of chemical contamination than the general population that eats a lower amount of fish and at a lower frequency (USEPA, 2011).

This study aimed to assess the dietary exposure of the artisanal fishermen and small-scale fish vendors and estimate the potential health risks associated with the consumption of fish contaminated with PCBs in Dar es Salaam city, in comparison to the general population by analysis of indicator PCBs (PCB 28, PCB 52, PCB 101, PCB 138, PCB 153 and PCB 180) in the preferred fish species among fishermen and fish vendors.

3.2 Materials and Methods

3.2.1 Study area description

The study was carried out in Dar es Salaam in mainland Tanzania. As the most industrialized and populous city in Tanzania with 5.4 million people (2022 census), the city is the major hotspot for the discharge of industrial and urban wastes in the Indian Ocean.

3.2.2 Fish consumption survey

A food frequency questionnaire-based (FFQ) survey was conducted to assess the fish consumption habits among artisanal fishermen and fish vendors community at the Kivukoni Feri fish market in Dar es Salaam. The survey involved 200 randomly selected volunteers constituting 100 fishermen and 100 fish vendors who use the market as their landing site. The participants were asked to recall the frequency at which they consume fish over a given period (per day, in a week, month, or never) while estimating the serving size per meal. Participants were also asked to identify the preferred fish species and factors affecting their preference for consumed species.

3.2.3 Sample collection and preparation

Fish samples were randomly collected from fishermen and fish vendors at the Kivukoni-Feri fish market in Dar es Salaam. Sixty fish samples, 30 fish samples for each of the two species, *Rastrelliger kanagutra* (Indian mackerel) and *Siganus sutor* (spine-foot rabbitfish), the most preferred fish species among fishermen and fish vendors (according to the initial food frequency survey conducted in this study), were randomly collected. The collected samples were placed in a container, marked appropriately, and immediately transferred into a cool ice box and transported to the laboratory where they were held at -18℃ until analysis.

3.2.4 Sample extraction and clean-up

Sample extraction was conducted by using a slightly modified Quick, Easy, Cheap, Efficient, Rugged, and Safe (QuEChERS) extraction method described in detail by Norli *et al.* (2011), where five grams of fish filets were weighed into 50 ml polypropylene centrifuge tubes marked M for *R. kanagutra* and R for *S. sotur* samples. Five milliliters of water were added and vortexed for one minute to homogenize the mixture. Ten milliliters of acetonitrile were then added to the mixture and vortexed for two minutes then sonicated for 30 minutes to enhance extraction. The mixture was then centrifuged for five minutes at 4000rpm and placed into a freezer and kept at – 20℃ for one hour, then immediately centrifuged for five minutes at 4000rpm. The supernatant was then transferred into a tube containing 300 mg $MgSO_4$ and 50 mg PSA and the mixture was vortexed and centrifuged for five minutes, then the supernatant was transferred into the vials for GC analysis.

3.2.5 Instrumental analysis

The indicator PCBs detection and quantification were determined by using a gas chromatography-tandem mass spectrometer (GC-MS/MS – Agilent 7890B – 7000D) operating with HP-5MS capillary column (30 m x 0.25 mm x 025 µm). Quantification was performed in multiple reaction monitoring (MRM) mode, where MRM transitions (256 > 186 for PCB 28, 289.9 > 220 for PCB 52, 323.9 > 253.9 for PCB 101, 357.8 > 287.9 for PCB 138, 359.8 > 287.9 for PCB 153 and 393.8 > 323.9 for PCB 180) were monitored. The inlet temperature was set at 280^0C and the program for oven temperature was, 80 ^{0}C hold for 1 min, 80 – 170^0C at 50^0C min^{-1}, then 170 – 240 ^{0}C at 5 ^{0}C min^{-1}, hold for 1 minute, finally 240 – 280^0C at 20^0C min^{-1}. Helium was used as carrier gas flowing at a constant flow rate of 1.2 ml/min. The injection volume was 1 µL in splitless mode into the GC column.

3.2.6 Quality control and assurance

Quality control measures were taken to avoid cross-contamination and contamination from outside environments. In-house quality control samples (spiked sample, blank sample, and blank solvent) which were processed after a run of each 10 samples were used for monitoring and recovery analyses. The recovery efficiency of the method was estimated from a spiked sample of the six indicator PCBs standards and analysed with the same analytical method used to analyse the samples. The concentration (ngg^{-1}) obtained in samples was corrected by using the blank values obtained for each batch.

3.2.7 Dietary exposure assessment

Exposure assessment to estimate the fishermen and fish vendors' daily intake of PCBs, was carried out by a single-point deterministic dietary exposure estimation approach where food consumption data is combined with a single value of chemical concentration found in food (IPCS, 2020). The Equation applied was adopted from Sirrot *et al.* (2012);

$$E = \frac{\mathrm{IR} * \mathrm{C}}{\mathrm{BW}} \quad (1)$$

Where E is the exposure of the subject to the contaminant, IR is the fish consumption rate by the subject (400.5 $gday^{-1}$, from this study's consumption survey), C is the mean concentration of a contaminant in fish and BW is the body weight of the subject (70kg for adults, for Africans and European consumers (Yu *et al.,* 2010)). Total PCBs exposure was assumed to be twice the exposure to the six indicator PCBs since the six indicator PCBs represent approximately 50% of all PCB contamination in environments and food (Sirot *et al.,* 2012). The obtained chemical contaminant data that were below the limit of quantitation (LOQ) were left-censored according to the EFSA recommendations, and therefore estimations were conducted in two scenarios (lower bound and upper bound) for which in the lower bound scenario (LB), data below LOQ were treated as zero and in the upper bound scenario (UB), data below LOQ were treated as equal to LOQ (EFSA, 2010). Exposure assessment was carried out by considering only the chemical contamination data from fish species analysed in this study and where mean consumption amount represented normal/regular fish consumers and 95th percentile consumption amount represented high fish consumers.

3.2.8 Risk characterization

3.2.8.1 Carcinogenic health risks (CR)

The lifetime cancer risk (C_R) which represents the possibility of occurrence of any type of cancer was estimated using Equation (2) adopted from Ibeto *et al.* (2019)

$$C_R = \mathrm{CADI} * \mathrm{SF} \quad (2)$$

Where CADI is the chronic average daily intake of PCBs and SF is the cancer slope factor/potency, 2 per mg/kg/day (ATSDR, 2018) and CADI were estimated using Equation 3 adopted from Ibeto *et al.* (2019)

$$\mathrm{CADI} = \frac{\mathrm{C} * \mathrm{IR}}{\mathrm{BW}} \quad (3)$$

Where C is the mean concentration of contaminant in fish, IR is the consumption rate of fish by fishermen and fish vendors and the Kivukoni-Feri fish market.

Description of lifetime cancer risks was based on (ATSDR, 2018) standards as follows; very low when the estimated value is Cr ≤ 10E-06, low when 10E-06<value≤10E-04, moderate when 10E-04<value≤10E-03, high when 10E-03<value≤10E-01 and very high when estimated value is above 10E-01.

3.2.8.2 Non-carcinogenic health risks (HI)

Non-carcinogenic risks were estimated using the hazard index quotients (HQs) of each indicator PCBs measured using Equation 4 (Lin *et al.,* 2020).

$$\mathrm{HQ} = \frac{\mathrm{CADI}}{\mathrm{RfD}} \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots \quad (4)$$

Where RfD is the Reference dose (estimate of daily oral exposure, 0.02 mg/kg/day (Lin *et al.,* 2020), and the overall non-carcinogenic risk was estimated using Equation 5 adopted form Lin *et al.* (2020).

$$\mathrm{HI} = \sum \mathrm{HQs} \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots \quad (5)$$

Where ∑HQs is the sum of the hazard quotients of each indicator PCBs measured
Non-carcinogenic risks were evaluated according to recommended health guidelines where, no carcinogenic health risk will occur when HI < 0.1, low non-carcinogenic risks might occur when HI value range from 0.1 to 1, and when HI > 1 there is high potential for non-carcinogenic risks to occur (Lin *et al.,* 2020; ATSDR, 2018).

3.2.9 Data analysis

Statistical analysis was conducted using IBM SPSS software v.20 in which data were presented in mean. One sample t-test was used to compare obtained ∑PCBs levels to the set maximum residue level (MRL) and the independent t-test was used to compare concentration between species. A level of $p < 0.05$ was chosen as the criterion for statistical significance.

3.3 Results and Discussion

3.3.1 Consumption characteristics of the respondent

The food frequency survey showed that 35% of fishermen and fish vendors consumed *R. kanagutra more* than other types of fish, while 16% preferred *S. sutor* over other types of fish for consumption (Fig. 3.1). Furthermore, the survey showed that 54% of the respondent population consume fish every day (Fig. 3.2). The preference of *R. kanagutra* and *S. sutor* for consumption by fishermen and fish vendors may be due to their availability in the daily catches and lower prices (Breuil and Bodiguel, 2015), making them more affordable to be used as meal compared to other species that are sold at higher prices.

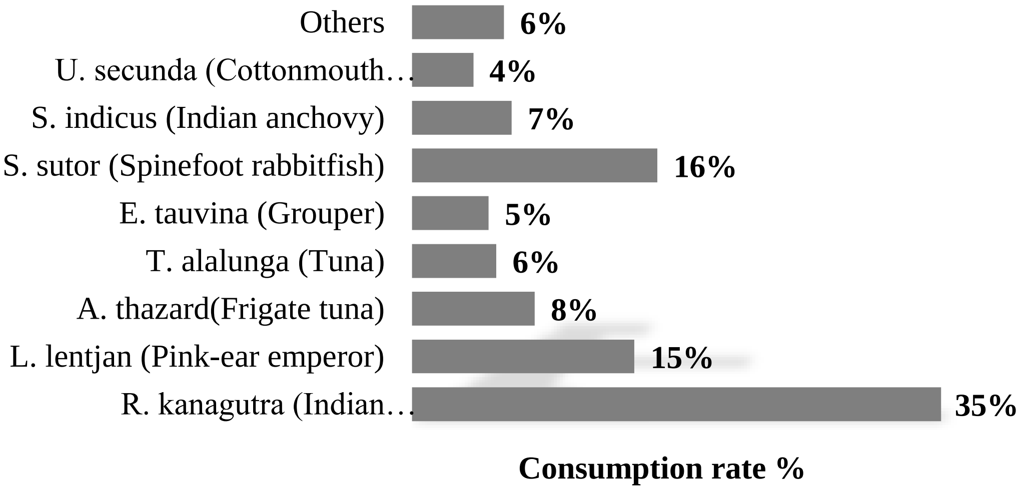

Figure 3.1: Fish consumption preference/rate among fishermen and fish vendors

R. kanagutra is one of the most caught fish species and is economically important in countries along the East coast of the Indian Ocean including Tanzania (Mziray and Kimirei, 2016; Sekadende *et al.,* 2020). In addition, the consumption of fish on most days within a week by respondents may be due to awareness of the health benefits associated with the consumption of fish and other white meat (Wenaty *et al.,* 2018).

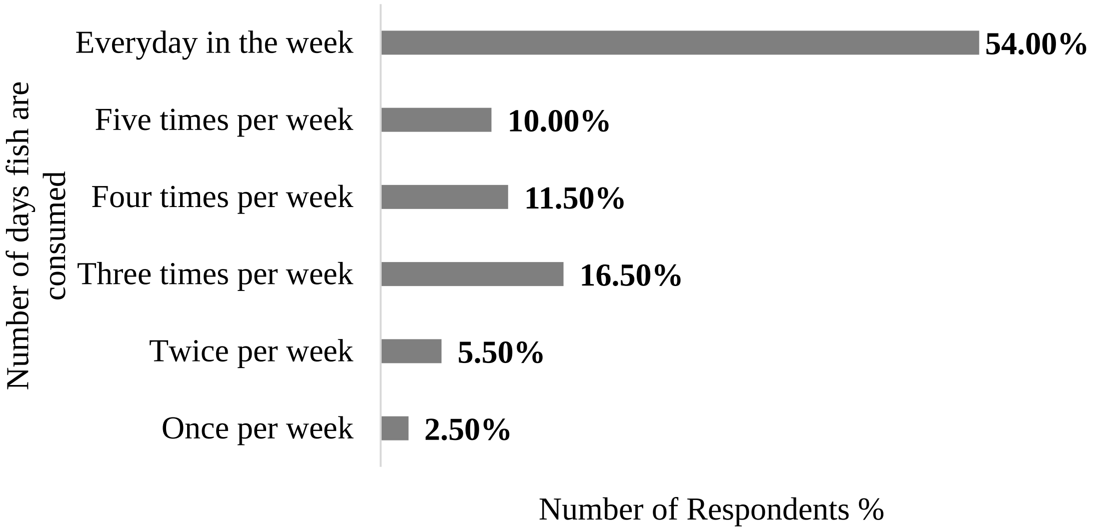

Figure 3.2: Consumption frequency of fish among fishermen and fish vendors

Sixty-four percent of respondents consume two to three servings of fish per day (Fig. 3.3). These findings are similar to that observed by (Cisneros-Montemayor *et al.,* 2016) where most of the low-income coastal communities, fishermen, and fish vendors depend on fish catches for their daily meal and income. The study further revealed that 50% of respondents consume at least 500 grams of fish per day (Fig. 3.4) with the average amount consumed by respondents estimated to be 400.5 gday^{-1}.

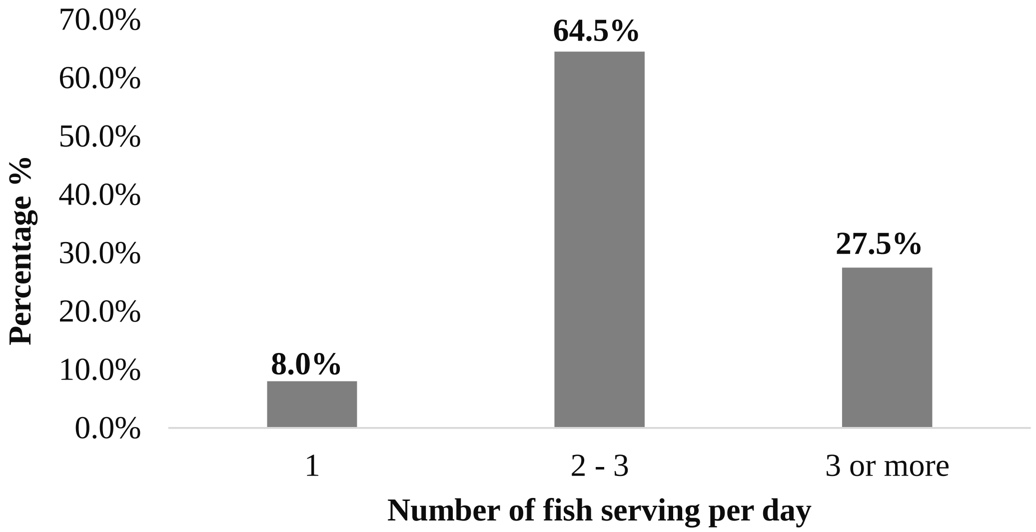

Figure 3.3: Fish consumption servings per day among fishermen and fish vendors

The consumption rate of fisherman and fish vendors was higher than the national per capita fish consumption of 23 gday^{-1} (URT, 2021) by the general population. The consumption trend observed in this study is in line with that observed by Wenaty *et al*. (2018) from fishing communities in areas surrounding Lake Victoria (Tanzania side) where the observed average fish consumption was 370 gday^{-1}. The consumption rate observed in the present study was also higher than the global per capita consumption of 20.5 kg (approx. 56 gday^{-1}) (FAO, 2020).

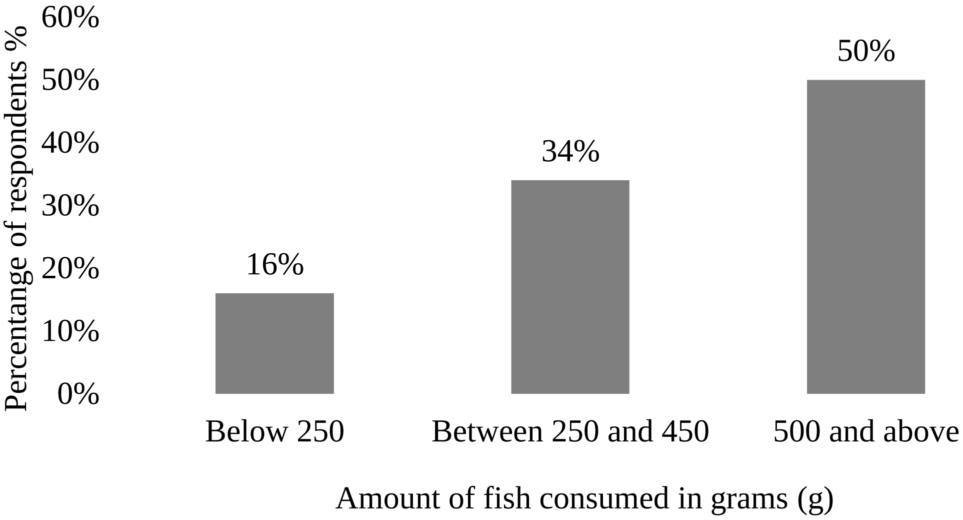

Figure 3.4. Fish consumption amount per day among fishermen and fish vendors

3.3.2: Occurrence of PCBs in analysed fish muscles

The analytical results obtained in this study showed the presence of indicator PCBs in the analysed fish, where 73.33% of *R. kanagutra* samples and 20% of *S.sutor* samples were contaminated with PCBs. The levels of the six indicator PCBs obtained were relatively low in samples of both fish species (Table 3.1) with the highest mean concentration of PCBs ($\sum_6$PCBs) of 3.71 ngg^{-1} w/w. Furthermore, the results showed that PCBs 28 and 52 were below their detection limits (<LOD). The individual mean levels of detected indicator PCBs ranged from <LOD to 1.55 ngg^{-1} w/w, while the sum of the six indicator PCBs ($\sum_6$PCBs) levels ranged from 0.12 to 3.71 ngg^{-1} w/w in both fish species (Appendix 1.2, Appendix 1.3). PCBs levels were dominated by the three congeners PCB 138, 153, and 180 with PCB 153 being the most prevalent. The prevalence of these three congeners is explained by their higher chlorination number which makes them more resistant to degradation in the environment, their ability to bioaccumulate due to their higher lipophilicity, and is less biodegradable in organisms (Lin *et al.,* 2020; Wenaty *et al.,* 2019a; Mwakalapa *et al.,* 2018).

Table 3.1. The mean concentration of PCBs contaminants in fish species *R. kanagutra* and *S. sutor* from the Kivukoni-Feri fish market in ngg^{-1} wet weight (w/w).

		R. kanagutra			*S. sutor*			
PCB Congener	N	Mean	SD	PSD(%)	Mean	SD	PSD(%)	EU – MRL
PCB 28	30	<LOD	-	0	<LOD	-	0	-
PCB 52	30	<LOD	-	0	<LOD	-	0	-
PCB 101	30	0.008	0.029	7	<LOD	-	0	-
PCB 138	30	0.220	0.270	56	0.008	0.032	7	-
PCB 153	30	0.396	0.416	73	0.026	0.072	13	-
PCB 180	30	0.216	0.257	63	0.016	0.042	13	-
∑PCBs	**30**	**0.840**	**0.932**		**0.050**	**0.124**		**75**

Key: N: Total number of samples analysed, EU-MRL: Maximum residue limit set by European Commission (EC, 2011), w/w: Wet weight, LOD: Limit of Detection, SD: Standard deviation, PSD; Percentage of samples PCBs were detected.

3.3.3 Dietary exposure assessment

Human exposure to indicator PCBs via consumption of fish was estimated using mean consumption for normal fish consumers, and using 95th percentile consumption, for high fish consumers in two scenarios, where in the first scenario (lower bound) the overall mean PCBs contamination in both species, 0.45 ngg^{-1} were used and in the second scenario (upper bound) the overall mean PCBs contamination level used was 0.98 ngg^{-1}. The estimated dietary exposure to indicator PCBs were as tabulated in (Table 3.2). The mean exposure of the fishermen and fish vendors to total PCBs was estimated by doubling the exposure estimated by indicator PCBs levels, as total PCBs exposure is assumed to be twice the exposure to the six indicator PCBs (Sirot *et al.,* 2012).

The estimated exposure of the fishermen and fish vendors to the total PCBs were 5.1 and 12.56 ngkg^{-1} bw.day^{-1} in the first scenario for normal and higher fish consumers respectively and 11.18 and 27.56 ngkg^{-1} bw.day^{-1} in the second scenario for normal and higher consumers respectively. The general population exposure to total PCBs was estimated at 0.14 and 0.31 ngkg^{-1} bw.day^{-1} in lower bound and upper bound estimation scenarios respectively. The estimated dietary exposure of the fishermen and fish vendors to total PCBs was higher compared to dietary intake exposure of total PCBs to the general population which was estimated using the national per capita fish consumption rate in both scenarios because fishermen and fish vendors consume a much higher quantity of fish (0.4 kgday^{-1}) compared to the general population (0.023 kgday^{-1}) (URT, 2021). The estimated exposure of fishermen and fish vendors to total PCBs in both scenarios was much higher in high fish consumers, which exceeded the recommended health guideline tolerable daily intake (TDI) for total PCBs of 20 ngkg^{-1} bw.day^{-1} (Bilau *et al.,* 2007; Lin *et al.,* 2020), which indicates the increased potential of occurrence of health risks to high fish-consuming fishermen and fish vendors. The finding shows that fishermen and vendors are 30 times more exposed to PCBs than the general population indicating how fishermen and fish vendors are more vulnerable to health risks related to toxic chemical contamination. Other dietary PCBs intake data from previous studies in Tanzania are limited, but the consumption data of communities involved with fishing activities, observed in the previous study by Wenaty *et al.* (2018) around Lake Victoria in Tanzania indicate the possibility of high intake of environmental contaminants because of their high fish consumption behavior, which was also observed by other previous studies (Cisneros-Montemayor *et al.,* 2016) and Soselisa *et al.,* 2021). Bilau *et al.* (2007) also reported the high risks of exposure to PCBs by recreational fishermen because they consume a high amount of self-caught fish (eels) than the general population and which had a high level of PCBs contamination.

Table 3.2. Estimated overall average daily intake of indicator PCBs for two consumer groups estimated in two possible scenarios

		Dietary Intake (ngkg^{-1}bw.day^{-1}) of six indicator PCBs	
Consumer Group	Scenario	Normal Consumer (Mean)	Higher Consumers (P95)
Fishermen & vendors	LB	2.55	6.28
	UB	5.59	13.78
General population	LB	0.14	
	UB	0.31	

Key: LB: Lower bound scenario, UB: Upper bound scenario, P95: 95th percentile

The study also showed the difference in dietary exposure with the consumption of different fish species. Consumers of *R. kanagutra* were more exposed to PCBs than consumers of *S. sutor* species (Table 3.3), because of the difference in PCBs contamination levels in the respective fish species which were caused by their difference in trophic levels and feeding habits. This finding suggests that fish consumers might be able to reduce their exposure to PCBs without losing the benefits of consuming fish by either diversifying their fish species consumption habit and reducing consumption amount for species such as fatty fish and predator fish species that are more likely to have a high amount of PCBs contamination.

Table 3.3: Estimated average daily intake of indicator PCBs for two consumer groups estimated in two possible scenarios for different fish species in ngkg-^{1}bw.day-^{1}

		R. kanagutra		*S. sutor*	
Consumer Group	Scenario	Normal Consumer (Mean)	Higher Consumers (P95)	Normal Consumer (Mean)	Higher Consumers (P95)
Fishermen & vendors	LB	4.8	18.3	0.29	0.71
	UB	7.31	19.81	3.86	9.5
General population	LB	0.3		0.02	
	UB	0.4		0.22	

Key: LB: Lower bound scenario, UB: Upper bound scenario, P95: 95th percentile

3.3.4 Risk characterisation

3.3.4.1 Lifetime cancer risks assessment

Potential human health risks as a result of consumption of PCB-contaminated fish were estimated using equations 2, 4, and 5, where risks from intake of six indicator PCB were estimated (Table 3.4). The lifetime cancer risk estimated from intake of total PCBs were 1.9E-05 and 4.7E-05 for normal and high fish consumers respectively in the first scenario. In the second scenario, the estimated lifetime cancer risk values were between 2.9E-05 and 7.2E-05 for normal and high fish consumers respectively. The estimated lifetime cancer risks for the general population from intake of total PCBs were 1.1E-07 in the first scenario and 5.0E-07 in the second scenario. For fishermen and fish vendors, the estimated risks for normal and high consumers in both scenarios (lower and upper bound) were above the critical value of 10E-6 which indicates a higher risk of cancer-related health problems and indicates the likely potential for the occurrence of cancer-related health problems to fishermen and fish vendors as a result of consumption of analysed fish species within Dar es Salaam based on the Agency for Toxic Substances and Disease Registry (ATSDR) risk estimation criteria (ATSDR, 2018). However, the estimated lifetime cancer risks for the general population in both scenarios were below the recommended critical value of 10E-6, indicating that they are within recommended health guidelines for lifetime PCBs exposure based on the ATSDR risk estimation criteria, because of the lower amount of fish they consume in average in comparison to the fishermen and vendors.

Table 3.4: Lifetime cancer risks (LCR) for two consumer groups estimated from intake of six indicator PCBs in two possible scenarios

		Lifetime cancer risks (LCR) from intake of indicator PCBs	
Consumer Group	Scenario	Normal Consumer (Mean)	Higher Consumers (P95)
Fishermen & vendors	LB	5.1E-06	1.3E-05
	UB	1.1E-05	2.8E-05
General population	LB	2.9E-07	
	UB	6.3E-07	

Key: LB: Lower bound scenario, UB: Upper bound scenario, P95: 95th percentile

3.3.4.2 Non-carcinogenic health risks

The estimated hazard indices (HI) from intake of the six indicator PCBs for fishermen and fish vendors are indicated in (Table 3.5). Hazard indices estimated from intake of total PCBs ranged from 0.48 to 1.81 for normal and high fish consumers in lower and upper bound scenarios respectively, while the estimated hazard indices for the general population from intake of total PCBs ranged from 0.027 to 0.041 in lower and upper bound scenarios respectively. The estimated hazard indices from intake of total PCBs for normal fish consumers were higher than 0.1, which indicate that low non-carcinogenic health risks might occur (ATSDR, 2018; Wenaty *et al.,* 2019a; Lin *et al.,* 2020) to normal fish consumer in fishermen and fish vendors communities in both estimation scenarios (lower and upper bound). Hazard indices for high fish consumers in both scenarios were above the critical value of 1, indicating higher risks of occurrence of non-carcinogenic health problems for high fish-consuming fishermen and fish vendors than for normal fish consumers due to the high intake of total PCBs. However, for the general population, non-carcinogenic health risks estimated from intake of total PCBs were much lower, less than 0.1, which suggests that there are no risks of non-carcinogenic health problems occurring as a result of consuming the studied fish species in the study area based on the Agency for Toxic Substances and Disease Registry (ATSDR) risk estimation criteria (ATSDR, 2018).

Table 3.5. Hazard index (HI) for non-carcinogenic health risks for two consumer groups estimated from intake of six indicator PCBs in two possible scenarios

		Non-carcinogenic health risks from intake of indicator PCBs	
Group	Scenario	Normal Consumer (Mean)	Higher Consumers (P95)
Fishermen & vendors	LB	0.127	0.314
	UB	0.279	0.689
General population	LB	0.007	
	UB	0.016	

Key: LB: Lower bound scenario, UB: Upper bound scenario, P95: 95th percentile

3.4 Conclusion

The present study findings show that exposure levels due to intake of PCBs are a cause of serious concern among the artisanal fishermen and fish vendors community in Dar es Salaam. Fishermen and fish vendors are more vulnerable to exposure to unacceptable levels of PCBs of toxicological relevance in comparison to the general population because they consume a higher amount of fish. The general population hazard indices were much lower, indicating that lifetime consumption of analysed fish in the current PCBs levels would have no significant health effects. Further studies are recommended to include a wider range of fish species along the Tanzania coastal waters, to have more information that can help in the formulation of strategies to minimize PCBs exposure to consumers.

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CHAPTER FOUR

4.0 General discussion

4.1. Food frequency survey

The preference of *R. kanagutra* and *S. sutor* for consumption by fishermen and fish vendors (Fig. 3.1) in the study area may be due to their availability in the daily catch and lower prices (Breuil and Bodiguel, 2015). This makes these species more affordable to be used as a meal while other species in the catch can be sold. *R. kanagutra* is one of the most caught fish species and is economically important in countries along the East coast of the Indian Ocean including Tanzania (Mziray and Kimirei, 2016; Sekadende *et al*., 2020, 2021). Furthermore, the consumption of fish on most days within a week by respondents (Fig. 3.2), may be due to awareness of the health benefits associated with the consumption of fish and other white meat (Wenaty *et al*., 2018). The majority of respondent consumes two to three servings of fish per day (Fig. 3.3), which is similar to the findings observed by Cisneros-Montemayor *et al*. (2016) where most of the low-income coastal communities, fishermen, and fish vendors depends on fish catches for their daily meal and income. The consumption rate of fishermen and fish vendors (average of 400.5 $gday^{-1}$) was higher than the national per capita fish consumption (Fig. 3.4) of 23 $gday^{-1}$ (URT, 2021). The consumption trend observed in this study is in line with that observed by Wenaty *et al*. (2018) from a study in fishing communities in areas surrounding Lake Victoria (Tanzania side) where the observed average fish consumption was 370 $gday^{-1}$. The consumption rate observed in the present study was also higher than the global per capita consumption of 20.5 kg (approx. 56 $gday^{-1}$) (FAO, 2020).

4.2 Occurrence and levels of PCBs in fish muscles

The levels of the six indicator PCBs obtained were relatively low in samples of both fish species (Table 3.1) where the PCBs levels were dominated by the three congeners PCB 138, 153, and 180, with PCB 153 being the most prevalent. The prevalence of the these congeners can be explained by their higher chlorination number which makes them more resistant to degradation in the environment, their ability to bioaccumulate due to their higher lipophilicity, and are less biodegradable in organisms (Lin *et al.,* 2020; Wenaty *et al.,* 2019a; Mwakalapa *et al.,* 2018). These findings are similar to that observed by (Mwakalapa *et al.,* 2018, Haar *et al.,* 2021; Shang *et al.,* 2016) where PCB 153 congener was more dominant, but they are slightly different from studies from other locations in Tanzania (Wenaty *et al.,* 2019a; Polder *et al.,* 2014) where PCB 138 congener was more dominant congener, which suggests contamination originated from different sources of PCBs (Mwakalapa *et al.,* 2018).

Statistical analysis showed a significant difference in the mean PCBs levels between the analysed species samples at ($p< 0.05$). The mean PCBs concentration of indicator PCBs congener in *R. kanagutra* was significantly higher than *S. sutor* (Appendix 1.2). However, the PCBs contamination levels observed were below the maximum residue limits (MRL) of 75 ngg^{-1}w/w set by the European Commission, for fish meat and fishery products (EC, 2011). The higher concentration of PCBs in *R. kanagutra* fish species than in *S. sutor* species was probably due to the difference between their feeding habit and that *R. kanagutra* have higher trophic levels than *S. sutor* (Fishbase, 2017). *R. kanagutra* are omnivorous feeding on other small organisms such as polychaetes, ostracods, and small fishes such as anchovy and sardines which results in biomagnifacation (Mziray and Kimirei, 2016; Fishbase, 2017; Hakimelahi *et al.,* 2020) while the *S. sutor* are herbivorous fish feeding on seaweed and seagrasses (Mziray and Kimirei, 2016; Fishbase, 2017). These findings have a similar trend to results obtained by other studies (Shen *et al.,* 2017; Panseri *et al.,* 2019) that showed omnivorous fish and other predator fish (carnivorous) have more contamination levels of PCBs than herbivorous fish because of biomagnification. The findings also showed that the concentration of PCBs levels in marine fish in this study was relatively higher than that of fish from freshwater in Tanzania, where Polder *et al.* (2014) reported that tilapia from Lake Tanganyika (0.57 ngg^{-1} w/w), Lake Nyasa (0.02 ngg^{-1} w/w) and Lake Victoria (0.08 ngg^{-1} w/w) had lower PCBs levels than that obtained in the present study (0.84 ngg^{-1} w/w) with the same feeding habits (*R. kanagutra*). This trend is similar to other reported studies where PCBs contamination levels in marine fish were found to be higher than that obtained from freshwater fish (Ahmed *et al.,* 2016; Shang *et al.,* 2016) probably due to the vast amount of waste discharged into the ocean from multiple source channels and rivers from different parts of the country far from the ocean. However, the PCBs levels obtained in this study were lower than that obtained by other studies for marine fish with the same feeding habits in India (65.8 ngg^{-1} w/w) by Ahmed *et al.* (2016) and China (3.82 ngg^{-1} w/w) by Shang *et al.* (2016) involving *R. kanagutra* fish species which suggest higher level of contamination because of higher level of industrialisation of the areas which may lead to higher environment pollution.

4.2 Dietary exposure assessment

The estimated exposure of the fishermen and fish vendors to total PCBs were higher compared to the dietary exposure of the general population to total PCBs. The estimated exposure of fishermen and fish vendors to total PCBs in both scenarios was much higher in high fish consumers, where PCBs exposure exceeded the recommended health guideline of tolerable daily intake (TDI) for total PCBs of 20 ngkg^{-1}bw.day^{-1} (Bilau *et al*., 2007; Lin *et al*., 2020). These findings indicated the increased potential of occurrence of health risks to high fish-consuming fishermen and fish vendors. The fishermen and vendors are estimated to be 30 times more exposed to PCBs than the general population which indicates how fishermen and fish vendors are more vulnerable to health problems related to toxic chemical pollutants. Other dietary PCBs intake data from previous studies in Tanzania are limited, but the consumption data of communities involved with fishing, as observed in a previous study by Wenaty *et al.* (2018) around Lake Victoria in Tanzania indicate the possibility of high intake of environmental contaminants because of their high fish consumption behaviour, which is also observed by other previous studies Cisneros-Montemayor *et al*. (2016) and Soselisa *et al*. (2021). Bilau *et al.* (2007) also reported the high risks of exposure to PCBs by recreational fishermen because they consume a high amount of self-caught fish (eels) than the general population and which had a high level of PCBs contamination.

4.3 Risk characterisation

Estimated lifetime cancer risks among the general population for adults and children were observed to be below the critical values of 10E-6 for adults and 10E-04 for children (Lin *et al.,* 2020), which indicates that consumption of the *R.kanagutra* and *S. sutor* fish species from the Kivukoni Feri fish market will have negligible adverse health effects. The hazard indices (HI) values used to estimate the non-carcinogenic health risks were lower than 0.1 indicating that there are no non-carcinogenic risks that are likely to occur as a result of consumption of the *R.kanagutra* and *S. sutor* fish species from the study area (Paper 1).

However, the estimated risks for normal and high consumers in both scenarios of estimation (lower and upper bound) for fishermen and fish vendors, were above the critical value of 10E-6 (Table 3.4) which indicates the likelihood of the occurrence of cancer-related health problems for fishermen and fish vendors as a result of consumption of analysed fish species within the study area based on the Agency for Toxic Substances and Disease Registry risk estimation criteria (ATSDR, 2018). The estimated lifetime cancer risks for the general population in both scenarios were below the recommended critical value of 10E-6, indicating that the general population would have insignificant health risks because of the lower amount of fish they consume on average (23 gday^{-1}) in comparison to the fishermen and vendors (400.5 gday^{-1}). The estimated hazard indices from intake of total PCBs for the normal fish consumer were higher than 0.1, which indicate that low non-carcinogenic health risks might occur to normal fish consumer in fishermen and fish vendors communities in both estimation scenarios (lower and upper bound) (ATSDR, 2018; Wenaty *et al*., 2019b; Lin *et al*., 2020), while, the indices for high fish consumers in both scenarios were above the critical value of 1, indicating higher risks of occurrence of non-carcinogenic health problems for high fish-consuming fishermen and fish vendors than for normal fish consumers due to the high intake of total PCBs. However, for the general population, non-carcinogenic health risks estimated from intake of total PCBs were much lower, less than 0.1, which suggests that there are no risks of non-carcinogenic health problems that can occur as a result of consuming the analysed fish species from the study area based on the estimation criteria of the Agency for Toxic Substances and Disease Registry risk (ATSDR, 2018).

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CHAPTER FIVE

5.0 Conclusion and Recommendations

5.1 Conclusions

The present study assessed the occurrence, levels, dietary exposure, and probable human health risks of polychlorinated biphenyls to fishermen and fish vendors and the general population, in Dare es Salaam city, Tanzania by analysis of indicator polychlorinated biphenyls in selected marine fish species *R. kanagutra* and *S. sutor.* The findings showed that there is the presence of PCBs contamination of marine fish in the study area due to the historical release of PCBs. The detected concentration of the six indicator PCBs was relatively low in both fish species with the highest mean concentration ($\sum_6$PCBs), 3.71 ngg^{-1} w/w observed in *R. kanagutra*. The mean contamination levels of indicator PCBs in *R. kanagutra* were found to be statistically different from that of *S. sutor* fish species due to the differences in their trophic levels and feeding habits. However, the detected PCBs levels in both fish species were below the maximum residue limits (MRL) of 75 ngg^{-1}w/w set value by the European Commission (UC).

The average daily intake of total PCBs by high fish consumer fishermen and fish vendors exceeded recommended tolerable daily intake, (TDI) of 20 ngkg^{-1} bw.day^{-1} where estimated dietary exposure of fishermen and fish vendors to total PCBs ranged between 5.10 and 27.56 ngkg^{-1} bw.day^{-1} for normal and higher fish consumers depending on the estimation scenario, while that of general population ranged between 0.14 and 0.31 ngkg^{-1} bw.day^{-1} in lower and upper estimation scenario respectively.

Estimated lifetime cancer risks were above the recommended critical value of 10E-6, based on the Agency for Toxic Substances and Disease Registry (ATSDR) set criteria, for fishermen and fish vendors from intake of total PCBs where the estimated values were 1.9E-05 and 4.7E-05 for normal and high fish consumers respectively in the first scenario and between 2.9E-05 and 7.2E-05 for normal and high fish consumers in the first scenario. Estimated hazard indices indicating non-carcinogenic health risks, ranged from 0.48 to 1.81 for normal and o high fish consumers in respective estimation scenarios. The estimated hazard indices from intake of total PCBs for the normal fish consumer were lower than 0.1, which indicates low chances for the occurrence of non-carcinogenic health risks in the long run while for high fish consumers were above the critical value of 1, indicating higher risks of occurrence of non-carcinogenic health problems based on ATSDR risks estimation criteria.

However, the estimated value of lifetime cancer risks for the general population from intake of total PCBs was between 1.1E-07 and 5.0E-07 in the first scenario and second scenario respectively, below the recommended critical value of 10E-6 based on the ATSDR, while the estimated hazard indices for non-carcinogenic health risk were between 0.027 and 0.041 in both lower and upper bound scenarios which are below the critical value of 0.1 indicating very low non-carcinogenic health risk based on ATSDR risks estimation criteria meeting the recommended health guidelines for the average daily exposure to PCBs for the general population.

5.2 Recommendations

The findings of this study show the presence of PCBs contamination in marine fish which can likely result in unacceptable adverse health effects for consumers. Therefore, the study calls for;

i. Regular monitoring of PCBs contamination in environments including previously identified hot spots (storage facilities and their surroundings) and also in the foodstuff that is the main contributor to PCBs exposure (meat, fish, dairy products) by the relevant regulatory authorities (GCLA and NEMC)

ii. Awareness training among fish consumers on the potential adverse health risks of PCBs and the different ways to minimize exposure such as species diversifying and reducing consumption of fish species that are more vulnerable to PCBs contamination such as fatty fish, predator (carnivorous) and omnivorous fish while consuming more of herbivorous and low-fat fish species.

iii. Sensitization and training of maintenance workers and technicians working on PCBs-containing equipment about the effects of PCBs and the need to avoid new environment contamination through leakage and poor management of PCBs-containing equipment.

iv. Further studies on the assessment of marine fish species contamination along the Tanzanian coastal waters be carried out to assess get the full picture of contamination so that appropriate measures and cost-effective strategies to minimize PCBs exposure to the consumer are carried out.

APPENDICES

Appendix 1: Summary of analytical results

PCB Congener	LOD	LOD	Recovery (%)	Units
PCB 28	0.05	0.20	119.0	ng/g
PCB 52	0.08	0.23	104.0	ng/g
PCB 101	0.03	0.11	77.0	ng/g
PCB 138	0.05	0.12	85.0	ng/g
PCB 153	0.03	0.12	85.5	ng/g
PCB 180	0.02	0.11	83.0	ng/g

Appendix 2: *Rastrelliger kanagutra* (Indian Mackerel)

S/N	Sample ID	PCB 28	PCB 52	PCB 101	PCB 138	PCB 153	PCB 180	∑PCBs	Units
1	M01	<LOD	<LOD	<LOD	0.30	0.46	0.25	1.02	ng/g
2	M02	<LOD	<LOD	<LOD	0.19	0.34	0.16	0.69	ng/g
3	M03	<LOD	<LOD	<LOD	0.30	0.68	0.29	1.27	ng/g
4	M04	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	ng/g
5	M05	<LOD	<LOD	<LOD	<LOD	0.15	0.12	0.28	ng/g
6	M06	<LOD	<LOD	0.12	0.37	0.40	0.30	1.19	ng/g
7	M07	<LOD	<LOD	<LOD	0.33	0.64	0.28	1.24	ng/g
8	M08	<LOD	<LOD	<LOD	0.38	0.65	0.40	1.43	ng/g
9	M09	<LOD	<LOD	<LOD	0.23	0.41	0.22	0.86	ng/g
10	M10	<LOD	<LOD	<LOD	0.18	0.56	0.17	0.91	ng/g
11	M11	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	ng/g
12	M12	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	ng/g
13	M13	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	ng/g
14	M14	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	ng/g
15	M15	<LOD	<LOD	<LOD	0.24	0.35	0.26	0.86	ng/g
16	M16	<LOD	<LOD	<LOD	0.13	0.17	0.12	0.42	ng/g
17	M17	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	ng/g
18	M18	<LOD	<LOD	<LOD	<LOD	0.67	0.37	1.04	ng/g
19	M19	<LOD	<LOD	<LOD	0.68	1.33	0.66	2.67	ng/g
20	M20	<LOD	<LOD	<LOD	<LOD	0.12	<LOD	0.12	ng/g
21	M21	<LOD	<LOD	<LOD	<LOD	0.15	<LOD	0.15	ng/g
22	M22	<LOD	<LOD	<LOD	0.12	0.11	0.11	0.33	ng/g
23	M23	<LOD	<LOD	0.11	0.75	1.24	0.67	2.78	ng/g
24	M24	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	ng/g
25	M25	<LOD	<LOD	<LOD	0.39	0.49	0.31	1.19	ng/g
26	M26	<LOD	<LOD	<LOD	1.07	1.55	1.09	3.71	ng/g
27	M27	<LOD	<LOD	<LOD	<LOD	0.27	<LOD	0.27	ng/g
28	M28	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	ng/g
29	M29	<LOD	<LOD	<LOD	0.37	0.36	0.16	0.89	ng/g
30	M30	<LOD	<LOD	<LOD	0.57	0.79	0.53	1.89	ng/g

Appendix 3: *Siganus sutor* (Spinefoot Rabbitfish)

S/N	Sample ID	PCB 28	PCB 52	PCB 101	PCB 138	PCB 153	PCB 180	∑PCBs	Units
1	R01	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	ng/g
2	R02	<LOD	<LOD	<LOD	0.14	0.28	0.12	0.54	ng/g
3	R03	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	ng/g
4	R04	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	ng/g
5	R05	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	ng/g
6	R06	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	ng/g
7	R07	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	ng/g
8	R08	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	ng/g
9	R09	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	ng/g
10	R10	<LOD	<LOD	<LOD	0.10	<LOD	0.13	0.23	ng/g
11	R11	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	ng/g
12	R12	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	ng/g
13	R13	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	ng/g
14	R14	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	ng/g
15	R15	<LOD	<LOD	<LOD	<LOD	0.13	<LOD	0.13	ng/g
16	R16	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	ng/g
17	R17	<LOD	<LOD	<LOD	<LOD	0.24	0.12	0.36	ng/g
18	R18	<LOD	<LOD	<LOD	<LOD	0.13	<LOD	0.13	ng/g
19	R19	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	ng/g
20	R20	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	ng/g
21	R21	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	ng/g
22	R22	<LOD	<LOD	<LOD	<LOD	<LOD	0.12	0.12	ng/g
23	R23	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	ng/g
24	R24	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	ng/g
25	R25	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	ng/g
26	R26	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	ng/g
27	R27	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	ng/g
28	R28	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	ng/g
29	R29	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	ng/g
30	R30	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	ng/g

Appendix 4: Questionnaire for the assessment of public awareness of the health risks associated with the consumption of PCBs contaminated fish and quantities of fish consumed by household

Site Name: Kivukoni-Feri Fish Market **Region:** Dar es Salaam **District:** Ilala **Ward:** Kivukoni

Date of the interview …………………

Part A: Personal Information

1) Name: ……………………………………………………
2) Age: ………………………….
3) Sex: ……… …………………..
4) Education: ………………………………………………...
   a) No school
   b) Primary school
   c) Ordinary Secondary school
   d) Advanced Secondary school
   e) College (Diploma/Degree)
5) Experience (*how long working in the current job*):
   a) 1 – 6 months
   b) 6 – 12 months
   c) More than 12 months

Part B: Awareness of PCBs

1. Do you have any knowledge about the chemical wastes issues in the ocean especially polychlorinated biphenyls (PCBs), YES / NO
2. If the answer is yes, where did you receive this information?
   a. Newspaper
   b. Radio
   c. Television (TV)
   d. Internet
   e. Training
   f. Other means (explain) ……………………………………………………

3. Do you have any knowledge about the sources of polychlorinated biphenyls (PCBs) you know?
   a. Industries waste
   b. Municipal wastes
   c. Electric transformers and capacitors
   d. Others (specify)…………………………………………………………………….
   e. I don't know

4. Do you know any harmful health effects that can be caused by the consumption of PCBs-contaminated fish?
   a) Reproductive system impairment such as poor sperm quality and miscarriage
   b) Increased risk of prostate and breast cancer
   c) Endocrine system disruption
   d) Developmental effects in breastfed infants
   e) Others (specify)…………………………………………………………….
   f) I don't know

5. Does the management of the market conduct awareness training on PCBs and their potential effects on human health? YES / NO

6. If YES what was the training about?
    a) What are PCBs, cause and effects on health
    b) Control of leakage during maintenance
    c) Disposal of wastes suspected to contain PCB
    d) Storage of equipment containing PCB
    e) Others (specify) …………………………………………………………
    f) N/A

Part C: Fish Consumption Habit

7. Does your household use fish in as part of the meal? YES / NO
8. If YES, how many times per week do you consume fish?
    a) Once per week
    b) Twice per week
    c) Three time per week
    d) Four times per week
    e) Five times per week
    f) Every day of the week

9. If answer to question 7 is NO or once or twice per week, give the reason (s)
    a) Financial cost
    b) Health/Allergy
    c) Tradition taboos/beliefs
    d) Others (specify)

10. How many servings of fish does your household eat each day?
    a) 1- 2
    b) 3 – 4
    c) 5 – 6
    d) 7 or more

11. For how long does the household have been eating this number of daily servings of fish?
    a) Less than one month
    b) 1 - 3 months
    c) 4 - 6 months
    d) Longer than 1 year

12. Which type of fish do you usually eat?
    a) Mackerel
    b) Rabbitfish
    c) Others (specify) …………………………………………….

13. What is the reason for the answer in question (12) above?
    a) Price
    b) Nutrition value
    c) Taste
    d) Availability
    e) Easy to prepare

  f) Others (specify)

14. How many fish (number or in grams) on average do you consume per day? ....

15. Which parts of the fish do you eat?
    a) Fillets (muscles only)
    b) All parts (Fillet and Ofals)

16. Do you eat fish caught from other area apart from Kivukoni Feri Fish Market? YES / NO

17. How often do you eat fish caught from other areas apart from Kivukoni Feri Fish Market?
    a) Once per week
    b) Twice per week
    c) Three times per week
    d) Four times per week
    e) Five times per week
    f) Every day of the week
    g) Never

18. For each fish species listed, put a tick in a correct response indicating how often on average the household used last month

Type of fish	< 1/month	1 – 3/ month	1/ week	2-4/ week	5-6/ week	1/ day	2-3/ day	4– 5/day	>6/ Day
Mackerel									
Rabbitfish									
Others									

THANK YOU FOR YOUR PARTICIPATION