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## RESEARCH PAPER

## OPEN ACCESS

## Heavy metals and *lambda*-cyhalothrin levels in prawns, water and sediments along the Indian Coastline of Tanzania

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### Abstract

Heavy metals and pesticide residues in prawns, water and sediments, and sources of contamination were determined. Prawns, sediments and water samples were collected from agricultural farms and estuarine, along the coastline of Indian Ocean in Kisiju, Bagamoyo and Kilwa zones in Tanzania. The concentration of heavy metals; zinc (Zn), lead (Pb) and cadmium (Cd) was determined using Atomic Absorption Spectrophotometry (AAS), and Direct Mercury Analyzer (DMA) for mercury (Hg). Gas Chromatography-Mass Spectrophotometry (GC-MS) was also used to analyze for pesticide residues. Data on demographic characterization and sources of aquatic pollution were obtained through structured questionnaires. Results indicated that, fertilizers (60.7%), pesticides (59.8%) and herbicides (60.7%) were the main sources of aquatic pollution. *Cyberdip* and *Utupa* were the chemicals used in prawns. The distribution of heavy metals mean concentrations in prawns and sediments were found to be in order Zn>Pb>Cd>Hg, and Pb>Zn>Cd>Hg in water. *Lambda*-cyhalothrin was the only pesticide residue detected in prawns and sediments and not detected in water. Significant high levels of *lambda*-cyhalothrin in sediments were detected from Kisiju (16.49±2.36mg/kg) and Kilwa (12.21±3.24mg/kg) respectively. On the other hand, lowest *Lambda*-cyhalothrin level in prawns was detected in Kisiju (2.26±0.51mg/kg) and Kilwa (0.28±0.40mg/kg). Presence of heavy metals and *Lambda*-cyhalothrin indicate marine pollution. Marine pollutants may impair quality and safety of prawns and by-products to the public health. Monitoring of agricultural and other anthropogenic activities in the area especially in the Rufiji delta and around the mangroves where prawns spawn is inevitable.

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## Introduction

Heavy metals and pesticide residues are of global concern as they pose adversely affects to the ecosystem and human health (Marković *et al.*, 2010). Trace metals are always very essential at low levels but usually very toxic at higher concentrations (Rainbow, 2007). Human get affected through food chain causing various diseases and damages (Pandey and Madhuri, 2014). Both natural and anthropogenic activities attributes to heavy metals and pesticide residues in the environment. Increased pollution of heavy metals and pesticide residues have significant adverse health effects for invertebrates, fish, and humans (Islam and Tanaka, 2004). Heavy metal pollution in aquatic ecosystems is increasing due to the effects from urbanization and industrialization toward pollution (Lin *et al.*, 2009). Heavy metals pollution have been proven as health risks to more than 10 million people in various countries (Muhammad *et al.*, 2013). Fish is a potential indicator of presence of traces of heavy metals pollution (Bernet *et al.*, 1999).

Bioaccumulation of metal toxins in the food chain also results to disastrous effects on human health (Shakoor *et al.*, 2013). Some inorganic elements found in nature, such as cadmium (Cd), mercury (Hg), lead (Pb) and arsenic (As), are considered potentially dangerous for human health if absorbed beyond certain limits (Shakoor *et al.*, 2013). For instance, in Linfen China, people faced extreme loads of pollution, in Haina of Dominican Republic people suffered from a huge amount of lead poisoning, and in Ranipet a city of India, about 3.5 million people were affected by tannery waste (Shakoor *et al.*, 2013). General signs of these heavy metals (Cd, Zn, Hg, copper (Cu), arsenic (Ar) and Pb) includes gastrointestinal (GI) disorder, diarrhea, stomatitis, tremor, hemoglobinuria causing rust red color to stool, ataxia, paralysis, vomiting and convulsion, depression and pneumonia and the effect could be neurotoxic, carcinogenic, mutagenic or teratogenicity (Duruibe *et al.*, 2007). Sediments have been widely used as environmental indicators to assess metal pollution in the natural water (Förstner and Wittmann, 2012).

Deposited heavy metals and pesticide residues in a river system through different methods including rains undergo changes due to dissolution, precipitation and sorption which affect their performance and bioavailability (Alloway and Ayres, 1997). Furthermore, presence of heavy metals and pesticide residues in prawns, water and sediments indicates waste discharges on the riverine ecosystems from agricultural, anthropogenic and industrial sources (Bayen, 2012). Nowadays, heavy metals and pesticide residues pollution is a main problem in many developing countries (Odada *et al.*, 2004) Tanzania inclusive. High concentration of heavy metals such as Aluminum (Al), Zn, iron (Fe), Cu, cobalt (Co), chromium (Cr), Cd and Pb were reported to be discharged into the coastal area of Tanzania from agricultural, anthropogenic and industrial sources, which in turn pollutes marine ecosystems (Bonga *et al.*, 2008).

The Indian Ocean has high biodiversity hotspot of marine fauna and flora species including corals, mangroves, sea grasses, fishes, marine mammals, turtles, crustaceans and mollusks (Keesing and Irvine, 2005). Prawns have been harvested for a long time using tradition fishing techniques, marketed and consumed locally, as well as for export (Silas, 2011).

Bagamoyo, Kilwa and Kisiju zones are well known for prawns production and fishing along the Indian coastline of Tanzania. Fishing is the major source of daily income and food for people living along the coastline of Tanzania, more specifically in Bagamoyo area. The Wami, Ruvu and Pangani river estuaries also provide wide range of diversity of fish to Bagamoyo residents. Likewise, Kilwa and Kisiju zones are fed by fish from the Rufiji delta within the coastline of the Indian Ocean of Tanzania (Shaghude, 2004). In the Rufiji delta, various agricultural activities are simultaneous carried out with prawns farming. Pesticide residues from the rice farms in mangrove areas have been reported to affect the performance of prawns and jeopardize the health of people eating prawns (Bonga *et al.*, 2008).

According to Sarkar *et al.*, 2016, heavy metals and pesticide residues are potentially gathered in aquatic environments including prawns, water, and sediments. A study by (Machiwa, 1992) has reported accumulation of pesticides such as PAHs, PCBs, and heavy metals, in the coastal sediments. Heavy metals and pesticide residues in aquatic environment have been reported worldwide (Machiwa, 1992; Saha *et al.*, 2016; Sarkar *et al.*, 2016; Sobihah *et al.*, 2018; Wandiga *et al.*, 2002).

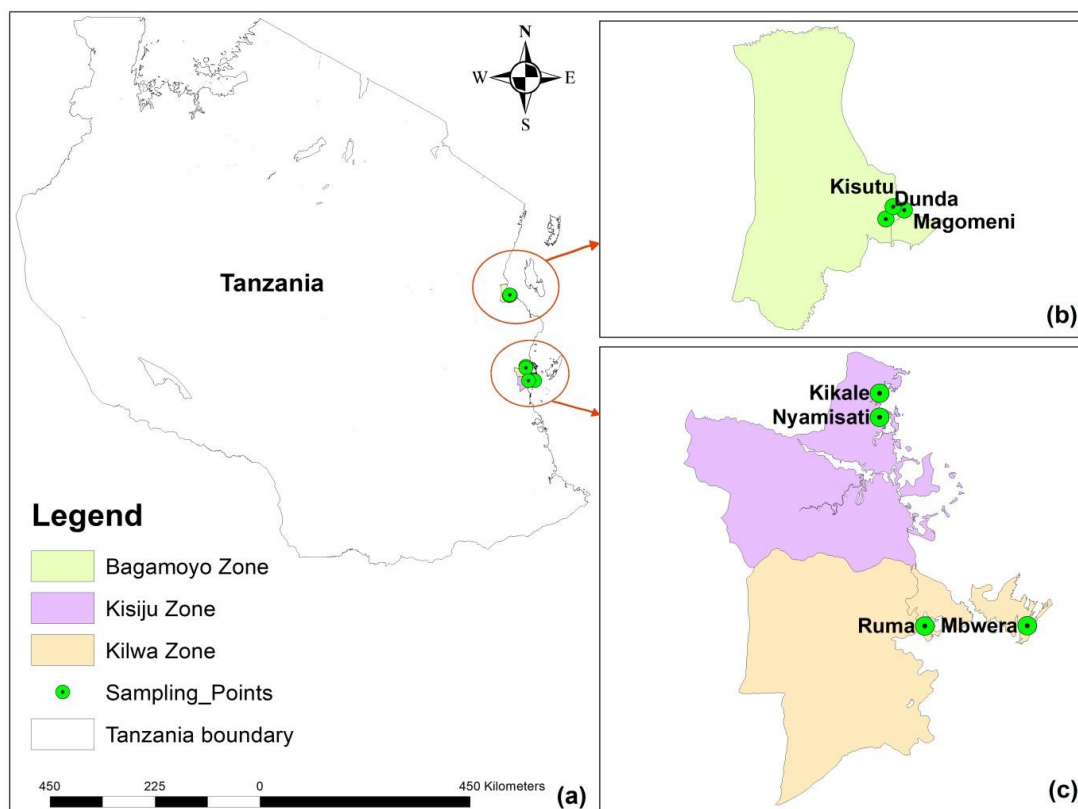
Prawns are the most important source of income and protein to the human populations in the Coast region of Tanzania. Heavy metals and pesticides contamination can be dangerous to public health as they can compromise with safety and quality of the prawns (Pigott, 2017). Rumisha *et al.*, (2016) is the only recent report on heavy metals contamination in prawns in Dar-es-Salaam, Tanzania. Besides, no studies have focused on heavy metal and pesticide residues in Bagamoyo, Kisiju and Kilwa zones. In other words, to date, there is no scientific research done on heavy metal pollution and pesticide residue in prawns, water and sediments in the area.

In this study, concentration of heavy metals and pesticide residues in prawns, sediments and water were determined and possible sources of contaminants assessed.

## Materials and methods

### Study area

This study was conducted in Bagamoyo, and Kibiti Districts in Coast region of Tanzania. The two coastal districts encompass three big prawns fishing zones of Bagamoyo (Zone 1), Kisiju (Zone 2) and Kilwa (Zone 3) (Fig. 1). The selected zones are potential for prawns production and fishing. Also, contributes to about 25-45% of the total annual prawn catches in Tanzania (PMP, 2012). Purposive sampling was done to select seven villages three in Zone 1 and two in each zones 2 and 3) interviews were conducted and samples collected. The dominant prawn species found are *Penaeus indicus* and *Metapenaeus monoceros* (Zones 1 & 2), and *Metapenaeus monoceros*, *Penaeus semisulcatus* and *Penaeus indicus* in zone 3 respectively. Bagamoyo zone is fed by Ruvu; Wami and Pangani rivers, whereas Kisiju and Kilwa zones receive fresh water from Rufiji river.



**Fig. 1.** Map of the prawns fishing zones along the coastline of Tanzania.

### *Chemicals and reagents*

Nitric acid (HNO<sub>3</sub>), perchloric acid (HClO<sub>4</sub>), dichloromethane (CH<sub>2</sub>Cl<sub>2</sub>) and cyclohexane (C<sub>6</sub>H<sub>12</sub>) were obtained from Blulux Lab-Pvt Ltd, India. Anhydrous sodium sulphate (Na<sub>2</sub>SO<sub>4</sub>), hydrogen peroxide (H<sub>2</sub>O<sub>2</sub>) and sulphuric acid (H<sub>2</sub>SO<sub>4</sub>) were obtained from Sentmenat, Spain. All other chemicals and reagents used in this study were of analytical grade.

### *Sampling*

#### *Interviews*

Pilot survey was done to 30 respondents to test the relevance of the questions, i.e., how easy or difficult were the questions to the respondents, especially on information about sources of pollutants from the study areas. A purposive sampling was done to select 7 villages (3 in Bagamoyo, 2 in Kilwa and 2 in Kisiju) where the samples were collected. About 20 respondents from each village were randomly selected and interviewed using structured questionnaires. A total of 122 willingly respondents were interviewed.

#### *Collection of prawn, sediment and water samples*

Water, sediments and prawns samples were collected from 15<sup>th</sup> May to 15<sup>th</sup> June 2018. Surface water samples were collected from about 10 cm below water surface using hand grab method and stored in 500mL sterile glass bottles. Fifty gram of NaCl was added to the samples and capped with caps lined with Teflon to preserve it. About 30g of sediments was collected from the same locations where water samples were taken and placed in an aluminum foil. Furthermore, about 30g of prawns was collected from the same spots and placed in an aluminum foil and preserved in a cooler box. All the samples were then stored in a separate cool box with ice cubes at -4°C and immediately transported to the laboratory for further analysis.

#### *Sample preparation*

Sediments and prawn's samples were dried in an oven at 80°C separately until constant weight was reached. The samples were later grounded with mortar and pestle, sieved using laboratory test sieve with an aperture size of 125µm and diameter of 2mm and preserved in a desiccator. Water samples were filtered for dissolved metal concentrations using 0.45µm membrane filters, (Merck, South Africa) and later

acidified with 0.24M HNO<sub>3</sub> and kept at 4°C in the dark until further analysis.

### *Heavy metal analysis*

#### *Prawns*

Five grams of dry weight of prawn's powder was placed in a 250mL beaker with a solution of 5mL HNO<sub>3</sub> and 5mL H<sub>2</sub>SO<sub>4</sub>. When the prawn's tissue stopped reacting with the acids, the mixture was heated in a hot plate at 60°C for 30 min. The mixture was cooled at room temperature and 10mL of HNO<sub>3</sub> (65%) was added and re-heated slowly to 120°C. The temperature was increased to 150°C and the mixture was removed from the hot plate when the sample turned black. The mixture was then allowed to cool at room temperature and 30% H<sub>2</sub>O<sub>2</sub> was added until the sample become clear. Then, the content from the beaker was transferred to 50mL volumetric flask and diluted to the mark with deionized water (Elmer, 1984).

#### *Sediments*

Two grams of dry weight sediments was placed in a 250mL beaker with a solution of 10mL HNO<sub>3</sub> and 10mL HCl. When the sediments stopped reacting with acids, the mixture was heated in a hot plate at 60°C for 30 min. After cooling at room temperature, 10mL of HClO<sub>4</sub> was added and re-heated at 100°C for 5 minutes then allowed to cool at room temperature. The mixture was then transferred to a 50mL volumetric flask and diluted to the mark with deionized water and filtered with a 125µm Whatman filter paper (Amicon, USA) similar to (Saria, 2016).

#### *Water*

Hundred milliliters of water sample was poured in a 250mL beaker with a solution of 10mL HNO<sub>3</sub> and 10mL HCl. When the water stopped reacting with acids, the mixture was heated on a hot plate at 60°C for 30 min until brown fumes were formed. The mixture was allowed to cool at room temperature and 10mL of HClO<sub>4</sub> was added, then reheated for 5 minutes and allowed to cool. The content from the beaker was transferred to 50mL volumetric flask and diluted to the mark with deionized water and filtered with 125µm Whatman filter paper (Amicon, USA) according to Adefemi and Awokunmi, 2010.

#### *Atomic Absorption Spectrophotometer analysis*

All the samples were analyzed for Pb, Cd and Zn by an Atomic Absorption Spectrophotometer (AAS) (Thermo Scientific iCE 3000 Series AA Spectrometer, UK) using an air acetylene flame with digital read out system. The limits of detection and wavelengths in parentheses of Pb, Cd and Zn were 0.04mg/L (217.0nm), 0.006mg/L (228.8nm) and 0.01mg/L (213.9nm) respectively. Then different concentration of standards of each metals were prepared to give a linear relationship, consequently a linear equation was used to quantify the concentration of unknown samples.

#### *Direct Mercury analysis*

A Direct Mercury Analyzer (DMA-80 TRICELL, Milestone, Italy) was used to analyze mercury in prawns, sediments and water. Amount of prawns, sediments and water ranges from 0.1463g to 0.1935g was placed on a nickel or quartz boat (sample holder), then introduced in the quartz furnace and heated up to 200°C (drying temperature) for 1 min and 650°C for 105 seconds to allow Hg reduction and volatilization. Oxygen (99.99%) was used as combustion and carrier gas. Mercury and combustion gases were flushed through the catalyst to retain NO<sub>2</sub> and SO<sub>2</sub> gases. Mercury was selectively trapped in the amalgamator, whereas combustion gases were removed from the detection cell. Mercury was then released from the amalgamator by heating at 850°C for 3 seconds and carried to the detector where the absorbance from the radiation emitted by Hg lamp was measured at 253.7nm for any of the three optic path lengths. The path length used to detect Hg in each sample was automatically selected depending on the Hg concentration and the amount of analyzed sample. The catalyst remained constant at 650°C throughout the analysis procedure and air was used as carrier gas. Limits of detection and quantification were determined using the standard deviation of the residuals from the linear regression plus the linear regression coefficient value of the calibration equation according to Melendez-Perez and Fostier, 2013.

#### *Pesticide residue analysis*

##### *Water*

Five hundred milliliter of water samples were placed in a separating funnel and mixed with 25mL of CH<sub>2</sub>Cl<sub>2</sub>.

The organic layer was separated and the process was repeated three times. Ten grams of anhydrous Na<sub>2</sub>SO<sub>4</sub> was added to dry the extract. Then CH<sub>2</sub>Cl<sub>2</sub> was removed by rotary evaporator (RE-5002, Shanghai, China) until 2mL of the extract was obtained as a final volume according to the method of (Borlongan and Chuan, 2004) with slight modification. The collected extract was further used for pesticide residues analysis using the GC-MS.

##### *Sediment*

Ten grams of sediments was dried in an oven at 120°C for 12h to determine water content. Another 10 g of sediments was mixed with 10g of anhydrous Na<sub>2</sub>SO<sub>4</sub> that was continually added until the sample flowed freely. The mixture was placed in a glass-stoppered bottle and 50mL of CH<sub>2</sub>Cl<sub>2</sub> was added. The sample was sonicated for 20 min and then filtered and evaporated in rotary evaporator (RE-5002, Shanghai, China) until the final volume was 2mL according to (Sumon *et al.*, 2018) with slight modification. The collected extract was further used for pesticide residues analysis using GC-MS.

##### *Prawns*

Ten grams of grounded prawns were taken and the same procedure for extraction in sediments was used, however no anhydrous Na<sub>2</sub>SO<sub>4</sub> was added. The sample was then cleaned up by eluting the extract with 10 g of activated carbon. The solvent was changed from CH<sub>2</sub>Cl<sub>2</sub> to C<sub>6</sub>H<sub>12</sub> and then washed with 10mL of C<sub>6</sub>H<sub>12</sub>. The solvent was evaporated in a rotary evaporator (RE-5002, Shanghai, China) until the final volume of 2mL was reached. The collected extract was further used for pesticide residues analysis using GC-MS (Borlongan and Chuan, 2004).

##### *Pesticide residue analysis*

The final extract of prawns, water and sediment were analyzed for pesticide residues using Gas Chromatography-Mass Spectrometry (GC-2010 Plus Shimadzu, Japan). Pesticide residues were recorded in a GC-MS-2010 Shimadzu instrument operating in Electron Ionization (EI) mode (MS) at 70 ev and Flame Ionization Detector (FID) for GC. A Restek-5MS column (30m x 0.25mm x 0.25µm) was used.



The temperature was maintained at 90°C for 2 min at the beginning then raised to 260°C for 5 min at the rate of 35°C per min. The injection temperature was 250°C with split less injection mode whereas injection sample volume was 1µL. The flow rate of a carrier gas, Helium was 1.21mL/min. The ion source temperature and interface temperature in MS were 230°C and 300°C, respectively. The pesticide residue was identified by scan method, which involved the use of Mass Spectral Library and Search Software (NIST). Quantification of pesticide residues in the samples was done using Peak Integration method where by ion allowance was 20% (Borlongan and Chuan, 2004).

#### Data Analysis

Statistical analysis of data was done in SPSS (IBM SPSS, Version 21, 2012). Data on the sources of aquatic pollution was analyzed and descriptive analyses to compare the responses of respondents. R software version 3.5.0 for Windows (R Development Core Team 2018), specifically the Tukey HSD package was used to quantify the data on heavy metals and pesticide residue. One-way ANOVA tests were performed to compare the dataset at 5% significance level.

## Results

#### Demographic characterization

Demographic characterization of the fishing zones is as shown in Table 1. About 52.5% of the respondents were fishermen, followed by peasants and businessmen. Men were more actively engaged in fishing activity, with age group of 30 – 39 and 40 – 49 years, whereas most of them attained primary education level (Table 1 & 2). Fishing activity was mostly done around the mangrove, rivers, shallow sea, and deep sea, mostly once a day. The prawn catch per day for some prawn fishers was either < 10kg per day (31.1%) or < 50 (29.5%) respectively. However, a 77% decline in prawns yield was also observed. The fishing methods employed in the study areas was the use of gillnets, ring nets and pins with average net size of 3 inches (Table 2).

#### Sources of aquatic pollution

Fertilizers, pesticides and herbicides were reported to be the possible sources of aquatic pollution in the

area. UREA, Booster, and NPK were reported to be used (Table 3). Furthermore, a number of pesticides (*Karate*, and *Ninja*) and herbicides (Roundup, and *Weedall*) were commonly used as agricultural inputs (Table 4). Other than fishing, about 79.5% of agricultural activities were conducted in the area mainly rice farming. Farm sizes were of small and medium scale respectively (Table 2). Ongoing use of agricultural inputs nearby prawns nursery and farming areas may contribute to chemical contaminants which deposits during heavy rainfalls and floods to the estuaries and ultimately to the ocean. Unexpectedly, fishing using chemicals (*Cyberdip* and *Utupa*), and dynamites (18.9%) were practiced in the areas (Table 4).

#### Pesticide residues

*Lambda*-cyhalothrin concentration in sediments and prawns are summarized in Table 5. Only *lambda*-cyhalothrin, an active ingredient in *Karate* and *Ninja* was detected as a pesticide residue. A significant variation ( $p < 0.05$ ) in *lambda*-cyhalothrin levels between zones was observed. *Lambda*-cyhalothrin levels in prawn was detected in order of Kisiju>Kilwa and Kisiju>Kilwa>Bagamoyo in sediments. Besides, *lambda*-cyhalothrin residue was not detected either in prawns from Bagamoyo nor water samples from the three zones.

**Table 1.** Demographic characteristics of respondents in the prawns fishing zones.

Attributes	N (%)	Cumulative percent
Gender		
Man	102 (83.6)	83.6
Woman	20 (16.4)	100
Marital status		
Un-married	10 (8.2)	8.2
Married	96 (78.7)	86.9
Separated	12 (9.8)	96.7
Divorced	2 (1.6)	98.4
Widow	1 (0.8)	99.2
Un-marriage	1 (0.8)	100
Age		
10-19	1 (0.8)	0.8
20-29	21 (17.2)	18.2
30-39	28 (23.0)	41.3
40-49	29 (23.8)	65.3
50-59	24 (19.7)	85.1
60-69	12 (9.8)	95.0
70-79	6 (4.9)	100.0
Education level		

Attributes	N (%)	Cumulative percent
Uneducated	17 (13.9)	13.9
Primary	90 (73.8)	87.7
Secondary	15 (12.3)	100
Occupation		
Fishermen	64 (52.5)	52.5
Peasants	44 (36.1)	88.5
Businessman	12 (9.8)	98.4
Others	2 (1.6)	100.0
Total	122 (100)	100.0

**Table 2.** Socioeconomic activities around the fishing zones.

Attribute	N (%)	Commulative (%)
Any agricultural activities around the area		
Yes	97 (79.5)	79.5
No	21 (17.2)	96.7
I don't know	4 (3.3)	100.0
Sizes of agricultural activities		
Small	37 (30.3)	30.3
Medium	46 (37.7)	68.0
Large	13 (10.7)	78.7
Not applicable	26 (21.3)	100.0
Types of crop harvested		
Rice	79 (64.8)	64.8
Maize	8 (6.6)	71.3
Cassava	5 (4.1)	75.4
Tomatoes	2 (1.6)	77.0
Cashew nuts	1 (0.8)	77.9
Vegetables	1 (0.8)	78.7
Not applicable	26 (21.3)	100.0
Any Industrial activities		
Yes	9 (7.4)	7.4
No	104 (85.2)	92.6
I don't know	9 (7.4)	100.0
Fishing location		
Rivers	22 (18.0)	18.0
Estuaries	2 (1.6)	19.7
Mangrove	42 (34.4)	54.1
Deep sea	12 (9.8)	63.9
Shallow sea	19 (15.6)	79.5
<i>Mwamba</i>	9 (7.4)	92.5
All	16 (13.1)	100.0
Prawns catch per day (kg)		
< 10	38 (31.1)	31.1
< 30	29 (23.8)	54.9
< 50	39 (29.5)	84.4
> 50	19 (15.6)	100.0
Types of fishing gears		
Gillnet	96(78.7)	78.7
Ring net	18(14.8)	93.4
Pin	8(6.6)	100.0
Size of the nets (Inches)		
1	1(11.5)	11.5
2	67(54.9)	66.4
3	6(4.9)	71.3
I don't know	35(28.7)	100
Frequency of fishing prawns (day)		
Once	84 (68.9)	68.9
Twice	32 (26.2)	95.1
Above twice	6 (4.9)	100.0

Attribute	N (%)	Commulative (%)
Status of prawns yield		
Decline	94 (77.0)	77.0
Raised	17 (13.9)	91.0
Same	11 (9.0)	100.0
Total	122 (100)	100.0

**Table 3.** Agricultural inputs used in the study area.

Attribute	N (%)	Cumulative (%)
Fertilizers		
UREA	38 (31.1)	31.1
NPK	10 (8.2)	39.3
Booster	26 (21.3)	60.7
Nil	48 (39.3)	100.0
Herbicides		
Roundup	41 (33.6)	33.6
Commander	8 (6.6)	40.2
<i>Weedall</i>	15 (12.3)	52.5
Herbicide	10 (8.2)	60.7
Nil	48 (39.3)	100.0
Pesticides		
<i>Karate</i>	38 (31.1)	31.1
<i>Ninja</i>	20 (16.4)	47.5
<i>Rungu</i>	3 (2.5)	50.1
<i>Cyberdip</i>	7 (5.7)	55.7
Commander	5 (4.1)	59.8
Nil	49 (40.2)	100.0
Total	122 (100)	100.0

**Table 4.** Chemicals used in fishing.

Attributes	N (%)	Cumulative (%)
Chemicals		
<i>Utupa</i>	8 (6.6)	6.6
<i>Cyberdip</i>	9 (7.4)	14.0
Bottles of oxygen gas	2 (1.6)	15.6
Théoden	1 (0.8)	16.4
Not applicable	96 (78.7)	95.1
I don't know	6 (4.9)	100
Dynamite fishing		
Yes	23 (18.9)	18.9
No	99 (81.1)	100.0
Total	122 (100)	100.0

**Table 5.** *Lambda*-cyhalothrin concentration in the fishing zones.

	Lambda- Concentration (Mean±SD;mg/kg)			P-values
	Bagamoyo	Kisiju	Kilwa	
Prawns	nd	2.26±0.51	0.28±0.40	0.017
Sediments	0.65±0.82	16.49±2.36	12.21±3.24	0.014
Water	nd	nd	nd	-

Nd: not detected; Error bars represent Mean±SD, (n=3), Means between the groups are significant at p < 0.05.



### Heavy metals

The concentration of Pb, Zn, Cd and Hg in prawns, water and sediments varied widely in the three zones (Table 6). Heavy metal concentration in prawns in all zones was in a range between 24.29 – 24.91, 107.16 – 166.06, 0.18 – 1.17, and 0.05 – 0.19mg/kg for Pb, Zn, Cd and Hg respectively. In sediments, the range for Pb, Zn, Cd and Hg was between 28.13 – 35.07, 133.44 – 153.49, 1.50 – 1.80 and 0.04 – 0.05mg/kg respectively. For water, the range was between 0.05 – 0.74, 0.42 – 0.53, 0.02 – 0.09 and 0.03 – 0.19mg/L for Pb, Zn, Cd and Hg respectively (Table 6). Generally, heavy metal concentration in prawns and sediments were detected in order of Zn>Pb>Cd>Hg, and Pb>Zn>Cd>Hg in water respectively. Furthermore, all the samples exhibited Zn highest concentration in the order of Kilwa>Kisiju>Bagamoyo. Mercury exhibited lowest concentration in all samples. Besides, significant low levels of Pb, Zn, Cd and Hg were observed in all water samples.

**Table 6.** Heavy metals concentration in the study zones.

Zones	Concentration (Mean±SD)			
	Pb	Zn	Cd	Hg
<b>Bagamoyo</b>				
Prawns	24.29±1.73	107.16±2.65	1.04±0.07	0.07±0.03
Sediments	31.66±3.43	152.50±20.80	1.80±0.13	0.05±0.01
Water	0.74±0.46	0.42±0.11	0.07±0.03	0.03±0.01
<b>Kilwa</b>				
Prawns	24.91±1.43	166.06±7.14	1.17±0.03	0.05±0.01
Sediments	35.07±10.54	133.44±35.98	1.50±0.24	0.04±0.01
Water	0.05±0.07	0.45±0.22	0.02±0.00	0.03±0.01
<b>Kisiju</b>				
Prawns	24.75±1.59	153.89±0.61	0.18±0.05	0.19±0.24
Sediments	28.13±3.44	153.49±9.35	1.63±0.08	0.05±0.01
Water	0.59±0.61	0.53±0.20	0.09±0.11	0.03±0.01

Note: Heavy metal mean concentrations for prawns and sediments (mg/kg), and water (mg/L).

## Discussion

### Demographic characterization

Fishing, particularly prawn fishing was among the economic activities of socioeconomic importance to the coastal communities. Artisanal fishing is a pillar to most coastal livelihood households thus calling for proper management of the fisheries resource (Hamidu, 2014). The marine fishery is very important for coastal population as their major sources of daily income and food for their families (Mkama *et al.*, 2013). From the current study, majority of men (30 – 49 years) engaged in fishing activities were fishermen

by occupation. Similarly, (Okayi *et al.*, 2013) observed more men (21 – 40 years) involvement in fishing activities with higher education level (secondary and tertiary) as opposed to most with low (primary) education level in this study. The age group and education levels of the respondents in the study zones suggest that majority were engaged in fishing activities immediately after completing their primary education.

Level of education has been shown to have an impact on illegal fishing methods (dynamite and chemicals uses) on ecosystem and human health (Katikiro, 2014). Besides, the current study did not assess the impact of level of education on fishing practices.

### Sources of aquatic pollution

Agricultural activity, largely medium and small scales is heavily dependable by the local communities for their livelihood (Table 2). A routine usage of fertilizers, pesticides and herbicides as farming inputs in addition to industrial activities may have resulted into high concentration of marine pollutants, of which contaminates fish and marine environment. Likewise, according to Odada *et al.*, 2004, industrial activities, disposal or incineration of waste, certain agricultural and the use of fertilizers, anti-parasites and feeds containing metals as well as natural causes have been reported to be sources of aquatic pollution.

Heavy rains and floods carry agricultural waste deposits and residues of pesticide into the oceanic waters (Nzung'a, 2018). A decline in prawns production around the delta has been linked to agriculture activities that disturb spawning grounds of prawns (Mmochi and Francis, 2003). Similarly, (Slade and Kalangahe, 2015) reported dynamite fishing along the coastline of Tanzania to be detrimental not only to the environment but also to consumers. Dynamite fishing has been reported to destroy coral reefs that destruct spawning grounds of fish (Wells, 2009). The decreases in abundance and biomass of fisheries resources, and prawn in particular is in line with decline of target species in various areas throughout the tropics (Hamidu, 2014). Likewise, a decline (77%) in prawn's yield was also reported in this study (Table 2).

### *Pesticide residue*

*Lambda-cyhalothrin* is a pyrethroid insecticide (He *et al.*, 2008) which can bioaccumulate and not able to dissociate in the environment under suitable conditions. *Lambda-cyhalothrin* is an active ingredient of *Karate* and *Ninja* used in pests' control. With intensive rice farming (64.8%, Table 2) around the fishing zones (Rufiji Delta), *Karate* and *Ninja* (*lambda-cyhalothrin*) are applied to control pests. Therefore, *lambda-cyhalothrin* contamination in prawns and sediments is suggested to originate from pesticide application from rice farms. According to (Senoro *et al.*, 2016), pesticide application is the most common method of controlling pests and weeds in rice and vegetable farming. Pesticides can migrate from treated fields to air, nearby land areas and water bodies. Also, pesticide residues may drain (Owa, 2014) through runoff, and sink down into sediments caused by upwelling (Alegria and Shaw, 1999). *Lambda-cyhalothrin* has been reported to be highly toxic to freshwater shrimp and *Tilapia* fingerling (Senoro *et al.*, 2016) with a LC<sub>50</sub> in shrimp 0.28 microgram/L (Wang *et al.*, 2007). Therefore, detection of *lambda-cyhalothrin* residue in prawns may indicate risks to human health as well as aquatic pollution.

### *Heavy metals*

Zinc in prawn and water samples in this study exceeded the FAO and WHO acceptable maximum limit of 100mg/kg for prawns according to Choi (2011), and 0.0766mg/L for water but it was below the acceptable limit of 7500mg/kg in sediments (Duruibe *et al.*, 2007). The exceeded limits for Zn in prawn and water samples is up to 1.7 and 5 – 7.0 folds respectively. The concentration of the Zn and Pb also exceed that of Rumisha *et al.*, 2016. The acceptable limits of heavy metals in sediments are 85mg/kg for Cd, 1mg/kg for Hg, and 420mg/kg for Pb respectively (Choi, 2011; CAC, 2016). Therefore, from this study concentration of Cd, Hg and Zn in sediments was below the acceptable limits.

The limit for Pb in prawns is 0.30mg/kg CAC (2016) and 0.5mg/kg FAO (1983) and for water is 0.0058mg/L (Duruibe *et al.*, 2007). Therefore, all the prawn samples contained 50-83 folds higher

concentration of Pb and 8-128 folds in water respectively. Higher accumulation of Pb in marine prawns and water was also reported in other studies (Abdennour *et al.*, 2000; Adefemi and Awokunmi, 2010; Förstner and Wittmann, 2012). In contrast, Pb concentration in sediments was found to be below acceptable limits of 420mg/kg.

For instance, Cd concentrations in all samples of prawns, sediments and water analyzed were above the European Union (EU, 2001) limit of 0.05mg/kg; United States Environmental Protection Agency (USEPA, 2000 and EUROPA (2004) 0.10mg/kg respectively. Ali *et al.*, 2016 reported that, anthropogenic activities can effortlessly generate heavy metals in sediments and water that pollute the aquatic environment. Conversely, Cd concentration in water was below the limit set by the Malaysian Food Regulation (1985) i.e., 1.0mg/kg.

According to Hong Kong Environmental Protection Department (HKEPD, 1987) the recommended limit is 2.00mg/kg, United States Food and Drug Administration (USFDA, 1993) is 3.70mg/kg of which the levels reported in this study were within the acceptable limits. Additionally, Hg detected was also below the acceptable limit in sediments and water; and 0.38 folds higher in prawns from Kisiju.

The anthropogenic activities around the coastal area and discharge of oil from cargo ships pollutes water, especially at Bagamoyo as a result of high concentrations of heavy metals (Khalifa *et al.*, 2010). Heavy metals cannot be destroyed or degraded and can enter in human bodies through food chain including prawns, water and air inhalation. Heavy metals have ability to accumulate in marine environmental (Lundy *et al.*, 2017) and marine organisms such as prawns. If the concentration exceeds the acceptable levels, it becomes toxic to prawns and may cause health problems to prawns (Mmochi and Francis, 2003) as well as posing health risks to human. Heavy metals and *lambda-cyhalothrin* contaminants indicate a potential safety risk of prawn consumers and marine environment.

## Conclusion

Heavy metal concentrations (Pb, Zn, Cd and Hg) and *lambda*-cyhalothrin were detected in prawns in varying levels in all study zones. Pesticides, herbicides and fertilizers used in agricultural activities were reported as possible sources of pollutants. Elevated human activities along the Indian coastline of Tanzania might have attributed to high concentration of Pb and Zn and *lambda*-cyhalothrin. To safeguard the ecosystem and human communities, immediate and frequent monitoring of anthropogenic activities around the nursery areas for prawns is necessary. In addition, a comprehensive risk assessment is needed to shade more light on the status quo of heavy metals and pesticide residues to the human population in the areas. Also, the findings from this study can serve as a baseline information for future monitoring and risk assessment studies.

## Author Contributions

GP conceived the idea, performed the experiment, analyzed the data and drafted the manuscript; AM and EM involved in the study design, and manuscript revision.

## Conflicts of Interest

The authors declares that there is no conflict of interest.

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