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Assessment of Health Risks Associated with Concentrations of Heavy Metals in Fish from the Coast of Tanzania

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***Abstract*—Heavy metals are serious threat because of their toxicity, long persistence, bioaccumulation and biomagnification in the food chain. This research deals with human health risk assessment of metal contamination through the consumption of commonly consumed fish from Dar es Salaam City. The fish sample of interest were Rastrellieger kanagurta, Lutjanus fulvus and Fenneropenaeus indicus. The aim is to determine the concentration of Pb, As, Cd, Fe and Cu contaminant in these fish samples. By using AAS the maximum concentration of Pb, As, Cd, Fe and Cu was 0.14, 1.09, 0.16, 60.29 and 12.11 mg/kg respectively. The estimated daily intake (EDI) of heavy metals with the respective type of fish can be arranged as Fe > Cu > As > Cd > Pb in which values are higher than Provisional Tolerable Weekly Intake (PTWI) for metals. Therefore the consumption of fish samples is questionable. Target Hazard Quotient (THQ) was used in the health risk assessment to determine carcinogenicity of the sample. The result shows that the concentration and THQ of As in all fish samples ranges from 1.173 – 2.325 which is > 1, hence signified that a daily exposure at this level is likely to cause effects during a person lifetime. It is well known that fishes can accumulate variety of toxic chemicals including persistent organic contaminants such as dioxins and chlorinated pesticides; hence similar study has to be conducted for such compounds at different sampling sites including river and personal fish ponds.**

Keywords—Target Hazard Quotient, Arsenic, Health Risk, Concentration, Heavy Metals

I. INTRODUCTION

In the recent years, World consumption of fish has increased simultaneously with the growing concern of their nutritional and therapeutic benefits. It is documented that fish is a good source of protein, essential minerals, vitamins and unsaturated fatty acids world consumption of fish and its products has increased mainly due to their health benefits such as preventing cardiovascular and other disease [1, 2]. Despite their recognized benefits, fish and seafood may represent a risk for human health since they can accumulate contaminants from aquatic environment and magnify them up the food chain [3]. In many studies, fish species have been employed as bioindicators of environmental contamination [2- 4]. However, factors such as time, catching place, habitat, gender and age may modify chemical constituents, and pollutant burden can diverge among different species and even among individuals of the same species [1].

For many years, scholars [3, 5] have tried to write on the effect of pollutants on aquatic flora and fauna because it came to notice that water bodies near urban areas (cities and towns) are highly polluted. This is the result of both garbage dumped by individuals and dangerous chemicals legally or illegally dumped by manufacturing industries, health centers, schools and market places. The issue of concern to researchers is the level of heavy metals detected in these marine animals [5]. These heavy metals pose threats to public water supplies and can also cause health hazard to human consumption of fish resources [6]. Heavy metals in aqueous form are easily taken up by aquatic organisms where they are strongly bound with sulfhydryl groups of proteins and accumulate in their tissues. The accumulation of heavy metals in the tissues of organisms can result in chronic illness and cause potential damage to the population.

Metals enter rivers and lakes from a variety of sources, such as rocks and soils that are directly exposed to surface waters, fallout of atmospheric particulate matter, and from man's activities, including the discharge of treated and untreated wastes into water bodies [7]. Excess amounts of these metals entering into the aquatic ecosystem may pollute the environment and also affect the food chain and ultimately pose serious human health risks to those who depend directly or indirectly on the water body for the supply of fish and water [8].

Lead toxicity can be expressed as it affects the hematopoietic, renal, reproductive and central nervous system, mainly through increased oxidative stress. Compared to other organ, the nervous system appears to be the most sensitive and chief target for lead induced toxicity [9]. Both the central nervous system and the peripheral nervous system become affected on lead exposure. The effects on the peripheral nervous system are more pronounced in adults while the central nervous system is more prominently affected in children.

Copper is an essential trace metal and micronutrient for cellular metabolism in living organisms on account of being a key constituent of metabolic enzymes [10]. However it can be extremely toxic to intracellular mechanisms in aquatic animals at high concentrations which exceed normal levels [11]. It is an abundant element which occurs as a natural mineral with a wide spread use. Copper pollution is through extensive use of fungicides, algaecides, molluscicides, insecticides and discharge of wastes [12]. Copper sulfate ($CuSO_4$) is often used as an algaecide in commercial and recreational fish ponds to control the growth of phytoplankton and filamentous algae and to control certain fish disease [13]. Fish can accumulate copper via diet or ambient exposure.

Excess concentration of copper is associated with liver cirrhosis in children [14]. Also the Wilson disease which causes the body to retain copper as it is not excreted by the liver into the bile. This disease, if untreated, can lead to brain and liver damage [15]. Also elevated free copper levels exist in Alzheimer's disease, which has been hypothesized to be linked to inorganic copper consumption [14].

Cadmium which is known carcinogenic compound makes the kidneys to lose their function to remove acids from the blood in proximal renal tubular dysfunction. The kidney damage inflicted by cadmium poisoning is irreversible [15]. Also, cadmium exposure is also associated with the development of kidney stones [15].

Arsenic compound is one of the cancer agent [16], which when exposure occurs over a brief period of time symptoms may include vomiting, abdominal pain, encephalopathy and diarrhea Long term exposure can result in thickening of the skin, darker skin, abdominal pain, diarrhea, heart disease and cancer. Arsenic exposure plays a key role in the pathogenesis of vascular endothelial dysfunction as it inactivates endothelial nitric oxide synthase, leading to reduction in the generation and bioavailability of nitric oxide.

Iron is one of very essential element in the body and there is no known toxicological effect of iron. There is no mechanism exists for excreting iron, toxicity depends on the amount of iron already in the body. Consequently, some animals develop clinical signs of toxicosis even when they receive doses that cause no problems in other animals. Iron is most toxic when given intravenously.

However, intake of heavy metal contaminated fish may pose a risk to the human health. Prolonged human consumption of unsafe concentrations of heavy metals in food stuffs may lead to the disruption of many biological and biochemical processes in the human body [17]. Intake of fish is an important path of heavy metal toxicity to human being. Dietary intake of heavy metals through contaminated fish may lead to various chronic diseases. Regular monitoring of these metals in fish product and in other food materials is essential for preventing excessive buildup of the metals in the food chain.

According to EPA [18], a human health risk assessment is the determination of likelihood that a given exposure or series of exposures may have damaged or will damage the health of an individual. This assessment is the process to estimate the nature and probability of adverse health effects in humans who may be exposed to chemicals in contaminated environmental media, now or in the future.

Human health risk assessment is a process that is accepted by most of international health agencies for evaluating the potential for chemical, biological and physical agents to cause adverse health effects in people. Although it is desirable to minimize exposures to some environmental chemicals, exposures to chemicals and physical agents cannot be avoided in many circumstances. Potentially harmful chemicals and physical agents can exist naturally and there were exposures prior to modern civilization. This is also true for most of the heavy metals.

As we are dealing with fish samples, there are issues to consider for the human health risk assessment. The USEPA standard uses the hazard quotient which assumes that there is a level of exposure below which it is unlikely to experience an adverse non-carcinogenic health effect [19]. The hazard Quotient values greater than 1 are predicted from consumption of certain fish under both the present conditions and the predicted post-impoundment conditions. Under post-impoundment conditions, Hazard Quotient values increase since the heavy metal concentrations in various fish are estimated to increase.

The aim of this study was to find the concentrations of heavy metals in vegetables in Dar es Salaam City in Tanzania. We have estimated the contribution of these heavy metals to the daily intake of normal human being. Also we have estimated the potential health risks associated with heavy metals via consumption of

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fish using the estimated daily intake (EDI) and health risk index (HRI) from single and combined heavy metals.

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II. MATERIAL AND METHODS

A. Study Area and Sampling Locations

Dar es Salaam is located in the eastern part of the Tanzanian mainland at 6º51'S latitude and 39º18'E longitude. With an area of 1,350 square kilometers (km^2), it occupies 0.19 percent of the Tanzanian mainland, stretching about 100 km between the Mpiji River to the north and beyond the Mzinga River in the south. The Indian Ocean borders it to the East. The beach and shoreline comprise sand dunes and tidal swamps. Coastal plains composed of limestone extend 10 km to the west of the city, 2-8 km to the north, and 5-8 km to the south. Inland, alluvial plains comprise a series of steep-sided U-shaped valleys. The upland plateau comprises the dissected Pugu Hills, 100-200 m in altitude. Dominated by limestones, sandy clays, coarse sands and mixed alluvial deposits, the soils of the Dar es Salaam region are not particularly fertile. The City is divided into three ecological zones, namely the upland zone comprising hilly areas to the west and north of the City, the middle plateau, and the lowlands, which include Msimbazi Valley, Jangwani, Mtoni, Africana and Ununio areas.

Dar es Salaam is the commercial city of the country; it is one of the fastest growing cities in Africa. It has a population of 4,364,541[20], with a population increase of 5.6 percent per year from 2002 to 2012, the city is the third fastest growing in Africa (ninth fastest in the world), after Bamako and Lagos [21]. The average income earner in Dar es Salaam is responsible for four people, this is a significant burden given a low level of earnings. Most workers are self-employed rather than wage earners. The majority of the poor is proprietors of small businesses and account for 20 to 40% depending on the area of the city.

Figure 1: Map of Dar es Salaam City Showing the Administrative Districts

B. Fish Sampling

Fish sample was collected from Kivukoni fish market at Dar es Salaam city in order to assure regularity in fishing methods. The fish was transported to the laboratory on the same day in the pre-cleaned polyethylene bags. All samples were frozen and stored at -18ºC immediately upon returning from the field. The collected samples were washed with distilled water to remove any contaminated particles. Muscle tissue of fish (dorsal muscle) was used in this study because it is the major target tissue for metal storage and is the most edible part of the fish. Fish tissues were cut and oven dried at 110°C to a constant weight [22]. A wet digestion method was used based on the Analytical Methods for Atomic Absorption Spectrometry. Prior to use, all glassware was previously soaked in diluted nitric acid for 24 h and then rinsed with distilled deionised water. The 2 g dry weight sample was put into a 50 ml beaker with 5 ml of HNO_3 and 5 ml of H_2SO_4. When the fish tissue stopped reacting with HNO_3 and H_2SO_4, the beaker was then placed on a hot plate and heated at 60°C for 30 min. After allowing the beaker to cool, 10 ml of HNO_3 was added and returned to the hot plate to be heated slowly to 120°C. The temperature was increased to 150°C, and the beaker was removed from the hot plate when the samples turned black. The sample was then allowed to cool before adding H_2O_2 until the sample was clear. The content of the beaker was transferred into a 50 ml volumetric flask and diluted to the mark with deionized water. All the steps were performed in the fume hood. The above procedures in this section followed the guidelines from the Analytical Methods for Atomic Absorption Spectroscopy [23].

III. Results and Discussion

The mean values of iron, manganese, copper, arsenic, lead, cadmium and iron concentrations in the studied three common fish species are given in Table 1.

Table 1: Levels of Selected Heavy Metals in Fish Samples (mg/kg)

Common Name	Scientific Name	Cd	As	Pb	Cu	Fe
Mackerel	Rastrellieger kanagurta	0.06	0.70	0.03	12.11	60.29
Snappers	Lutjanus fulvus	0.16	0.55	0.14	9.23	15.77
Prawns	Fenneropenaeus indicus	0.01	1.09	0.06	5.97	41.02
WHO/FAO		0.2	0.26	0.3	30.0	43

The concentrations of heavy elements in the selected studied species are varied quietly such as, Fe (15.77–60.29), Cu (5.97–12.11), Cd (0.01 – 0.16), Pb (0.03-0.14) and As (0.55 – 1.09) mg/kg. Fe was the most accumulated in all the fish samples with *Rastrellieger* (60.29 mg/kg) having the highest concentration and *Lutjanus fulvus* (15.77 mg/kg) the lowest. The result showed *Fenneropenaeus indicus* has a values very close the WHO recommended 40mg/kg in food. The concentration of iron observed in this study is comparable to levels reported by other author (Makimilua, and Afu, 2013) reported Fe mean concentration in fish samples 49 mg/kg where catfish has a concentration of 44.0 mg/kg and tilapia 53 mg/kg. Iron is an essential component of haemoglobin which is responsible for oxygen transportation in the body. Severe iron deficiency in human causes anaemia.

The metal cadmium known to be a highly toxic non–essential heavy metal and it does not have a role in biological process in living organisms. Thus, even at its low concentration, cadmium could be harmful to living organisms [16]. The concentration detected 0.16 mg/kg maximum concentration of Cadmium detected in the muscle of *Lutjanus fulvus* and the 0.06 mg/kg concentration in the muscle tissues of *Rastrellieger* as shown in Table 1 were exhibitive of high potential health effects to the majority of the patronizing fish consumer population at the study area. The grand mean concentration of Cadmium in both common fish species is comparable to the WHO/FAO maximum permissible limit of 0.2 mg/kg for food samples. However, the concentration is very much comparable with the recent study with the concentration from 0.17 – 0.32 mg/kg [6]. Though the levels seem to be lower than the acceptable levels by WHO/FAO, due to bioaccumulation of cadmium in fish could not be tolerated and the consumers might raise alarm if they were actually aware of its potential health risks.

The Pb concentration in the samples ranged from (0.03 - 0.14) mg/kg. It is documented that excessive accumulation of heavy metals like lead in food can result in serious systemic health problems in humans [24]. Therefore, the FAO, WHO and other regulatory bodies of various countries have established the maximum permitted concentrations of heavy metals in foodstuffs The maximum tolerable limit (MTL) of Pb in fish meat by the EU is 0.3 mg/kg [25]. The concentrations of Pb in all the fish samples from the samples are below the maximum permitted limit. Other researchers have also reported high levels of Pb in fish muscles. For example, reported Pb levels of 3.125 mg/kg in fish [26]..

The concentrations of Cu in the fish muscle were in the range 5.97- 12.11 mg/kg. The standard established limits for Cu in fish as 30.0 mg/kg for human health risk concerns [27]. The concentrations of Cu in these samples were far below this value therefore regular consumption of fish with such low amounts of Cu could not lead to any serious health risk so far as Cu is concerned.

Arsenic levels in the sample ranges from 0.55 – 1.09 mg/kg with prawns *(Fenneropenaeus indicus)* detected the highest and the lowest detected in snappers (*Lutjanus fulvus*). Arsenic is a carcinogen element that can traverse placental barriers and produce fetal death and malformations in many species of mammals. Although it is carcinogenic in humans, evidence for arsenic-induced carcinogenicity in other mammals is scarce. Paradoxically, evidence is accumulating that arsenic is nutritionally essential or beneficial. Arsenic deficiency effects, such as poor growth, reduced survival, and inhibited reproduction, have been recorded in mammals fed diets containing <0.05 mg As/kg, but not in those fed diets with 0.35 mg As/kg. The mean concentration of arsenic common fish species is comparable lower than the maximum permissible limit of 0.26 mg/kg for food samples [28].

Health Risk Assessment on Consumption of Fish Sampled

Previous studies [4, 6, 31], have shown that fish are able to accumulate and retain heavy metals from their environment and it has been shown that accumulation of metals in tissues of fish is dependent upon exposure concentration and duration, as well as other factors such as salinity, temperature, hardness and metabolism of the animals. In order to determine the health risk associated with consumption of fish, we need to estimate the daily intake (EDI) of these metals as was expressed based on USEPA guidelines and can be expressed in equation (i) [31].

$$EDI = \frac{E_F \times E_D \times F_{IR} \times C_f \times C_m}{W_{AB} \times T_A} \quad \ldots\ldots\ldots\ldots\text{(i)}$$

Then further we can determine the THQ which is the ratio between the exposure and the reference dose (a reference dose or RfD), is used to express the risk of non-carcinogenic effects. Ratio of less than 1 signifies non-obvious risk. It is important to note that an exposed population of concern will experience health risk if the dose is equal to or greater than the RfD [31]. The estimation of THQ of each heavy metal in this exposure pathway by using an equation (ii).

$$THQ = \frac{E_{Fr} \times E_{Dtot} \times F_{IR} \times C_m}{R_{fDO} \times BW_a \times ATn} \times 10^{-3} \quad \ldots\ldots\ldots\ldots\ldots\text{(ii)}$$

Where :

E_F = Exposure frequency (365 days/year)

E_{Dtot} = Exposure duration (average life time) 62 years for Tanzanian population

F_{IR} = Fresh food ingestion rate (48 g/person/day) (Ali and Hau, 2001)

C_m = Heavy metal concentration in fish sample

RfDo = Oral Reference dose (mg/kg/day)

BW_a = The average body weight (Considered to be 75kg)

ATn = Average exposure time for non-carcinogens (Equivalent to $E_P \times E_D$)

The RfD is an estimation of the daily exposure to which the human population is likely to be without any appreciable risk of deleterious effects during a lifetime. The values of R_{fD} for heavy metals were taken from Integrated Risk Information System and Department of Environment, Food and Rural Affairs [28].

1) Estimated Daily Intake

It should be taken note that most of heavy metals are accumulated in human tissues and hence they are harmful to human health. The Joint FAO/WHO Expert Committee on Food Additives has set limit for heavy metal intake based on body weight (bw) [28, 32]. For an average adult (70 kg bw), Provisional Tolerable Weekly Intake (PTWI) for metals like Pb and Cd are 0.025 and 0.007 mg/kg bw, respectively, while the Provisional Tolerable Monthly Intake (PTMI) for Fe and Cu is 0.8 and 0.5 mg/kg bw and the Benchmark Dose Lower Limit ($BMDL_{0.5}$) for As is 0.003 mg/kg bw [32].

Table 2: Estimated Daily Intake of Metals (EDI) and Target Hazard Quotients

Type of fish		Cd	As	Pb	Cu	Fe
RfD (USEPA (2000))		1×10^{-2}	3×10^{-4}	3.3×10^{-3}	3×10^{-1}	7×10^{-1}
Rastrellieger kanagurta	EDI	0.008	0.246	0.004	1.612	5.461
	THQ	0.038	1.493	0.048	0.194	0.055
Lutjanus fulvus	EDI	0.0213	0.194	0.019	1.229	2.099
	THQ	0.102	1.173	0.224	0.148	0.014
Fenneropenaeus indicus	EDI	0.001	0.384	0.008	0.795	5.461
	THQ	0.006	2.325	0.096	0.096	0.038

The estimated daily intake (EDI) of heavy metals with the respective type of fish is listed in Table 2. The rating can be organized as Fe > Cu > As > Cd > Pb. All type of fish, consumption questionable in terms of these metal levels because EDI is higher than the recommended intake values.

Target Hazard Quotients (THQ)

The HQ is a highly conservative and relative index. When HQ is < 1, there is no obvious risk from the substance over a lifetime of exposure, while HQ is > 1, the toxicant may produce an adverse effect. The Rfd of analysed metals is indicated on Table 2. The Body weight (Bo) is an average body weight of African male which is taken as 57 kg [33]. According to another study [6], an average mans needs a 48 g of fish per day. The target hazard quotients (THQs) of studied metals through consumption of fish for residents were derived and listed in Table 2.

Highest THQ value belongs to As observed to be more in *Fenneropenaeus indicus* followed by *Rastrellieger kanagurta* and lastly in *Lutjanus fulvus.* The values in As is higher compared to other metals it was higher than values for Cu, Fe, Pb and Cd.

From the result, the THQ values of all heavy metals in all fish samples were all below one (1) (except for As in all fish samples). The results indicate that there is no THQ value > 1, indicating that humans would not experience any significant health risk if they only consume metals from this species of fish. However, the high THQ for As observed in fish consumed had greatest potential to pose health risk to the consumer were probably exposed to some potential health risk through the intake of As via consuming all type of fish analyzed. Even though there was no apparent risk when each metal was analyzed individually, the potential risk could be multiplied when considering all heavy metals.

It has been reported that exposure to two or more pollutant may result in additive and/or interactive effects [31]. In this study the total THQ is treated as the arithemetic sum of the individual metal THQ values:

$$\text{Total THQ} = \text{THQ}_{(\text{Metal 1})} + \text{THQ}_{(\text{Metal 2})} + \text{THQ}_{(\text{Metal 3})} + \ldots\ldots\ldots$$

This research found that As was a major risk contributor for general population in that sample, accounted for about 91% of the total THQ. The intake of total arsenic in the human diet is usually dominated by organic arsenic derived from seafood. The tolerable weekly As intake limit recommended by the FAO/WHO for adults is PTWI 2.1 x10^{-3} mg/kg bw per day body weights [32]. Taking into account the average body weight of 55.9 kg for Tanzanian adults in these areas, the tolerable daily intake of As will be 0.12g.

The reported values in this study is higher than those detected in another study [30], where a total health hazard, based on hazard quotients (HQ) for fish eating human population from studied metals was ranged between 0.027- 0.13, which is much lower than safe limit of one (<1) suggesting negligible health risk.

IV. Conclusion

The selected fish individuals from coastal area of Tanzania analyzed reveal heavy metals concentrations of As is higher than permissible guidelines while other metals are lower than the guideline values. Since health risk for humans is given by the present consumption rate, hence data we obtained on ADD and EWI was lower than daily reference dose (RfD) and higher than provisional tolerable weekly intake (PTWI) of metals for the surrounding human population within the limit of our study area. Furthermore, the estimated risk in terms of total hazard quotient (THQ) from the metal concentrations does not have risk to human health. However, it is just a selective fish investigation; metal contamination levels should be carefully monitored on a regular basis in more fish species, to detect the change in their accumulation patterns. It is well known that fishes can accumulate variety of toxic chemicals including persistent organic contaminants such as dioxins and chlorinated pesticides; hence similar study has to be conducted for such compounds. It is further recommended to have more metals and several variety of fish samples so as to be able to conclude on human health status on consumption of fish in coastal area.

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