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Original Research Article

BIOACCUMULATION OF HEAVY METALS IN MACKEREL AND RED HYBRID TILAPIA COLLECTED FROM KIJAL AND PAKA, TERENGGANU, MALAYSIA

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A b s t r a c t

The major goal of this study is to determine the bioaccumulation of heavy metals in the tissues and bones of Red Hybrid Tilapia *(Oreochromis niloticus)* and Mackerel *(Rastrelliger kanagurta)* taken from two distinct sites in Terengganu state, Malaysia, namely Kijal and Paka. Zinc (Zn), copper (Cu), nickel (Ni), and lead (Pb)were the metals examined. Soil analysis also was carried out for the same elements. The tissues and bones of the fishes were separated before further analysis was made. Drying process technique was used to determine the moisture content for all samples at 80 °C for 24 hours. Water content ranged between 70 & 77%. Triplicate samples were evaluated by using Atomic Absorption Spectrophotometer (AAS) to find out heavy metals concentration. Oven and Teflon beakers were used to fully digest all solid samples at 120 ºC for 3 hours. Highest readings of Zn, Ni and Pb were found in the fish bone for the samples collected from both mentioned locations. However, different pattern was noticed for Cu where higher values were found in the tissues for the same samples. Values of Pb in the tissues were higher than the acceptable value in edible fish by Food and Agriculture Organization (FAO); 1 ppm and Malaysian Food Act (MFR) & World Health Organization (WHO); 2 ppm, whereas values of Cu, Zn were in the

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acceptable range set by the same organizations. No values recommended for Ni by MFR. In general heavy metals in the bones and tissues from both locations can be sorted in descending order as follows; Pb>Zn>Cu>Ni. Soil samples collected from both locations showed different pattern where high concentration was found for Zn followed by Pb then Cu and Ni. Moreover analysis for soil samples collected from Paka River showed higher content for all elements compared to Kijal. More investigation is needed to clarify whether this contamination is solely from industry or due to geology of the area.

Keywords: heavy metals, bio-monitoring, marine eco-system, rastrelliger kanagurta, oreochromis niloticus

1. INTRODUCTION

Heavy metals contamination due to industrial rapid growth as well as agricultural and mining activities has led to significant impact socially and economically. Contamination of estuarine and coastal marine ecosystem by heavy metals represents the major concern for the relevant agencies since heavy metals are the most significant anthropogenic contaminants in the estuarine and coastal marine ecosystems. Because of their toxicity and accumulative characteristics in aquatic species, exacerbating conditions can partially or completely damage both aquatic species diversity and ecosystems. Furthermore, if authorized organization do not implement reliable and immediate countermeasures to prevent heavy metal accumulation, local communities and consumers will suffer [1-3].

Bivalves and different species of fishes have been used to study the accumulation of heavy metal on water ecosystems either in their tissues or in their bones [4-5]. Chemical reactivity of ions with cellular structural proteins, enzymes, and membrane system in mammalian systems is primarily due to metal ion toxicity with cellular structural proteins, enzymes, and membrane system. The target organs for certain metal toxicity are usually the organs that accumulate the highest concentrations of metal in the body. Usually this depends on the exposure route and the chemical composition of the metal, volatility, valance, fat solubility, and so on.

People rely heavily on fish as a source of protein. According to the Food and Agriculture Organization (FAO) of the United Nations, it is man's most important single source of high-quality protein, accounting for more than 16 percent of the animal protein consumed by the world's population. It is a particularly important protein source in areas where livestock is scarce; 17% in Africa, 26% in Asia and 22% in China [6]. To ensure that heavy metals in fishery resources are safe for human consumption, researchers and government agencies have established allowable limits above which consumption becomes hazardous.

Biomonitoring by using aquatic organisms such as fish, shell fish, and bivalves have been found to be very efficient indicator for heavy metals contamination level [7-11]. Aquatic species well known as accumulator of heavy metals and have been widely used as bio-indicator for monitoring in many countries [12-20]. Accumulated concentration of metals in marine species has been found many times higher that present in water or sediments due to take up metals at different levels in their different body organs [21-22]. This study focuses on biomonitoring of heavy metals (Zn, Cu, Ni and Pb) that accumulated in tissues and bones of Mackerel *(Rastrelliger kanagurta)* and Red Hybrid Tilapia *(Oreochromis niloticus)* collected from two different locations in Terengganu state, Malaysia. Moreover to evaluate the results obtained with the acceptable value in edible fish by Malaysian Food Regulation (MFR), World Health Organization (WHO) and Food and Agriculture Organization (FAO). First sampling was performed in February 2018 just after the monsoon season (high rainfall usually occurs from November to January). Recent sampling was carried out in February 2021 however no significant change in the readings. Data shown in this study represent the average for triplicate readings.

2. EXPERIMENTAL

2.1 SAMPLING LOCATION

Sampling was carried out from two different locations in Terengganu as shown in Figure 1. Mackerel *(Rastrelliger kanagurta)* was collected from Kijal, whereas Red Hybrid Tilapia *(Oreochromis niloticus)* was collected from Paka River. Kijal area lies within latitude and longitude (4.345477 N, 103.507904 E) respectively. This area is used by the villagers for various activities such as fishing and recreational activities. South of Kijal there is industrial area in Teluk Kalong where effluents as well as domestic sewage/wastes are deposited into the sea or rivers with or without pretreatment. Meanwhile, Paka River lies within latitudes and longitude (4.669384, 103.402295) respectively and connected to the sea through Paka estuary. Paka River is near the Petronas industrial complex and has the freshwater cage-culture for Oreochromis niloticus as well as painting activities of the fishing boats. Hence, both areas assumed to have high concentrations of different kind of pollutants especially heavy metals.

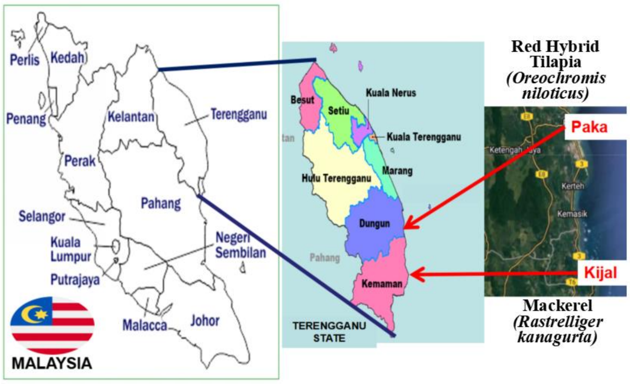

Fig. 1. Map of sampling areas

2.2. ANALYSIS

Drying process technique was used to determine the moisture content for all samples at 80 °C for 24 hours. The fish sample was separated into flesh and bone by using plastic knife. Both flesh and bone were cleaned using distilled water. After that, samples were put in beakers to measure their recorded weight. Dried samples (24 hrs, 80 ºC) were pulverized as well as soil samples after drying using stone mortar to produce a powdered form. Exactly 0.5 g for each sample either tissue, bone or soil were placed in Teflon beaker and closed tightly after adding 4 ml of concentrated Nitric Acid (HNO_3) and 1 ml of perchloric Acid (HClO). The samples were digested for 3 hours in the oven at 120 °C. After that, the digested samples were diluted into 100 ml volumetric flask and made mark with distilled water. Then the diluted samples were analyzed by using Atomic Absorption Spectrophotometer (AAS). A stock solution of Zn, Cu, Ni and Pb were prepared by using stock solution (1000 ppm) high purity grade. From this stock solution dilution was carried out to get the appropriate concentration to be used for AAS analysis. Precision performed by running triplicate analysis, therefore all data represent the average reading. Quality control of the analysis was secured by running standards from Dogfish liver and marine sediments standard reference materials. Analysis showed high confidence with the certified values (Table 1 & 2) where error for triplicate samples does not exceed ∓5%.

Table 1. Metal concentrations certified and measured sample (Dogfish liver; DOLT-4)

Element	Ni	Cu	Pb	Zn
Founded mg/kg or (ppm)	0.90	32.0	0.170	113.3
Real value; mg/kg or (ppm)	0.97	31.2	0.16	116

Table 2. Metal concentrations certified and measured sample (Marine sediment; MESS-3)

Element	Ni	Cu	Pb	Zn
Founded mg/kg or (ppm)	47.5	34.7	20.3	157.32
Real value mg/kg or (ppm)	46.9	33.9	21.1	159

3. RESULTS

The samples were dried in the oven at 80°C for 24 hours. Samples were weighed again, from the difference in weight moisture content can be calculated. Water content in the samples ranged between 70 & 77%. As depicted in Figure 2, Mackerel *(Rastrelliger kanagurta)* that was collected from Kijal contains high concentration of lead (Pb) 153.7 ppm in the bones while tissues contain less concentration 112.4 ppm. Zinc (Zn) comes next where accumulation of Zn followed the same manner. Nickel (Ni) also has a high content in the bones compared to tissues. On the contrary, the same samples show higher concentration for copper (Cu) in tissues. Regarding the samples collected from Paka for Red Hybrid Tilapia *(Oreochromis niloticus)*, the concentrations of Pb was 228.3 ppm in the bones as illustrated in Figure 3. On the contrary, the same samples show higher concentration for Cu in tissues. Nickel and Zn followed the same pattern of lead where high concentration was observed in the bones. Soil analysis for both locations was carried out to check the contamination of heavy metals in the soil. Recorded concentration was in the order Zn>Pb>Cu>Ni. In general Paka samples have higher readings compared to Kijal as shown in Figure 4.

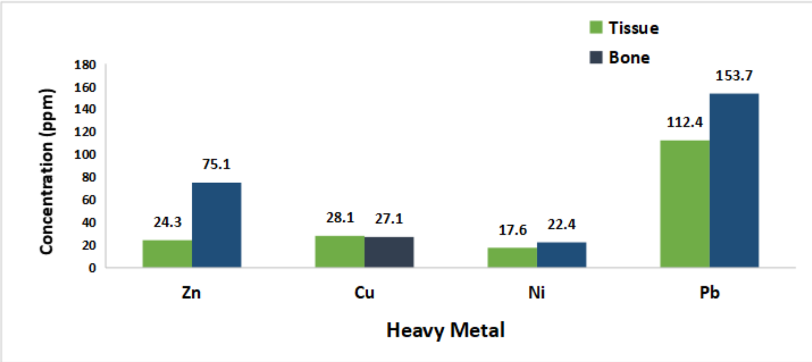

Fig. 2. Heavy metals in Mackerel *(Rastrelliger kanagurta)* collected from Kijal, Kemaman

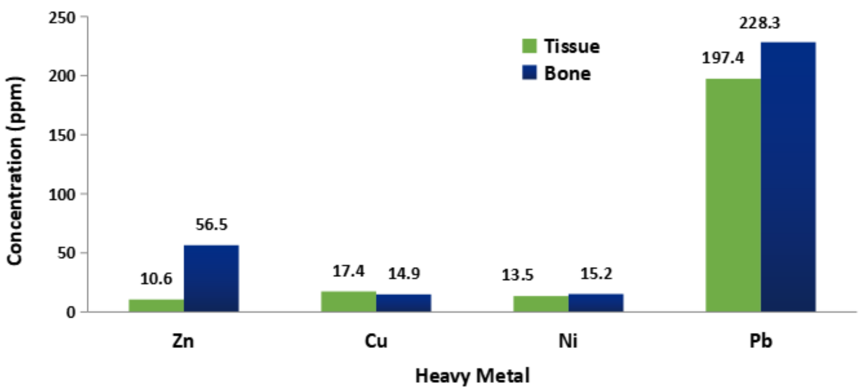

Fig. 3. Heavy metals in Red Hybrid Tilapia *(Oreochromis niloticus)*.collected from Paka River

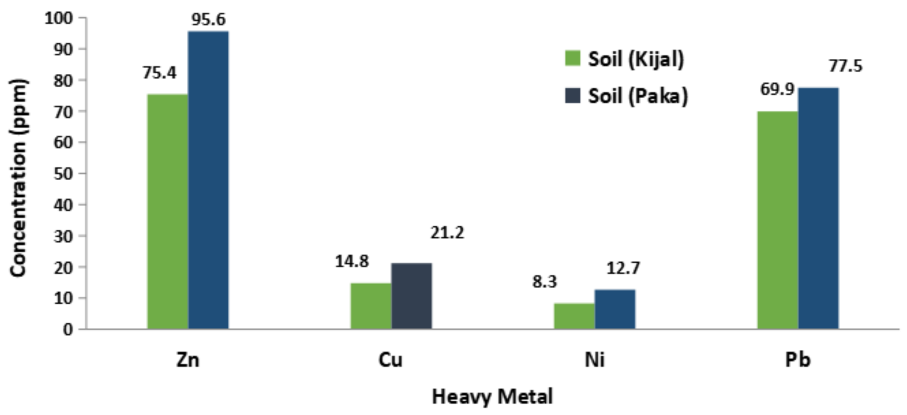

Figure 4. Heavy metals in the soil from Kijal and Paka Rivers.

4. DISCUSSION

To meet the ever-increasing demand for fish consumption, aquaculture has expanded rapidly and is now considered to be the fastest growing food industry in the world. FAO estimates that by 2030, more than half of the fish consumed by the people who live in ASEAN region will come from aquaculture. The same organization estimates that over one billion people worldwide depend on fish as the primary source of animal protein [6]. Since fishes are the inhabitants that cannot escape from the detrimental effects of heavy metal pollution due to their intimate contact with contaminated medium, regulations become very important tools to ensure good quality of edible marine species. Malaysia through the Malaysian Food Act (Malaysian Food Act 1983, has established the permissible level in which beyond this level consumption considered harmful [23]. This work goes online with this concept in which bio-accumulation of some heavy metals in the bones and tissues for two different fish species collected from two different locations (Paka River and Kijal) were evaluated. Paka River is connected to the sea through Paka estuary. The estuary is a very active area where large amounts of organic matter and trace metals flow into the marine system during the outflow from rivers and the rainy season. Heavy metals tend to be trapped in estuary sediments and remain a source of pollutants after the end of their discharge [24]. The study area is located in the moist tropics, where high rainfall is recorded during the monsoon period from November till January.

Results show that the highest Pb content in *Rastrelliger kanagurta and Oreochromis niloticus* bones were observed (153.7 and 228.3 ppm) respectively, as shown in Figures 2 and 3. The MFR (2014) limit in fish for Pb is 1 ppm, however for the World Health Organization (WHO) and the Food and Agriculture Organization (FAO) 2 ppm, indicating Pb concentrations in both fishes exceeding the tolerances set by various regulatory agencies for human consumption indicating the possibility for health risks associated with consumption. Lead has been identified by WHO as one of ten chemicals of major public health concern that require decision makers to take action to protect the health of workers, children, and women of reproductive age. WHO has made a variety of lead-related resources available on its website, including policymaker resources, technical guidance, and advocacy materials [25].

Lead poisoning can have serious consequences for children's health. Lead attacks the brain and central nervous system at high levels of exposure, causing coma, convulsions, and even death. Children who survive severe lead poisoning may develop intellectual disabilities as well as behavioural disorders. Lead is now known to cause a spectrum of injury across multiple body systems at low levels of exposure that cause no obvious symptoms. Lead, in particular, can harm children's brain development, resulting in lower intelligence quotients (IQ), behavioural changes such as decreased attention span and increased antisocial behaviour, and lower educational attainment. Lead exposure also causes anaemia, hypertension, renal impairment, immunotoxicity, and reproductive organ toxicity. Lead's neurological and behavioural effects are thought to be irreversible [26-27].

In addition, the Pb concentration of the samples collected from Paka River was higher than that of Kijal. The high Pb values recorded can result from the disposal of industrial waste that may contain large amounts of Pb, or from contaminated areas near sampling points. Another reason could be due to spilled or leaded fuel on nearby fishing vessels and dust from the burning of cars. The main business in Kijal is fishing. Moreover, Teluk Kalong region (south of Kijal), has seen significant and increasing growth recently especially in industry, and is considered the main hub of oil and gas activity in the region. Both anthropogenic and natural sources could be the major causes of heavy metal contamination.

Similar patterns were observed in Zn, with the highest concentrations in mackerel and tilapia bones, reaching 75.14 and 56.5 in both fishes respectively. The maximum value set by FAO and MFR guidelines for edible fish is 100 ppm whereas by WHO 40 ppm. All samples analyzed from the Paka River and Kijal were below the acceptable limits for Zn concentrations. The high Zn levels observed in sediment may be due to the disposal of industrial waste, which may contain higher Zn content, and the paint used to prevent corrosion of the boat. Zinc is one of the most important elements for animals and humans as copper. Zinc deficiency is characterized by delayed growth, loss of taste, and hypogonadism, which leads to reduced childbirth. Zinc toxicity is rare, but concentrations up to 40 mg/l (ppm) in water can cause toxicity characterized by symptoms such as hypersensitivity, pain, muscle stiffness, nausea, and loss of appetite [28].

In contrast to Pb and Zn, results show that Cu showed the greatest accumulation in the tissue. The distribution of copper in the tissues of mackerel and tilapia was 28.1 and 17.4 ppm, respectively. On the other hand, bone concentrations were 27.1 and 14.9 ppm for *Rastrelliger kanagurta and Oreochromis niloticus* respectively. According to MFR and WHO, the permissible limit of Cu is 30 ppm. In the current study, the accumulation level is below the value set by MFR and WHO. The reason for this may be that most Cu minerals are relatively insoluble in water, so natural water contains very little Cu. It is an essential component of human metabolism, but excessive amounts can cause bone and connective tissue damage, anemia, and liver damage. The toxicity of Cu depends on the hardness and pH of the water. The effects of high concentrations of Cu on fish are not well documented. However, there is some evidence that high levels of Cu can possess toxicity.

The most comprehensive study by Davis for sediment analysis where they reported that the average heavy metal concentration in the sediment decreased in the order of Zn> Pb> Cu> Ni between the Newport Bridge and the estuary. In the estuary system, heavy metals in the water column were observed to exhibit both conservative and non-conservative behavior along the salt gradient [29]. However, this study shows that heavy metals in sediments follow the same pattern, but with higher concentrations, as shown in Figure 4. The time lag between the two studies should be taken into account. According to Kamaruzzaman, most of the Zn in the sediments is of natural origin, probably due to the weathering of ultramafic rocks [30]. However, some higher concentrations at certain depths are expected to contribute to anthropogenic activity, especially near fishing ports and shipyards. The painting activities of fishing vessels and the use of rust preventive paints in the fishing industry can affect the concentration of Zn in sediments. Another reason can be traced back to the fact that over the past year there have been strong emissions from municipal and industrial outflows as a result of the rapid development of the region due to the expansion of industrialized areas and population growth. Steel and petrochemicals are major industries in this area and they are the catalysts to develop and support other industries in the region.

5. CONCLUSION

This study showed that Mackerel (*Rastrelliger kanagurta)* and Red Hybrid Tilapia (*and Oreochromis niloticus*) collected from Kijal & Paka respectively, both have relatively high concentration of Pb in their bones and tissues that exceeded the permissible limit set by FAO, WHO, and MFR. Significant concentration of Zn was also revealed from both samples however the concentration was below the allowable level for the edible fish set by the same organizations. Other elements analyzed from the same regions were below the permissible level. Zn in sediments found to be higher than other elements followed by lead.

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