Heavy metal assessment in water, sediment and biota from Ennore creek and Vellar estuary, southeast India.

ABSTRACT

The impact of anthropogenic activities on the accumulation of heavy metals in water, sediment and fish of both Ennore creek and Vellar estuary were observed in this study.Water and sediment sample were collected from both sampling stations and fish was collected just from onlyEnnore creek. Sediment and fish samples were subjectedto a total digestion technique (HNO_3 – HCl – HF conc.) and analysed using a spectrophotometer. Four elements Cu, Cd, Zn and Pb (tTrace elements) were determined. Heavy metal concentration of Ennore creek was significantly higher than the concentration of samples from Vellar estuary. The order of metal levels in water sample from Ennore creek is Cu > Zn > Cd > Pb and the sequence order of Vellar estuary water sample is Zn > Pb > Cu > Cd. The concentration of heavy metals in the sediment samples of Ennore creek showed in the following sequential order: Zn > Cu > Pb > Cd and the sequence order of Vellar estuary: Zn > Cd > Cu > Pb. In the Ennore creek, the accumulation of Zn in the muscle tissue is observed in the order, *O.mosambique*(7.3mg/g), *M.cephalus*(7.13mg/g) and *A.djedaba*(6.76mg/g). *M.cephalus*showed high levels of Cu (5.25mg/g) followed by *A.djedaba*(4.63mg/g) and *O.mosambique*(4.24mg/g).Based on the overall average of heavy metals present in different organs of the fish samples, trend of metal accumulation in different organs are observed as followsin the following manner; Cu: Muscle > liver > gills and Zn: Muscle > liver > gills.

Keywords: Heavy metals, Trace metals, Ennore creek, Vella estuary, Accumulation.

INTRODUCTION

Environmental pollution and its dangers are the most vital issues of societies and living creatures. However, extended populace with the improvement of era and manufacturing can purpose a lack of interest to environmental protection (Saraviet *et al.*, 2009). Industrialization leads to the pollution of ecosystem. Among all the environmental pollutions, the onepollution of water resources is a matter of incredible situation. Negative and growing international locations are at excessive chance due to lack of wastewater treatment technologies. Increasing infection of aquatic resources with massive variety of pollution is not handiest endangering the aquatic biota but also growing a global shortage of leisure waters (Sai, 2000).Heavy metals are almost in traces, which do not biodegrade in the habitats where released, and hence get biomagnified in the exposed organisms (Pandiyan *et al.,* 2021).

Among the different biotopes, estuaries play a vital role as they serve as areas of interaction between fresh and salt water. Estuaries are a unique, dynamic, complex ecosystem with a connection with the open sea through which the seawater enters accordingly to the rhythm of the tides (Kennish, 1991).The health status and the biological diversity of the Indian estuarine ecosystem are deteriorating day by day through man-made activities and dumping of enormous quantities of sewage into the estuary which in turn has drastically reduced the population of the fishes. Several studies have demonstrated metal contamination in seawater,sediments and bioaccumulation in coastal and estuarine ecosystems of India

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(Bhuvaneshwari*et al.,* 2012; Dhanakumar *et al.,* 2015; Gupta *et al.,* 2009; Jayaprakash *et al.,* 2015, 2005;Veerasingam*et al.,* 2012).

Singh *et al*. (2004) stated that rapid industrialization for sustaining economy is due to unsafe disposal of industrial effluents.India falls among the first ten industrialized countries of the world. In India about twenty-two million tons of water is discharged into aquatic bodies annually from industries.The Chennai coastal region is a distinctive example for numerous recreational and commercial activities that not only degrade the quality of coastal water but also pose a serious health hazard to marine biota and man (Beiras*et al*., 2003; Palanisamy *et al*., 2006). Ennore coast receives untreated sewage from Royapuram sewage outfall, untreated/ treated industrial effluents from Manali industrial belt, which houses many chemical industries. The dredging activities in Ennore area result in changes in the landscape, sediment transport, and dust pollution to the coast by quarrying process (Subramanian &Mohanachandran, 1990; Palanisamy*et al.,* 2006).

MATERIALS AND METHOD

Water sample

Samples were collecteddduring January to march from Ennore creek and Vellar estuary (Fig. 1 and Fig. 2). The samples were collected using clean polythene bottles. Metal concentration in water samples were analysed based on APDC-MIBK extraction procedure (Grasshoff, 1976) in which100 ml of water sample was placed in an acid cleaned separating funnel and its pH adjusted to 2.2 with concentrated HNO_3. After the addition of 2 ml of aAmmonium pyrrolidinedithiocarbonate (APDC), the chelates were extracted into 10 ml of iso-butyl methyl ketone (MIBK) under agitation. The aqueous phase was removed and the metals present in the MIBK were backtitrated with concentrated HNO_3 and distilled water. The acidic extractants were evaporated on a low temperature hot plate to remove traces of the organic solvent (Jayaprakash *et al.*,2005).

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Sediment sample

The sediment sample were collected from both Ennore creek and Vellar estuary. The samples were collected using spoon and stored in clean polythene bags. Samples were dried at 60˚C in a hot air oven for 24 hourshrs, and ground into fine powder using pestle and mortar and passed through 150 µm sieve. 0.5g of the homogenized samples were taken in Teflon tubes and digested with the combination of 5ml HNO_3, 2ml of HF and 1ml of $HClO_4$ (Perchloric acid) using microwave digestion unit. After acid evaporation to dryness, the residue was dissolved in 0.1 M HNO_3solution and diluted to 50 ml for metal analysis (Peerzada and Dickinson, 1988). The instrument was calibrated with standard chemical solutions prepared from commercially available chemicals (Merck, Germany). Analytical blanks were run in the same way as the samples and the concentrations were determined using the standard solutions prepared in the same acid 40 matrix. The quality of the data was checked by the analysis of standard reference material (BCSS-1, , obtained from National Research Council, Canada).

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Fish sample

The fishes *Mugilcephalus, Oreochromismossambicus,* and*Alepesdjedaba* were collected only from Ennore creek. All the fish collected fromEnnore creek were immediately stored in the ice chest containing ice cubes and brought to the laboratory for heavy metals analysis. The collected fishes were thoroughly cleaned before analysis to remove the sediment and biological material that are commonly present on the fish body surface. After cleaning the fishes , they were weighed and measured. The fishes were dissected in a pre-cleaned dish.

The gills, liver, and muscle tissue were separated and washed with de-ionised water. These sample were then oven dried at 70°C~~c~~ until constant weight. The dried sample were powdered using mortar and pestle and kept in vials. 1 to 2 g~~rams~~ of dried powdered samples were taken in a beaker and digested with HNO_3 and $HCIO_4$ using microwave assisted digester. Then they were evaporated to dry at room temperature. The residues were dissolved and diluted to 50ml. The trace elements were analysed using an inductively coupled plasma system (ICP) (Optical Emission Spectrophotometer by using the instrument Optima 2100DV and quantified against known standards) (Ay *et al.*, 1999).

RESULTS

Heavy metal analysis in the water sample of Ennore creek and Vellar estuary

The concentration of metals in water samples from Ennore creek and Vellar estuary were shown in Figure 3. The concentration of Cd from Ennore creek was 24.18 µg/l and concentration of sample from Vellar estuary was 0.16 µg/l. The concentration of Cu from Ennore creek was 123.56 µg/l and concentration of sample from Vellar estuary was 0.57 µg/l. The concentration of Zn from Ennore creek was 59.57 µg/l and concentration of sample from Vellar estuary was 8.25 µg/l. The concentration of Pb from Ennore creek was 6.20 µg/l and concentration of sample from Vellar estuary was 0.66 µg/l.

Heavy metal analysis in the sediment sample from Ennore creek and Vellar estuary

The concentration of metals in sediment sample from Ennore creek and Vellar estuary are~~were~~ shown in Figure 4. The concentration of Cd from Ennore creek was 1730 µg/g and concentration of sample from Vellar estuary was 0.72 µg/g. The concentration of Cu from Ennore creek was 13918.52 µg/g and concentration of sample from Vellar estuary was 0.56 µg/g. The concentration of Zn from Ennore creek was 24734.31µg/g and concentration of sample from Vellar estuary was 0.82 µg/g. The concentration of Pb from Ennore creek was 2783.31µg/g and concentration of sample from Vellar estuary was 0.07µg/g.

HEAVY METALS IN BIOTA (FISH) SAMPLE

Mugilcephalus

The concentration of selected heavy metals in the fish *M. cephalus*at Ennore estuary are shown in the Table 1. The concentration of Cu, Cd, Zn, and Pb accumulated in the muscle tissue are 5.25mg/g, BDL (Below Detection Level), 7.13mg/g, BDL respectively. The concentration of Cu, Cd, Zn, and Pb accumulated in the gills are 3.4mg/g, BDL, 27.6mg/g, BDL respectively. Similarly, the concentration of Cu, Cd, Zn, and Pb accumulated in the liver are 5.83mg/g, BDL, 41.2mg/g, BDL. (Fig. 5, 6 and~~&~~ 7)

Oreochromismosambique

The concentration of selected heavy metals in the fish *O. mosambique*at Ennore estuary are shown in the Table 1. The concentration of Cu, Cd, Zn, and Pb accumulated in the muscle tissue are 4.24mg/g, BDL, 7.3mg/g, BDL respectively. The concentration of Cu, Cd, Zn, and Pb accumulated in the gills are 4.2mg/g, BDL, 17.9mg/g, BDL respectively. Similarly, the concentration of Cu, Cd, Zn, and Pb accumulated in the liver are 7.39mg/g, BDL, 34.6mg/g, BDL, respectively. (Fig. 5, 6 and~~&~~ 7)

Alepesdjedaba

The concentration of selected heavy metals in the fish *A.djedaba*at Ennore estuary are shown in the Table 1. The concentration of Cu, Cd, Zn, and Pb accumulated in the muscle tissue are

4.63mg/g, BDL, 6.76mg/g, BDL, respectively. The concentration of Cu, Cd, Zn, and Pb accumulated in the gills are 5.3mg/g, BDL, 17.6mg/g, BDL respectively. The concentration of Cu, Cd, Zn, and Pb accumulated in the liver are 6.39mg/g, BDL, 26.6mg/g, BDL, respectively~~.~~(Fig. 5, 6 and~~&~~ 7).

DISCUSSION

The water and sediment samples were collected from both Ennore creek and Vellar estuary.The concentration of Cd from Ennore creek was 24.18 µg/l and concentration of sample from Vellar estuary was 0.16 µg/l. The concentration of Cu from Ennore creek was 123.56 µg/l and concentration of sample from Vellar estuary was 0.57 µg/l. The concentration of Zn from Ennore creek was 59.57 µg/l and concentration of sample from Vellar estuary was 8.25 µg/l. The concentration of Pb from Ennore creek was 6.20 µg/l and concentration of sample from Vellar estuary was 0.66 µg/l. The concentration of Cd from Ennore creek was 1730 µg/g and concentration of sample from Vellar estuary was 0.72 µg/g. The concentration of Cu from Ennore creek was 13918.52 µg/g and concentration of sample from Vellar estuary was 0.56 µg/g. The concentration of Zn from Ennore creek was 24734.31µg/g and concentration of sample from Vellar estuary was 0.82 µg/g. The concentration of Pb from Ennore creek was 2783.31µg/g and concentration of sample from Vellar estuary was 0.07µg/g.

The concentration was higher in Ennore creek due to the ~~many numbers~~number of thermal power plant located near the mouth of the Ennore river. This higher concentration is due to the discharge of effluents from the various industries and power plants situated in the locality. On the other hand, the concentration in water sample from Vellar estuary, when compared to Ennore creek, was distinctively low. This is due to the limited number of power plants and industries in the studied~~y~~ area. As shown in figure, the present investigation reveals that the concentration of Ennore creek and Vellar estuary are vastly different in both water and sediment. The order of metal levels in water sample from Ennore creek is shown as Cu > Zn > Cd > Pb and the sequence order of Vellar estuary water sample is Zn > Pb > Cu > Cd.The concentration of heavy metals in the sediment samples of Ennore creek showed in the following sequential order: Zn > Cu > Pb > Cd and the sequence order of Vellar estuary: Zn > Cd > Cu > Pb.Sediments are known to accumulate trace metals in aquatic ecosystems. They act as storage sinks for various pollutants. If the storage function overtakes its role, they cause problems such as pollution. This is particularly true of depositional sediments in close proximity to anthropogenic metal discharges.Inlow flow conditions most of themetals are precipitated or absorbed onto the finer sediments inthe estuarine region with organic particles and due to the sulphidic nature of sediments (Zwolsman*et al.*, 1997; Jiann *et al.*,2005; Jayaprakash *et al.*, 2015).The chemical composition of the sediments can be used as a powerful tool to determine the provenance (Vital &Stattegger, 2000). Sediments can be sensitive indicators for monitoring contaminants in aquatic environments. The sediments may be polluted with various kinds of hazardous and toxic substances (Kamaruzzaman *et al.*, 2006).

Rajathy & Azariah (1996) stated that the levels of zinc and copper in water and sediment samples showed seasonal fluctuations in the Ennore and Adyar estuaries of Chennai, India. In the present study only pre monsoon analysis was conducted which concluded both zinc and copper being high in water and sediment in both Ennore creek and Vellar estuary. In addition, the high concentration of metals in Ennore creek is due to the low salinity region and input of sedimentary fluxes. The levels of dissolved cadmium in Ennore water recorded in the present study (24µg/l) were in the same magnitude reported by Jayaprakash *et al.* (2005) in Ennore

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estuary. The concentration of copper observed in the waters of Ennore creek were way higher than Vellar estuary. Thismay be closely associated with suspended particulate matter (SPM) inputs either land-originated or mainly linked with re-suspension and remobilization processes with an active role of benthic biogenic particles.

The contamination level of zinc is higher than that of copper and cadmium in the sediment of Ennore creek. High values may be attributed to the continental input and runoff from the Araniyarriver in Ennore creek.Higher levels of lead and zinc (435 and 1090 µg/g, respectively) observed in the sediment samples of Tamilnadu coast reported by Srinivasalu *et al.* (2005).Distribution of trace metals in the sediment samples of Bay of Bengal, India (Selvaraj *et al.*, 2004) indicating enrichment of trace metals in the river basin which is due to the large-scale industrial input into the river on the upstream side as well as in the coastal region.Srinivasalu*et al.* (2007) reported copper in the northern, central sectors indicating a twofold increase in concentration (3320-6230 µg/g) than the background samples in Kalpakkam, India. On the other hand, Vellar estuary used only for fishing activities and there is low occurrence of industries in these areas.Estuarine sediment contamination is receiving increasing attention from the scientific community, since it is recognized as a major source of ecosystem health stress (Chapman & Wang, 2001; Riba*et al.*, 2002).

High accumulation of heavy metals recorded in the benthopelagic fishes compared to the demersal fishes could be due to the feed availability from both benthic and pelagic zones of the lake. In the present studies, *M.cephalus*, a benthic carnivore, *O.mosambique,* a benthic omnivore and *A.djedaba,* a pelagic carnivore have accumulated high amount of copper and zinc.It is reported that, accumulation of heavy metals in pelagic and benthopelagic planktons will be high in estuaries due to the disturbances in the benthic zone caused by natural (tidal influx and out flux, discharge from rivers etc.) and anthropogenic (dredging, transportation etc.) reasons. Heavy metal enters into the benthic organisms through their dietary preferences, food chain and food webs (Pandiyan et al., 2020).Upwelling of water from the bottom to the pelagic zone due to such disturbances, which in turn enriches the water with metals and make it available to zoo and phyto planktons in these zones. This could be one of the reasons for high metal accumulation in the above-mentioned fishes (Muraleedharan*et al*., 2006; Rejomon*et al*., 2010).Based on the overall average of heavy metals present in different organs of the fish samples, trend of metal accumulation in different organs are observed in the following manner;Cu: Muscle > liver > gills and Zn: Muscle > liver > gills.

In the Ennore creek, the accumulation of Zn in the muscle tissue is observed in the order, *O.mosambique*(7.3mg/g), *M.cephalus*(7.13mg/g) and *A.djedaba*(6.76mg/g). *M.cephalus*showed high levels of Cu (5.25mg/g) followed by *A.djedaba*(4.63mg/g) and *O.mosambique*(4.24mg/g). The accumulative capacity of heavy metals in fish muscles has been well established (Uysal *et al.*, 2008; Rahman *et al.,* 2012; Yi and Zhang, 2012; Leung *et al.*, 2014). Muscle formsmore than 60 % of the total body weight of fishes (Storelli *et al.*, 2005).The accumulation of Zn in the gills is observed in the order, *M.cephalus*(27.6mg/g), *O.mosambique*(17.9mg/g) and *A.djedaba*(17.6mg/g). The highest level of Cu was recorded in *A.djedaba*(5.3mg/g) then *O.mosambique*(4.2mg/g) and *M.cephalus*(3.4mg/g).Gills serve as a good indicator of water quality. They are sensitive to any change of water components since gill filaments and lamellae provide a very large surface area for direct and continuous contact with contaminants in water (Au. DWT, 2004). The accumulation of Zn in the liver is observed in the order, *M.cephalus*(41.2mg/g), *O.mosambique*(34.6mg/g) and

A.djedaba(26.6mg/g). The highest level of Cu was found in *O.mosambique*(7.39mg/g) then *A.djedaba* (6.39mg/g) and *M.cephalus*(5.83mg/g).The fish liver is a vital organ concerned with basic metabolism and is the major organ of accumulation, biotransformation and excretion of contaminants in fish (Figueiredo *et al.*, 2006). In spite of living in the same ecological conditions, thesix different fish species demonstrated variations in accumulation ofmetals which might be attributed to their feeding habits, trophiclevels and the contamination gradients of these sources (Jayaprakash *et al.*, 2015). Impact of contaminants on aquatic ecosystems can be evaluated by measuring biochemical parameters in the liver of fish that respond specifically to the degree and type of contamination (Barhoumi *et al.*, 2012).Metal accumulation in aquatic organisms varies depending on species, tissue, ambient concentration, sampling region and season (Çiftçi *et al.*, 2021)

CONCLUSION

The present study investigates the heavy metal concentration of Ennore creek and Vellar estuary in water, sediment and biota(fish). Among the two areas selected for studies, Ennore creek was highly polluted by Cu, Cd, Zn and Pb. This may bedue to the industrial activities and anthropogenic introduction of pollution also resulted from local people. This study has enlightened thefact the persistent pollutants like metals should be regularly monitored and any variation from the normal distributional pattern can furnish an idea about the proper management of the coastal area. It is necessary that biological monitoring of the water and fish meant for consumption should be done regularly to ensure continuous safety of the seafood. Safe disposal of domestic sewage and industrial effluents should be practiced and where possible, recycled to avoid these metals and other contaminants from going into the marine environment.

In the present study, the accumulation of heavy metals in the biota of Ennore creek has been undertaken to establish a baseline data on the heavy metal profile of Ennore creek environment. The fish sample *Mugilcephalus, Oreochromismossambicus*, and*Alepesdjedaba*were subjected to analyses. These selected fish sample have high commercial value in this particular locality and it is important to monitor the heavy metal content in their body. As through consumption the metal concentration may enter into the human system. The concentration in muscles, gills and liver was measured individually. The results of these studies indicate the concentration of heavy metals in the water and sediment from Ennore creek to be higher than the concentration of heavy metal in the water and sediment from Vellar estuary. This analytical study proves that the presence of numerous amounts of industries and power plant in and around Ennore creek has potentially reduced the quality of the water and sediment of the area.The values reported in this study can serve as baseline data to monitor future anthropogenic activities along the coast.

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List of tables and figure

Table 1. Heavy metal concentration in muscle, gill and liver of selected fishes.

FISH SAMPLE	MUSCLE				GILL				LIVER			
	Cu	Cd	Zn	Pb	Cu	Cd	Zn	Pb	Cu	Cd	Zn	Pb
Mugilcephalus	5.25	BDL	7.13	BDL	3.39	BDL	27.59	BDL	5.83	BDL	41.2	BDL
Oereochromismosambique	4.24	BDL	7.3	BDL	4.15	BDL	17.91	BDL	7.39	BDL	34.63	BDL
Alepesdjedaba	4.63	BDL	6.76	BDL	5.33	BDL	17.64	BDL	6.39	BDL	26.59	BDL

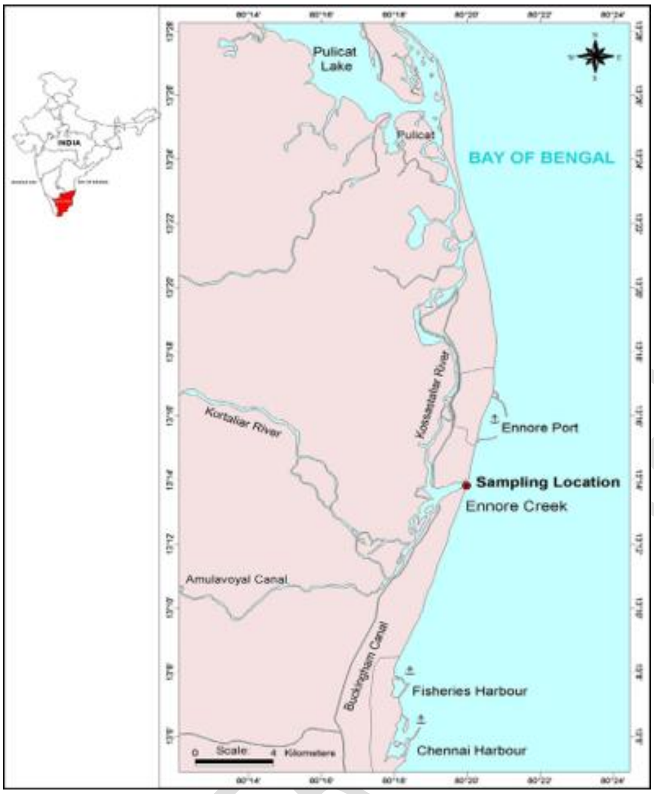

Figure 1. Sampling station – Ennore creek

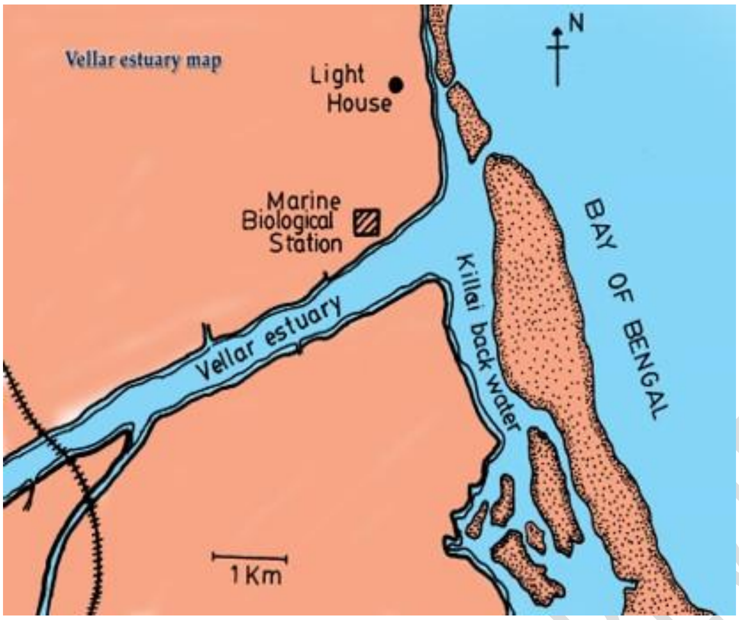

Figure 2. Sampling station – Vellar estuary

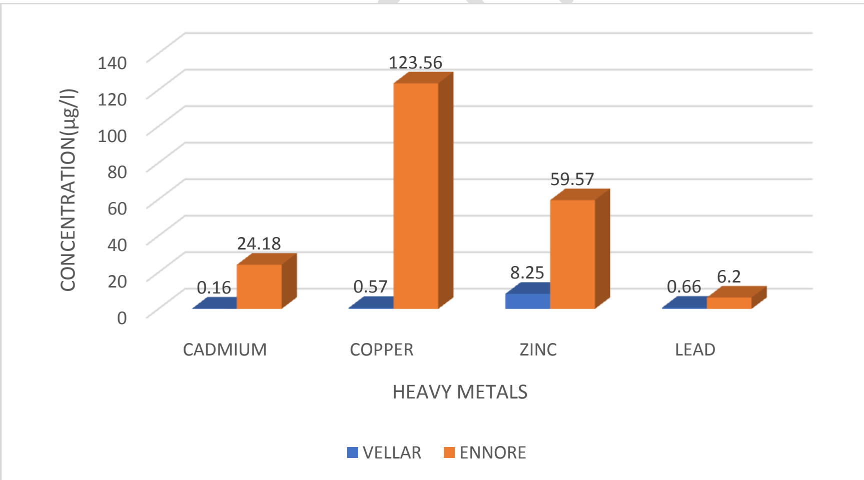

Figure 3. Heavy metal concentration in water sample from Ennore creek and Vellar estuary

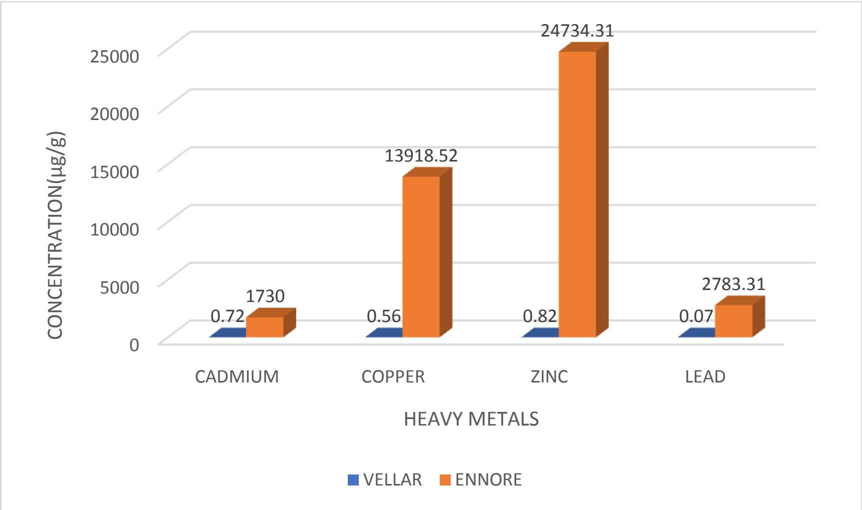

Figure 4. Heavy metal concentration in sediment sample from Ennore creek and Vellar estuary

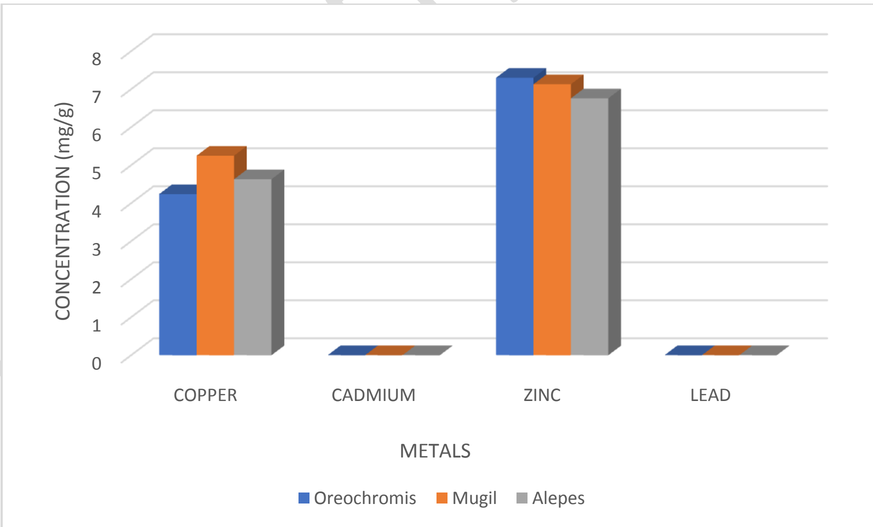

Figure 5. Heavy metal concentration in muscle tissue of selected fishes

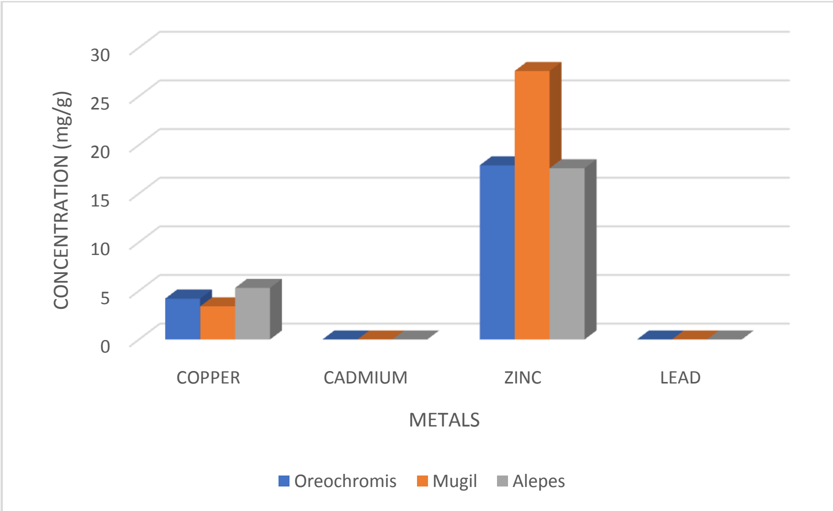

Figure 6. Heavy metal concentration in gills of selected fishes

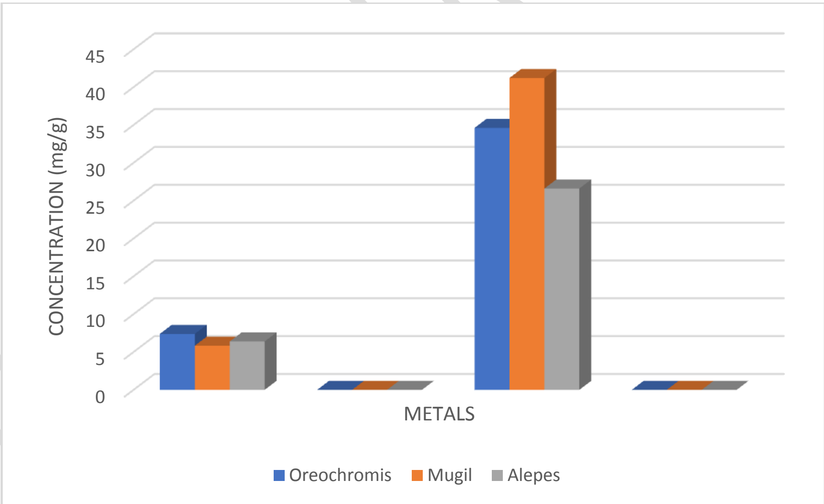

Figure 7. Heavy metal concentration in liver of selected fishes